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Deterministic and random models of limited aggregation and gelation phenomenon

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École Doctorale de Sciences Mathématiques de Paris Centre

THÈSE

présentée pour obtenir le grade de

Docteur de l'Université Paris VI - Pierre et Marie Curie

Discipline : **Mathématiques**

présentée par

Raoul NORMAND

**Modèles déterministes et aléatoires d'agrégation
limitée et phénomène de gélification**

dirigée par **Jean BERTOIN** et **Lorenzo ZAMBOTTI**

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Modèles déterministes et aléatoires d'agrégation limitée et phénomène de gélification

Résumé

Dans cette thèse, nous étudions des modèles d'agrégation limitée, qui modélisent la coalescence de particules ayant des “bras”, c'est-à-dire un nombre fixé de liens potentiels. Une particule ne peut donc créer plus de liens que son nombre de bras. On s'intéresse en particulier à une variante de l'équation de Smoluchowski introduite par Jean Bertoin.

Ce document comprend, outre l'introduction, trois chapitres. Le premier est dévolu à l'étude d'un modèle sexué de coagulation, où les particules ont des bras mâles et femelles et seuls des bras de sexes opposés peuvent se joindre. Ce modèle généralise et unifie ceux de Bertoin, dont on peut en particulier retrouver les résultats.

Le second chapitre comprend un travail en collaboration avec Lorenzo Zambotti. On s'y intéresse à l'unicité des solutions d'équations de coagulation après gélification, en particulier l'équation de Smoluchowski avec noyau multiplicatif et l'équation d'agrégation limitée. En particulier, on donne des preuves rigoureuses de certaines heuristiques de la littérature physique, par exemple en calculant précisément le temps de gélification. Dans le cas d'agrégation limitée, on obtient aussi des formules particulièrement simples pour les concentrations limites.

Pour expliquer celles-ci, on étudie dans le dernier chapitre un modèle microscopique pour l'équation de Smoluchowski d'agrégation limitée. Ceci est un travail commun avec Mathieu Merle. On parvient à décrire précisément l'état microscopique du système à tout temps et ainsi à retrouver les formules du second chapitre. Une caractéristique frappante de ce modèle est qu'il possède une propriété de criticalité auto-organisée.

Mots-clefs

Équation de Smoluchowski, Modèles d'agrégation limitée, Gélification, Systèmes infinis d'équations différentielles, EDP non linéaires, Graphes aléatoires, Modèle de configuration, Criticalité auto-organisée

Deterministic and random models of limited aggregation and gelation phenomenon

Abstract

In this PhD thesis, we study limited aggregation models, modeling coalescence of particles with “arms”, i.e. a fixed number of potential links. Hence, a particle cannot create more links than its number of arms. We are particularly interested in a variant of Smoluchowski’s equation introduced by Jean Bertoin.

This document contains, after the introduction, three chapters. The first is devoted to the study of a sexed model of coagulation, where particles have male and female arms, and only arms with opposite genders can bind. This model generalizes and unifies Bertoin’s model and we can in particular recover his results.

The second chapter contains a joint work with Lorenzo Zambotti. We deal with the uniqueness to various coagulation equations after gelation, in particular Smoluchowski’s equation with a multiplicative kernel and the equation with limited aggregations. We provide in particular rigorous proofs to several heuristics of the Physics literature, e.g. by computing precisely the gelation time. In the case of limited aggregations, we obtain particularly simple formulas for the limiting concentrations.

To explain them, we study in the last chapter a microscopic model for Smoluchowski’s equation of limited aggregation, what is a joint work with Mathieu Merle. We manage to describe precisely the microscopic state of our system at any time, and thus recover the formulas from the previous chapter. A striking feature of this model is that it exhibits a phenomenon of self-organized criticality.

Keywords

Smoluchowski’s equation, Limited aggregation models, Gelation, Infinite systems of ODEs, Non-linear PDEs, Random graphs, Configuration model, Self-organized criticality

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Chapter 1

Introduction

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1 Content of the thesis

This PhD thesis focuses on the pre- and post-gelation behavior of Smoluchowski's equation for limited aggregation introduced by Bertoin [3], meant to model pairwise coalescence of particles with a certain number of potential links, called *arms* in the text. We shall focus on the continuous deterministic model as well as on a microscopic model. The latter is related to random graph theory, more precisely to the configuration model.

The main material of this text consists in three articles, each accounting for a chapter of the thesis.

- The first [43] studies an alternate (continuous) model for limited aggregation, where the particles are given two types of arms, called female and male, and where only male and female arms can bind. This is quite natural when we think of ionic bonds. The results we prove are similar to those obtained for Smoluchowski's equation, with or without limited aggregations, and in fact allow to recover the results proved by Bertoin [3].
- The second is a joint work [44] with Lorenzo Zambotti. We prove therein the uniqueness of the solutions to Smoluchowski's equation (with or without limited aggregations) and provide representation formulas which allow to compute general relevant quantities. The strikingly simple expressions obtained for the final state of that model calls for a probabilistic interpretation, given in the last part of this work.
- The last chapter contains a joint work [41] with Mathieu Merle to be submitted soon. It deals with a microscopic model for Smoluchowski's equation with limited aggregations, aiming at giving a probabilistic interpretation for some formulas obtained in the previous chapter. This is achieved by describing precisely at any time the shape of the “small” clusters in that model and observing a striking feature called *post-gelation self-organized criticality*.

The goal of this introduction is threefold. We first provide a presentation of the one-century-old Smoluchowski equation, which has been extensively studied but has also raised many interesting and unanswered questions. Though we will mainly consider other models (of limited aggregation), the intuition and ideas gained from earlier (mathematical or not) works on Smoluchowski's equation is at the very heart of many of our results and proofs. Moreover many properties of Smoluchowski's equation still hold for other models, up to some quite obvious modifications.

One of these properties, and a most difficult one to handle, is the *gelation* phenomenon, which heuristically corresponds to the appearance of giant clusters of particles. Oddly, it turns out that this phenomenon is quite well understood heuristically (in what Aldous [1] calls “scientific modeling literature”), but that few rigorous results have been obtained. The third and fourth part of this thesis are actually devoted to studying rigorously this phenomenon.

The second goal of this introduction is to present our main interest, namely Smoluchowski's equation for limited aggregations, as introduced in 2009 by Bertoin [3]. It is closely related, on the one hand to Smoluchowski's equation, on the other hand to a random graph model called the configuration model which we shall explain. The paper [3] is actually the very starting point of this thesis, which gave rise to most of the questions we tried to answer, and we shall thus state and explain the results it contains.

The last part of this introduction is devoted to the precise statements and descriptions of the results of the three chapters. As already said, many results or models are easy to understand when we know the results or heuristics concerning Smoluchowski's equation, and could with a little luck be guessed down to the last comma¹. We hope however that some of them are more unexpected.

¹But hopefully, not the proofs!

2 A short history of Smoluchowski's equation

2.1 Presentation

2.1.1 A first motivation

Coagulation phenomena arise in many areas of science, such as chemistry (formation of polymers from monomers), meteorology (creation of water droplets), biology (coagulation of algae), astronomy (formation of planets) or, in mathematics, genealogy or random graph theory. Aldous' review [1] is a good introduction to the subject. Depending on the phenomenon which is to be modeled, many mathematical formulations, random or deterministic, can be given. Among the deterministic tools, one of the most usual is Smoluchowski's equation introduced in [57], which is meant to model *pairwise* coalescence of particles (or clusters of particles) in a mean-field setting.

2.1.2 A matter of vocabulary

Let us be careful about the terminology we will use. Except in a part of the third chapter of this thesis, the masses will always be discrete, integer-valued to be specific. We may thus assume that the elementary entities of our system are *particles* of mass 1. When particles bind together, they create a *cluster*, which has a mass $m \in \mathbb{N}^*$ being the number of particles it contains.

The coagulations are seen as the creation of a link between particles. This also creates a link between the clusters to which these particles belong. Hence, depending on the point of view we wish to adopt, we will speak of coagulation of particles or of clusters, though this is obviously the same.

2.1.3 The equation

To understand Smoluchowski's equation, imagine that clusters of different masses move through space. Two of them, of mass, say, m and m' , may, if sufficiently close, merge into a cluster of mass $m + m'$. Obviously, taking into account all parameters (mass, position, velocity, shape, ...) is rather complicated. In a mean-field model, the whole dynamic is thus subsumed into a symmetric kernel $\kappa(m, m')$, modeling the "rate" at which two clusters of mass m and m' coalesce. This is to be understood in the following sense: call $c_t(m)$ the concentration of clusters of mass m at time t , i.e. the number of such clusters per unit volume. Then, the average number of coagulations

$$\{m, m'\} \rightarrow m + m'$$

in a time interval $[t, t + dt]$ is given by

$$\frac{1}{2} \kappa(m, m') c_t(m) c_t(m') dt.$$

The evolution of $c_t(m)$ as time passes thus obeys Smoluchowski's equation, actually an infinite system of nonlinear ODEs,

$$\frac{d}{dt} c_t(m) = \frac{1}{2} \sum_{m'=1}^{m-1} \kappa(m, m') c_t(m') c_t(m - m') - \sum_{m' \geq 1} \kappa(m, m') c_t(m) c_t(m'), \quad (1.1)$$

for $m \in \mathbb{N}^*$. The first term of the RHS accounts for creation of clusters of mass m , by coagulation of clusters of mass m' and $m - m'$. The second accounts for the disappearance of such clusters by coagulation with another cluster.

2.2 What is obvious?

From a physical point of view, at least two facts should be intuitively obvious. Here and below, for nonnegative functions c and f on \mathbb{N} , we write

$$\langle c, f \rangle = \langle c, f(m) \rangle = \sum_{m \geq 1} c(m) f(m).$$

- First, the c_t 's, supposed to model concentrations, should be nonnegative. Obviously, we will only consider nonnegative initial conditions, and it is then easy to see that any solution to (1.1) or any of its variants remains nonnegative. We give a proof of this fact in the second chapter, but we will anyway take it for granted in the whole text.

- Secondly, the mass

$$\langle c_t, m \rangle = \sum_{m \geq 1} m c_t(m)$$

should remain constant, since none is lost when a coagulation occurs. This is in fact quite a tricky question and a major breakpoint in the theory, as we shall now see.

2.3 Multiplicative kernel and gelation

The behavior of Equation (1.1) depends heavily on the kernel κ , which itself depends on the process we wish to model (see [1] for some examples). However, three kernels are of particular interests, namely the constant $\kappa(m, m') = 1$, additive $\kappa(m, m') = m + m'$ and multiplicative $\kappa(m, m') = mm'$ kernels. For the first two kernels, the system is quite easy to solve, for instance by considering a PDE involving the generating function of $(c_t(m))$. The constant case has actually been solved by Smoluchowski himself [57], the additive case by Golovin in [20].

The multiplicative kernel has a much more interesting behavior, first pointed out by McLeod [39].

Theorem 2.1 (McLeod, 1962). *For initial conditions $c_0(m) = \mathbb{1}_{\{m=1\}}$, Smoluchowski's equation (1.1) with a multiplicative kernel has a unique solution for $t \in [0, 1]$. This solution enjoys the following properties.*

- For all $t < 1$,

$$\sup_{s \in [0, t]} \langle c_s, m^2 \rangle < +\infty,$$

and

$$\lim_{t \rightarrow 1^-} \langle c_t, m^2 \rangle = +\infty.$$

- This solution is mass-conservative, that is, for all $t \in [0, 1]$,

$$\langle c_t, m \rangle = \langle c_0, m \rangle = 1.$$

Moreover, there is no mass-conservative solution defined on a greater interval $[0, T]$ for $T > 1$.

Hence, if there is actually a solution on a larger time-interval, then the mass changes after 1, and it is easy to see that it can only decrease then. This is a phenomenon called *gelation*. Though it seems physically rather counterintuitive, it may be interpreted as the

appearance of a giant cluster with infinite mass but zero concentration, accounting for a positive fraction of the total mass of the system².

We may think of this phenomenon from a chemical point of view, where our clusters experience a chemical reaction in a solution, which eventually leads to the formation of huge clusters which precipitate. They then fall to the bottom of the solution where they become inert. Hence, the mass in solution decreases. We will call these large clusters by the generic name of *gel*. Throughout this text, we shall adopt this heuristic point of view, using related vocabulary and metaphors.

Now, obviously, these questions only make sense if we know that a solution does exist on a larger interval than $[0, 1]$. It can actually be proved that this holds, by using either deterministic [34] or random [45] techniques. In the first case, this is done by considering a truncated system taking into account only a finite number of masses. In the second case, by exhibiting a discrete random model, which is shown to be tight, and such that any converging subsequence solves Smoluchowski's equation (1.1). Now is thus probably the right time to introduce these models.

2.4 Microscopic model and Flory's equation

2.4.1 Marcus-Lushnikov model

Let us now deal with a microscopic model underlying Smoluchowski's equation (1.1). To keep the model as simple as possible, we shall only deal with monodisperse initial conditions $c_0(m) = \mathbb{1}_{\{m=1\}}$. The most natural model is to consider a pure-jump Markov process $t \mapsto \mathbf{X}_t^{(N)} = (\mathbf{X}_t^{(N)}(m))_{m \in \mathbb{N}^*}$, with values in $N^{\mathbb{N}^*}$. For $m \in \mathbb{N}^*$, the variable $\mathbf{X}_t^{(N)}(m)$ should be thought of as the number of clusters with mass m at time t . In particular, we start from initial condition $\mathbf{X}_0^{(N)}(1) = N$ and $\mathbf{X}_0^{(N)}(m) = 0$ for $m \geq 2$. Define

$$\Delta_{m,m'}(i) = \mathbb{1}_{\{i=m+m'\}} - \mathbb{1}_{\{i=m\}} - \mathbb{1}_{\{i=m'\}}, \quad i \in \mathbb{N}^*.$$

Then, the transition

$$\mathbf{X}^{(N)} \rightarrow \mathbf{X}^{(N)} + \Delta_{m,m'}$$

occurs at rate $\kappa(m, m')$. In other words, two clusters of mass m and m' coalesce at rate $\kappa(m, m')$. This is called a Marcus-Lushnikov process, as it was introduced and studied by Marcus [38] and Lushnikov [37].

One would thus expect that, after proper space- and time-rescaling, this would converge to a solution of Smoluchowski's equation. To be precise, let $n_t^{(N)} = X_{t/N}^{(N)}/N$. We wish to study its possible limit c in the Skorokhod space $\mathbb{D}(\mathbb{R}^+, \ell^1(\mathbb{N}^*))$ of càdlàg process with values in $\ell^1(\mathbb{N}^*)$, expecting c to be a solution to (1.1). This is actually only partly true.

In the case of a sublinear kernel (i.e. such that $\kappa(m, m')/m \rightarrow 0$ as $m \rightarrow +\infty$ for all m'), it has been first proved by Jeon [27] that this indeed holds. However, sublinear kernels do not exhibit gelation. Norris [45] considered this model for kernels which may exhibit gelation, such as the multiplicative one. He shows that the convergence to Smoluchowski's equation holds *only before the gelation time*.

2.4.2 What is the gelation time?

Let us take advantage of this last result to insist on a slight issue regarding the definition of the gelation time. For the continuous model (1.1), the most natural definition is the time

²Actually, there may just as well be several giant clusters, though we talk of “the” giant cluster by abuse of language.

when the mass starts to decrease. For instance, it is 1 in the context of McLeod’s result. It turns out that this is also the moment when the second moment $\langle c_t, m^2 \rangle$ explodes, though this may be purely coincidental.

There is actually an easy implication: as long as the second moment remains finite, the mass remains constant (this is just dominated convergence). The converse is not so obvious, but it is commonly admitted to be the case. We shall prove in the third chapter of this text that this is actually true for the multiplicative kernel.

In most cases where this relation is still unknown, as in Norris’ result mentioned above, the gelation time is defined as the first time when the second moment explodes, mainly because this is a stronger definition.

We may also want to define gelation times for microscopic models. Several definitions exist in that case, which may yield different times. We refer to Jeon [27] for more details. Concerning the present text, we will define in the last chapter a gelation time for our microscopic model, and prove that it converges to the gelation time for the corresponding continuous model.

2.4.3 Flory’s equation

In addition to Norris’ result mentioned above, Fournier and Giet [17] have shown the following: when $k(m, m')/m'$ converges to some $l(m)$ as $m' \rightarrow +\infty$ (as is the case of the multiplicative kernel), then, on the whole of \mathbb{R}^+ , a limit point c of $(n^{(N)})$ solves *Flory’s equation*:

$$\begin{aligned} \frac{d}{dt}c_t(m) &= \frac{1}{2} \sum_{m'=1}^{m-1} \kappa(m, m') c_t(m') c_t(m - m') - \sum_{m' \geq 1} \kappa(m, m') c_t(m) c_t(m') \\ &\quad - l(m) c_t(m) \sum_{m' \geq 1} m' (c_0(m') - c_t(m')). \end{aligned} \tag{1.2}$$

This is Smoluchowski’s equation with an extra term added, modeling an interaction between the gel and the clusters in solution. Note indeed that $l(m)$ may be interpreted as the rate of coagulation of clusters of mass m with clusters of infinite mass, and that

$$\sum_{m' \geq 1} m' (c_0(m') - c_t(m'))$$

is precisely the missing mass of the system, or, in other words, the mass in the gel.

The convergence of the discrete model to Flory’s rather than to Smoluchowski’s equation actually makes perfect sense. Indeed, in the discrete model, any cluster can interact with any other, whatever their sizes are, whereas Smoluchowski’s equation does not allow any interaction with the gel. Hence, a decent microscopic model for the latter would prevent the “big” clusters to interact with the others. This idea has been formalized by Fournier and Laurençot in [19]. In that paper, they allow only the clusters of size less than $\alpha(N)$ to coagulate, where $1 \ll \alpha(N) \ll N$, and show that the process converges (up to a subsequence) to a solution of Smoluchowski’s equation. That is the idea we will take over to build our microscopic model in the last chapter.

2.4.4 Other microscopic models

The model we consider above is quite simple and probably does not really convey the physical idea of coagulation. We may rather want to identify the clusters and actually “see” them grow bigger as time passes.

- On the one hand, we can forget about the space and study the combinatorial structure of our clusters. To be more precise, say that they are formed of elementary particles. A coagulation is seen as the creation of an edge between particles, and the clusters are the connected components of the graph which is formed this way. We can then study the typical structure of such a graph and how it evolves. Probably the best known model of that type is Erdős-Rényi's [10, 11], where two vertices are bound after an exponential time. The evolution of the size of the connected components is closely related to Smoluchowski's equation (1.1) with a multiplicative kernel [2]. Another classical model, called the configuration model, is related to Smoluchowski's equation for limited aggregation. We will see the relation between the two in the next section.
- On the other hand, we may only keep track of the mass (or size) of the clusters, assume that they move through space and may coalesce when sufficiently close. When the motion of the clusters is a Brownian motion, it has been shown [32, 22, 23] under various assumptions that, after proper rescaling, the cluster-size distribution converges to a solution of Smoluchowski's equation (1.1), with a kernel depending on the model. This is probably the most striking evidence that this equation is a good continuous model of coalescence.

3 Smoluchowski's equation of limited aggregations

3.1 Bertoin's model

3.1.1 The equation

Except in the third chapter, this thesis will mainly focus on a variant of Smoluchowski's equation introduced by Jean Bertoin [3], which we will call the *modified* Smoluchowski equation. In contrast, Equation (1.1) will be called the *standard* Smoluchowski equation.

Two main ideas underlie the introduction of this equation. One is its link with the configuration model, which we shall explain later. Another is the following important issue concerning the standard Smoluchowski equation: an elementary particle can (and eventually will if the kernel is positive) be bound to infinitely many other particles. However, physically, one may want to think of coagulations as covalent bonds, whose number is limited. A way around this problem is to initially give each particle a certain number of arms. These arms are used to create the bonds, in that two arms are used to create a link and cannot be used again. Then, a cluster is characterized by its mass m and its number of (free) arms a , i.e. the total number of non-used arms of the particles it consists in. This model has first been considered by Bertoin in [3], and the evolution of concentrations $c_t(a, m)$ of (a, m) -clusters, i.e. clusters with a arms and mass m , is given by

$$\begin{aligned} \frac{d}{dt}c_t(a, m) = & \frac{1}{2} \sum_{a'=1}^{a+1} \sum_{m'=1}^{m-1} a'(a+2-a')c_t(a', m')c_t(a+2-a', m-m') \\ & - \sum_{a' \geq 1} \sum_{m' \geq 1} aa'c_t(a, m)c_t(a', m'). \end{aligned} \quad (1.3)$$

As for (1.1), the first term of the RHS accounts for the creation of (a, m) -clusters by coagulation of (a', m') and $(a+2-a', m-m')$ -clusters. The second term accounts for the disappearance of such clusters, by coagulation with other clusters.

Note also that here as well, the kernel is multiplicative. The reason for that choice is the relation with the configuration model explained in Section 3.3. We may anyhow

ask natural questions regarding this equation, in particular wonder if there is a unique solution, and if we may observe the same phenomenon of gelation as for Smoluchowski's equation with a multiplicative kernel. Intuitively, this depends on the number of arms on each particle. For instance, nothing happens if particles have no arm. If they all have one arm, clusters of two particles only are created as time goes by. On the other hand, if all particles have a huge number of arms, big clusters may very well appear.

3.1.2 Normalization

It is important to insist here on the normalization considered for the initial conditions. Here and below, we denote, for $c, f : \mathbb{N} \times \mathbb{N}^* \rightarrow \mathbb{R}^+$,

$$\langle c, f \rangle = \sum_{(a,m) \in \mathbb{N} \times \mathbb{N}^*} c(a,m) f(a,m)$$

writing a for the function $(a,m) \mapsto a$, m for $(a,m) \mapsto m$ and so on.

In the text, we will consider two types of initial conditions:

- Either monodisperse initial conditions $c_0(a,m) = \mu(a) \mathbb{1}_{\{m=1\}}$. They have two main advantages: the first is that they allow to obtain quite simple expressions for $c_t(a,m)$. The second is that, when we consider a corresponding microscopic model, the initial state is just constituted of a set of elementary particles, with no links created between them.
- Or polydisperse initial conditions, where there can be particles of any mass at time 0. Obviously, the interest is to obtain more general results. It is however harder to study a corresponding microscopic model, and we shall not use this approach.

We may also adopt two conventions for the normalization, namely either have initial conditions c_0 with unit mean, i.e. $\langle c_0, a \rangle = 1$, or c_0 with unit concentration, i.e. $\langle c_0, 1 \rangle = 1$. It really does not matter if $\langle c_0, a \rangle < +\infty$, since this only leads to a linear time-change in the models.

- When considering the continuous model given by Smoluchowski's equation (1.3) (as in the second and third chapter), we will mainly choose the first normalization, since it makes computations easier. For monodisperse initial conditions $c_0(a,m) = \mu(a) \mathbb{1}_{\{m=1\}}$, this means, from a microscopic point of view, that we consider
 - a large number N of particles,
 - with i.i.d. number of arms given by $\mu / \langle \mu, 1 \rangle$,
 - in a volume $\langle \mu, 1 \rangle$.
- When considering a microscopic model (as in the fourth chapter), we will only consider monodisperse initial conditions $c_0(a,m) = \mu(a) \mathbb{1}_{\{m=1\}}$ with the second normalization, i.e. such that μ is a probability. It means that we consider
 - a large number N of particles,
 - with i.i.d. number of arms given by μ ,
 - in a volume 1.

However, for the most part of the third chapter, no particular normalization is chosen, so as to give the most general results.

3.1.3 Result

In his paper [3], Bertoin considers initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$ for a measure μ with unit mean and finite second moment. He then answers the standard questions on Smoluchowski's equation. Denote in the sequel $K = \langle \mu, a(a-1) \rangle$ and

$$T_{\text{gel}} = \begin{cases} +\infty & \text{if } K \leq 1 \\ \frac{1}{K-1} & \text{if } K > 1. \end{cases} \quad (1.4)$$

Theorem 3.1. *For initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$, Equation (1.3) has a unique solution defined on $[0, T_{\text{gel}})$. This solution has the property that, for all $T < T_{\text{gel}}$,*

$$\sup_{t \in [0, T]} \langle c_t, a^2 \rangle < +\infty$$

and, if $T_{\text{gel}} < +\infty$,

$$\lim_{t \rightarrow T_{\text{gel}}^-} \langle c_t, a^2 \rangle = +\infty.$$

Moreover, for $t < T_{\text{gel}}$, these solutions are given by explicit formulas.

By analogy with the result of McLeod (Theorem 2.1), the time T_{gel} is interpreted as the *gelation time*. When $T_{\text{gel}} = +\infty$, the explicit formulas allow to compute the limiting concentrations. To this end, we define a probability measure ν by $\nu(k) = (k+1)\mu(k+1)$, $k \in \mathbb{N}$, and assume that $\nu \neq \delta_1$. Its m -th convolution product is written ν^{*m} .

Corollary 3.2. *When $T_{\text{gel}} = +\infty$, the following convergence holds:*

$$\lim_{t \rightarrow +\infty} c_t(a, m) = c_\infty(m)\mathbb{1}_{\{a=0\}}$$

where, for $m \geq 2$,

$$c_\infty(m) = \frac{1}{m(m-1)}\nu^{*m}(m-2). \quad (1.5)$$

Those limiting concentrations can be interpreted in terms of the total progeny of some Galton-Watson process. A rigorous explanation of this relation is given in the paper [4], and we shall give an intuitive presentation of the result. This is achieved thanks to the *configuration model*, which deserves an introduction of its own.

3.2 Configuration model

3.2.1 Definition

The configuration model is a random graph model aiming at producing a graph with prescribed degrees. The simplest definition is to consider a set of N particles with a_1, \dots, a_N arms (or half-edges, or stubs), and then to take a uniform pairing of these arms, i.e. a partition of the set of arms in pairs. If the total number of arms is odd, we leave an arm unpaired. We then link the two arms in each pair to produce an edge, so, as a result, we get a random graph. Note that the number of pairings is finite so that this variable is well-defined. Note also that we may actually obtain a multiple graph. In the standard definition of the configuration model (see e.g. [54]), one conditions on the graph being simple, but this is not relevant in our case.

There are multiple ways of constructing a configuration model, other than naively (and algorithmically extremely costly) picking a pairing uniformly at random.

- A first way is to choose a random uniform ordering of the arms, and to bind together the first and the second arm, the third and fourth, and so on. To do so, one may for instance set i.i.d. exponential clocks on the arms, which induce a natural ordering of them.
- Another way is to first remove an arm uniformly at random if the number of arms is odd. Then, pick a particle arbitrarily. Choose one of its arms, and bind it with another one uniformly chosen among all other arms. Then, take any available (i.e. not yet paired) arm of this newly-formed cluster, and bind it to another arm uniformly chosen among all the other available ones. Go on until no more arms are available in the cluster, then start over with any other particle. It is important to note that, at each step, the choice of the first vertex or arm really is arbitrary, in that we may choose them according to any random or deterministic rule.

To get a microscopic model for (1.3), an intuitive idea is to consider a dynamical version of this algorithm. Obviously, the same issues as for microscopic models for Smoluchowski's equation arise. When no gelation occurs, this should however provide a good model, as we shall see.

3.2.2 Giant component

A classical question about random graphs is the existence of a giant component, i.e. a connected component with size C_N such that $C_N = \Theta(N)$. This problem is entirely solved in the case of the configuration model. The main assumption is that the empirical measure

$$\frac{1}{N} \sum_{i=1}^N \delta_{a_i}$$

converges to a probability μ with a second moment, and that so do its two first moments.

An important notion which we will use throughout the text is the size-biased shifted-by-one probability $\hat{\pi}$ deduced from a probability π . It is defined, whenever $\pi \neq \delta_0$ (in which case we let $\hat{\pi} = \delta_0$) and π has a finite mean, by

$$\hat{\pi}(k) = \frac{(k+1)\pi(k+1)}{\sum_{i \geq 1} i\pi(i)}, \quad k \geq 0.$$

Its interests lies in the following remark. Consider N particles with empirical distribution of arms π (i.e., there are $\pi(k)N$ particles with k arms). When one picks a particle uniformly at random, its number of arms has law π . Now, pick an *arm* uniformly at random, and consider the *outer* number of arms of the particle to which it is attached, i.e. its number of arms but the one picked. Then this number has law $\hat{\pi}$. Notice that the size-bias stems from the fact that we pick an arm, not a particle, uniformly at random; the shift from the fact that we consider the outer number of arms.

The main result about the configuration model [42, 26, 54] is that a giant component exists with probability tending to 1 as $N \rightarrow +\infty$ if and only if $\langle \hat{\mu}, k \rangle > 1$, and else no giant component exists with probability tending to 0.

This condition is in fact closely related to the occurrence of gelation in Smoluchowski's equation. To be precise, consider initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$, thus considering a different normalization from that of Theorem 3.1. This only induces a linear time-change, and obviously does not matter to whether $T_{\text{gel}} < +\infty$ or $T_{\text{gel}} = +\infty$. Then, $T_{\text{gel}} < +\infty \Leftrightarrow \langle \hat{\mu}, k \rangle > 1$, i.e. there is gelation if and only if a giant component appears

in the corresponding configuration model. This is natural if we think of the configuration model as a microscopic model for Smoluchowski's equation (1.3), and gelation as the appearance of a giant cluster.

3.3 Microscopic model

3.3.1 Definition

A nice microscopic model for Smoluchowski's equation with limited aggregation (1.3) should be obtained as a dynamical version of the configuration model. To this end, proceed as follows. Consider first a large number N of particles with i.i.d. number of arms with law $\mu_i := \mu / \langle \mu, 1 \rangle$. Then, set i.i.d. exponential clocks on each *pair* of arms, and bind the two arms whose clock rings first. Then, wait for the next clock to ring on a couple of arms $\{a, b\}$. If a or b has already been paired, do nothing, and else, pair them. Go on until all arms (except possibly one) are paired.

This construction provides a process such that two clusters with a and a' available arms coalesce at rate close to aa' . This is not the exact rate since there is a positive probability that the two arms we bind belong to the same cluster, in which case no coagulation occurs. However, this event has very small probability when N is large, and so this does not play any role in the limit. This is the reason for the rate aa' in Equation (1.3), and why it is interesting to study this one amongst all.

Now, the same result as for Smoluchowski's equation, as explained in Section 2.4, holds. To be precise, after space- and time-rescaling, the concentrations converge to the solution to Smoluchowski's equation (1.3) if there is no gelation. Else they converge to a solution to Flory's equation for limited aggregations, which will be introduced in Chapter 3.

3.3.2 Limiting concentrations

Now, the final state of this construction should explain the limiting concentrations (1.5). But this final state is just a uniform pairing of the arms, or, in other words, a configuration model. Hence, a typical cluster may be built thanks to the second algorithm of Section 3.2.1.

Recall from Section 3.2.2 the following facts.

- The law of the number of arms of a particle picked uniformly at random is μ_i .
- When one picks an arm uniformly at random, the number of outer arms of the particle to which it is attached is $\nu = \hat{\mu}_i$.
- Moreover, when few particles have been picked, these laws do not vary much.

Now, let us wonder what a typical cluster looks like, that is, consider a particle uniformly at random, call it 1, and let us build its cluster. Since N is large, it is very unlikely that two of its arms bind together, or grab the same particle. The law of the number of particles to which it is bound is thus approximately μ_i .

Then, take an arm b . By construction, b chooses an arm uniformly at random, attached to a particle j , and hence, from the above remarks, the law of the number of outer arms of j is very close to ν . We can go on and see that the number of outer arms of any particle but 1 is approximately ν . When few particles are paired, there is only a very small probability that cycles, loops or double-edges occur and thus, the cluster of 1 is approximately a Galton-Watson tree $\mathbb{GW}_{\mu_i, \nu}$, whose ancestor has reproduction law μ_i and

the other individuals have reproduction law ν . We may check a posteriori that ν has mean less than 1 (since we assumed $T_{\text{gel}} = +\infty$), so these trees indeed are finite.

To recover the limiting concentrations from this remark, consider $p(m)$, the probability that a particle, picked uniformly at random, is in a cluster of size m in the final state. On the one hand,

$$p(m) = \frac{mc_\infty(m)}{\langle c_\infty, m \rangle} = \frac{mc_\infty(m)}{\langle \mu, 1 \rangle}$$

by definition. On the other hand, we just saw that this is (approximately) the probability that the size $T(\mu, \nu)$ of a $\mathbb{G}\mathbb{W}_{\mu, \nu}$ -tree is m , so that we get

$$c_\infty(m) = \frac{1}{m} \langle \mu, 1 \rangle \mathbb{P}(T(\mu_i, \nu) = m).$$

But, by Dwass' formula [9],

$$\mathbb{P}(T(\mu_i, \nu) = m) = \frac{1}{\langle \mu, 1 \rangle} \frac{1}{m-1} \nu^{*m}(m-2),$$

hence verifying the result. This argument is made rigorous in the work [4] by Bertoin and Sidoravicius subsequent to [3].

4 A two-type limited aggregation model

4.1 Model

The second chapter (following this one) of this thesis deals with a two-type limited aggregation model. The material of this chapter is taken from an article [43] published in the *Journal of Statistical Physics* and entitled “A model for coagulation with mating”.

The title is explained by the “sexed” model we consider. We indeed assume that we are given particles with two types of arms, namely the male and female arms. Only male and female arms can bind, and these arms are used to perform the coagulations: two clusters coalesce when a male arm of one binds with a female one of the other. There are two main motivations to this work. The first is the idea of ionic bonds, which is natural when considering a medium filled with ions, which bind to create polymers. The second motivation is to bring together the two models considered by Bertoin in [3]. We shall however only discuss the relation with the *symmetric* model (in the terminology of [3]) (1.3) described above. The relation with the other model is explained in Chapter 2.

As coagulations occur in the sexed model, clusters of particles are created. These clusters are characterized by a triplet (a, b, m) , where $a \in \mathbb{N}$ is its number of available (i.e. not yet linked) male arms, $b \in \mathbb{N}$ its number of available female arms and $m \in \mathbb{N}^*$ its mass, i.e. the number of particles it consists in.

Two clusters may coagulate when one has an (available) female arm and the other has an (available) male arm, and when a coagulation occurs, the used arms merge and create an edge. Hence, we may only observe the transition

$$\{(a, b, m), (a', b', m')\} \rightarrow (a + a' - 1, b + b' - 1, m + m').$$

We will assume that this transition occurs with a rate given by the number of pairs formed of a female arm and of a male arm, that is $a'b + ab'$.

Mathematically, define $c_t(a, b, m)$ the concentration of clusters with a male arms, b female arms and mass m . Then these concentration abide by the *sexed Smoluchowski*

equation given by

$$\begin{aligned} \frac{d}{dt}c_t(a, b, m) &= \frac{1}{2} \sum_{m'=1}^{m-1} \sum_{a'=0}^{a+1} \sum_{b'=0}^{b+1} (a'(b+1-b') + b'(a+1-a')) \times \\ &\quad c_t(a', b', m') c_t(a+1-a', b+1-b', m-m') \\ &\quad - \sum_{m' \geq 1} \sum_{a' \geq 0} \sum_{b' \geq 0} (ab' + a'b) c_t(a, b, m) c_t(a', b', m'). \end{aligned} \quad (1.6)$$

As for (1.1) and (1.3), the first term of the RHS accounts for the creation of (a, b, m) -clusters by coagulation of (a', b', m') and $(a+1-a', b+1-b', m-m')$ -clusters. The second term accounts for the disappearance of such clusters, by coagulation with other clusters.

Our goal is to study this equation as done in [3] for (1.3). More precisely, we want to know about the existence and uniqueness to a solution, and whether we may observe similar phenomena as for (1.3), namely whether gelation occurs, whether we can give explicit solutions and compute the limiting concentrations.

4.2 Results

4.2.1 Well-posedness

Our main result is existence and uniqueness of a solution to (1.6) up to a critical time. Let $S = \mathbb{N} \times \mathbb{N} \times \mathbb{N}^*$. For two nonnegative functions $c, f : S \rightarrow \mathbb{R}^+$, define

$$\langle c, f \rangle = \sum_{p \in S} c(p) f(p).$$

We also write a for the function $f(a, b, m) = a$, b for $f(a, b, m) = b$, and so on.

In all the statements and proofs, we are given nonnegative initial concentrations c_0 such that $\langle c_0, 1 \rangle < +\infty$, $\langle c_0, a \rangle = \langle c_0, b \rangle = 1$ and $\langle c_0, a^2 + b^2 \rangle < +\infty$. We may then define

$$M = \langle c_0, ab \rangle + \sqrt{\langle c_0, a^2 - a \rangle \langle c_0, b^2 - b \rangle}$$

and

$$T_{\text{gel}} = \begin{cases} +\infty & \text{if } M \leq 1 \\ \frac{1}{M-1} & \text{if } M > 1. \end{cases} \quad (1.7)$$

We thus obtain the following result.

Theorem 4.1. (i) *Smoluchowski's equation (1.6) with initial conditions c_0 has a unique solution (c_t) defined on $[0, T_{\text{gel}})$.*

(ii) *For $t \in [0, T_{\text{gel}})$, $\sup_{s \in [0, t]} \langle c_s, a^2 + b^2 \rangle < +\infty$, and $\langle c_t, a^2 + b^2 \rangle \rightarrow +\infty$ when $t \rightarrow T_{\text{gel}}$.*

(iii) *The total mass $\langle c_t, m \rangle$ is constant on $[0, T_{\text{gel}})$.*

It is obviously very similar to Theorem 3.1, so carrying out the same discussion is probably not worth it. Let us rather insist on its consequences.

4.2.2 Explicit solutions

In [3], Bertoin obtained explicit solutions for (1.3) starting from monodisperse initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$. However, this cannot be achieved in our case for any initial conditions $c_0(a, b, m) = \mu(a, b)\mathbb{1}_{\{m=1\}}$, but only in some special cases. Probably the most interesting case is the one which allows to recover Bertoin's results for (1.3).

To do so, a natural idea is to give, at time 0, i.i.d. numbers of arms to each particle and then give each of these arms independently a gender with probability 1/2. The knowledge of the genders should then be irrelevant, and forgetting about them should allow to recover Equation (1.3). So consider

$$\mu(a, b) = \mu_1(a + b) \binom{a + b}{b} \frac{1}{2^{a+b}},$$

where μ_1 is a measure with mean 2, so that $\langle c_0, a \rangle = \langle c_0, b \rangle = 1$. In other words, this means that the particles have i.i.d. number of arms given by $\mu_1/2$, and that each arm is independently with probability 1/2 male or female.

Denote $c_t(k, m)$ the solution to (1.3) with monodisperse initial conditions $c_0(k, m) = (\mu_1/2)(k)\mathbb{1}_{\{m=1\}}$. Then, we can see that

$$\sum_{a+b=k} c_t(a, b, m) = 2c_t(k, m)$$

and

$$c_t(a, b, m) = \frac{1}{2} \binom{a + b}{b} \frac{1}{2^{a+b}} c_t(a + b, m).$$

Hence, at any given time, the distribution of the number of male (or female) arms is still binomial. So, if at some time we choose to reassign to each arm a gender uniformly and independently, and let the system evolve on from this state, no difference would be observed. Or we could forget about the genders at some time t and observe the state at time t of Bertoin's model (1.3). Or, finally, we could watch a system evolve like (1.3) starting from an arm distribution $\mu_1/2$, and then at some time give the arms a gender uniformly at random and independently. The evolution afterwards will be the evolution of the sexed model with initial arm distribution μ .

Note also that the factor 2 in these equations is irrelevant, since it comes only from the normalization: in our model, the total concentration of arms in the medium is 2, while it is 1 in Bertoin's model. Finally, it is worth noticing that the gelation time for this model is the same as in the symmetric model with initial distribution $\mu_1/2$.

4.2.3 Limiting concentrations

As in Bertoin's result, we may want to compute the limiting concentrations whenever the gelation time T_{gel} is infinite. So let us start from monodisperse initial conditions $c_0(a, b, m) = \mu(a, b)\mathbb{1}_{\{m=1\}}$ with $\langle c_0, a \rangle = \langle c_0, b \rangle = 1$. We may then define the probability measures $\mu_m(a, b) = (b + 1)\mu(a, b + 1)$, $\mu_f(a, b) = (a + 1)\mu(a + 1, b)$ and $\mu_i(a, b) = \mu(a, b)/\langle \mu, 1 \rangle$.

Let $T(\mu_i, \mu_m, \mu_f)$ be the size of a two-type Galton-Watson tree $\mathbb{GW}_{\mu_i, \mu_m, \mu_f}$, whose ancestor has reproduction law μ_i , and the other individuals have reproduction law μ_m for the males, μ_f for the females. The assumption that $T_{\text{gel}} = +\infty$ ensures that it is a subcritical Galton-Watson tree. When it is not degenerate (i.e. has a positive probability

of being infinite even though it is subcritical, see [24]), we may show that we have the convergence

$$c_t(a, b, m) \rightarrow c_\infty(m) \mathbb{1}_{\{a=b=0\}}$$

where

$$c_\infty(m) = \frac{1}{m} \langle \mu, 1 \rangle \mathbb{P}(T(\mu_i, \mu_m, \mu_f) = m).$$

In this formula, the factor $\langle \mu, 1 \rangle$ stems from the normalization and is irrelevant. Having said that, this formula may be interpreted as in Bertoin's case with only one type of arm (see Section 3.3.2). Note indeed that μ_i is the law of the number of (male and female) arms of a particle picked uniformly at random, and that, when a male (resp. female) arm is picked uniformly at random, μ_m (resp. μ_f) is the law of the outer number of arms of the particle to which this arm is attached. In other words, a typical cluster in solution should be a $\mathbb{GW}_{\mu_i, \mu_m, \mu_f}$ -tree.

4.3 A microscopic model

The last section of this first work is devoted to providing a microscopic model to (1.6), so as to give a more proper justification to this equation. As pointed out earlier, the convergence of this kind of models has been first established by Jeon [27] and Norris [45]. The framework here is slightly different because we consider particles with arms. Moreover, we only study a multiplicative kernel, and the proof is made quite simpler by using PDE techniques developed earlier in the paper.

Let us define the model a bit informally (in particular, the jump rates are not exactly those below). Define, for $p = (a, b, m)$, $p' = (a', b', m')$, $p \circ p' = (a + a' - 1, b + b' - 1, m + m')$ and

$$\Delta_{p,p'}(q) = \mathbb{1}_{\{q=p \circ p'\}} - \mathbb{1}_{\{q=p\}} - \mathbb{1}_{\{q=p'\}}, \quad q \in S.$$

Then, we define a pure-jump Markov process $(\mathbf{X}_t^{(N)}(p), p \in S)_{t \geq 0}$, where the jumps

$$\mathbf{X}_t^{(N)} \rightarrow \mathbf{X}_t^{(N)} + \Delta_{p,p'}$$

occur at rate $ab' + a'b$. Rescale the process by letting $c_t^{(N)} = \frac{1}{N} X_{t/N}^{(N)}$, and assume that the initial conditions are non-random and verify

- for every $p \in S$, $c_0^{(N)}(p) \rightarrow c_0(p)$ for some $c_0(p) \geq 0$,
- $\langle c_0^{(N)}, a \rangle \rightarrow \langle c_0, a \rangle = 1$, $\langle c_0^{(N)}, b \rangle \rightarrow \langle c_0, b \rangle = 1$,
- $\langle c_0, a^2 + b^2 \rangle < +\infty$.

In particular, $c^{(N)}$ belongs to

$$E = \left\{ C \in [0, 1]^S, \sum_{(a,b,m) \in S} (a + b + m) C(a, b, m) \leq M \right\}$$

for some large enough M , where E , endowed with the ℓ^1 distance, is a Polish space. If we define T_{gel} as in (1.7), then we have the following convergence result.

Theorem 4.2. *The process $(c_t^{(N)})_{t \in [0, T_{\text{gel}}]}$ converges in the Skorokhod space $\mathbb{D}([0, T_{\text{gel}}], E)$ to the unique solution of Smoluchowski's equation (1.6) on $[0, T_{\text{gel}}]$.*

This gives a rigorous justification for the equation (1.6) we consider. It is also worth noticing that the convergence takes place only before the gelation time, as was to be expected upon knowing the results on the standard Smoluchowski equation (1.1).

5 Post-gelation uniqueness of coagulation equations

The second part of this thesis is mainly constituted of a joint work [44] with Lorenzo Zambotti. We prove therein that there is a unique solution on the whole of \mathbb{R}^+ to Smoluchowski's equation (1.1) or (1.3), in particular when gelation occurs. As far as we know, this is the first rigorous uniqueness result proved in that case, except for Kokholm's result [29] on (1.1) for monodisperse initial conditions.

5.1 Post-gelation behavior for the standard Smoluchowski equation

5.1.1 Statement

Let us give here our precise result concerning Equation (1.1). The actual result of [44] deals with a version of the equation with continuous masses, but we only give here the statement for discrete masses which is easier to understand. We start from nonzero initial conditions c_0 such that $\langle c_0, 1 \rangle < +\infty$, and we define

$$T_{\text{gel}} = \frac{1}{\langle c_0, m^2 \rangle} \in [0, +\infty).$$

Theorem 5.1. *Smoluchowski's equation (1.1) with the multiplicative kernel has a unique solution (c_t) on \mathbb{R}^+ . It has the following properties.*

1. *The total mass $M_t = \langle c_t, m \rangle$ is continuous on \mathbb{R}^+ . It is constant on $[0, T_{\text{gel}}]$ and strictly decreasing on $[T_{\text{gel}}, +\infty)$. It is analytic on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$.*
2. *Let $m_0 = \inf\{m \geq 1, c_0(m) \neq 0\} \in \mathbb{N}^*$. Then, when $t \rightarrow +\infty$,*

$$M_t \sim \frac{1}{m_0 t}.$$

3. *The second moment $\langle c_t, m^2 \rangle$ is finite for $t \in [0, T_{\text{gel}})$ and infinite for $t \in [T_{\text{gel}}, +\infty)$.*

5.1.2 Remarks

This result really explains rigorously the heuristics given at the beginning of this introduction and in several papers from the Physics literature [13, 12, 36, 53, 59]. In particular, the time T_{gel} at which the second moment $\langle c_t, m^2 \rangle$ explodes is precisely the gelation time, as we already discussed in Section 2.4.2. This is obviously also reminiscent of McLeod's result (Theorem 2.1).

A most interesting feature is the continuity of the mass. This is a priori not obvious, since at the gelation time, we may think that a positive fraction of the mass would fall into the gel. Proving this fact is actually at the very heart of the proof, the other results being quite easily deduced from this one.

Finally, let us note that the mass behaves asymptotically like $1/t$, a phenomenon which was already observed in the monodisperse case [29]. The factor of $1/t$ depends on the smallest masses in the system at time 0 (and thus at any time). It is however quite intriguing that a tiny fraction of particles of unit mass at time 0 is enough to ensure a factor of 1. There is in particular no continuity in that respect. In the case of continuous masses, the mass may decrease slower than $1/t$ if there exists *dust*, i.e. particles with arbitrarily small mass (see Chapter 3 for some details).

5.1.3 Comparison with Flory's equation

Without stating the result, let us end with a few remarks on Flory's equation. In that case, we also obtain the existence and uniqueness of a solution, result already obtained by Norris [46] in a much more general setting. We can also show that, starting from the same initial conditions, the mass is, after gelation, strictly smaller in Flory's than in Smoluchowski's case. The mass even actually decreases exponentially fast, to be compared with the decrease in $1/t$ above.

This may at first sight seem natural, since in Flory's model, the gel attracts the clusters, so the rate at which clusters fall into the gel should be higher. However, it turns out³ that the clusters not in the gel are larger in Smoluchowski's than in Flory's model. Hence, in the second case, fewer clusters fall into the gel because they coagulate and reach a giant size. But they do fall because of interactions with the gel, which do not exist in Smoluchowski's model. A priori, it is not obvious which effect should prevail.

5.2 Post-gelation behavior for the model with limited aggregations

Let us now describe the results concerning (1.3) since they are the motivation for the study undertaken in the last part of this thesis. We start with initial concentrations $(c_0(a, m), a \in \mathbb{N}, m \in \mathbb{N}^*)$ such that $\langle c_0, 1 \rangle < +\infty$, $A := \langle c_0, a \rangle \in (0, +\infty]$ and $K := \langle c_0, a(a-1) \rangle \in [0, +\infty]$. Then $K = +\infty$ whenever $A = +\infty$. We may then let

$$T_{\text{gel}} = \begin{cases} \frac{1}{K-A} & \text{if } A < K < +\infty \\ 0 & \text{if } K = +\infty \\ +\infty & \text{if } K \leq A < +\infty. \end{cases} \quad (1.8)$$

Theorem 5.2. *Equation (1.3) has a unique solution (c_t) defined on \mathbb{R}^+ . When $T_{\text{gel}} < +\infty$, it enjoys the following properties.*

- *The mass $M_t := \langle c_t, m \rangle$ is continuous, constant on $[0, T_{\text{gel}})$, strictly decreasing on $[T_{\text{gel}}, +\infty)$, and analytic on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$.*
- *The mean number of arms $A_t := \langle c_t, a \rangle$ is continuous and strictly decreasing on \mathbb{R}^+ , and analytic on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$.*
- *The second moment $\langle c_t, a^2 \rangle$ is finite on $[0, T_{\text{gel}})$, infinite on $[T_{\text{gel}}, +\infty)$.*

Moreover, representation formulas for the generating function of c_t are given. They are however significantly more cumbersome than the explicit formulas of [3].

There are obvious parallels to be made with the result above, on which we shall not dwell. The only difference lies in the decrease of A_t , which is actually quite similar to that of the mass. Indeed, the "normal" (if no gelation were to occur) behavior of the mass is to be constant. The gelation time is precisely the time when this stops holding. Similarly, the normal behavior of A_t is to be equal to $A_0/(1 + tA_0)$, which corresponds to the continuous limit of the pure-death process obtained by killing a couple of arms when its clock rings, as explained in Section 3.3. The gelation time is once again precisely the time when this normal behavior is altered.

³But this will be clear only after the study of a microscopic model for Smoluchowski's equation undertaken in the last part of this thesis. See however Section 6.3 for more explanations.

5.3 Limiting concentrations

The representation formula which are obtained during the proof of the result above allows to compute basically any quantity of interest. The expressions are particularly simple when starting from monodisperse initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$, where μ has a second moment, and having $t \rightarrow +\infty$. Define $\nu(k) = (k+1)\mu(k+1)$. We assume that

- there are particles with one arm, or, in symbols, that $\mu(1) > 0$, since otherwise it is easy to see that nothing but the initial particles with no arm remains in solution in the final state,
- and that $T_{\text{gel}} < +\infty$, i.e. that $\langle \mu, a \rangle < \langle \nu, a \rangle$.

In particular, this readily implies that the equation

$$G_\nu(\eta) = \eta G'_\nu(\eta) \tag{1.9}$$

has a unique solution, where G_ν denotes the generating function of ν .

Corollary 5.3. *When $t \rightarrow +\infty$,*

$$c_t(a, m) \rightarrow c_\infty(m)\mathbb{1}_{\{a=0\}}$$

in $\ell^1(\mathbb{N} \times \mathbb{N}^*)$, where

$$c_\infty(m) = \frac{1}{m(m-1)}\beta^{m-1}\nu^{*m}(m-2) \tag{1.10}$$

and β is given by

$$\beta = \frac{\eta}{G_\nu(\eta)} = \frac{1}{G'_\nu(\eta)}. \tag{1.11}$$

Moreover, the mass at infinity is given by

$$\langle c_\infty, m \rangle = G_\mu(\eta). \tag{1.12}$$

The link with the result without gelation is once again really clear, the only difference lying in the appearance of the factor β in the expression of $c_\infty(m)$. However, at this point, it is not clear where it stems from, and giving a probabilistic interpretation for that constant is actually the incentive for the last part of this thesis.

6 A microscopic model for Smoluchowski's equation

The last chapter is dedicated to the study of a microscopic model for Smoluchowski's equation, whose stated goal is to explain Formula (1.10), as was done in [4] in the case gelation does not occur. The methods used are however quite different and rely heavily on random graphs considerations. This part is extracted from a joint work [41] with Mathieu Merle.

6.1 Model

The model is similar to those already mentioned in Sections 2.4 and 3.3. We start from N particles with empirical distribution of arms $\mu^{(N)}$ (i.e., for each k , there are $N\mu^{(N)}(k)$ particles with k arms). We then set i.i.d. exponential clocks on each arm. When a clock rings, the corresponding arm is *activated*, so an arm will be said to be *active* if the corresponding clock has rung, and *available* if it has not.

At each time, the model provides a multigraph \mathcal{C}_t . We say that a particle or arm is in solution if its cluster (i.e. connected component) has size less or equal than some $\alpha(N)$, and is in the gel otherwise. We then join pair of arms of particles whose respective clocks ring consecutively *when they are both in solution* and when they are not already paired. To be more precise, when an arm b is activated:

- if b is in the gel at that time, or if it is still in solution *and* there is no other unpaired and active arm in solution, nothing happens.
- If on the other hand, b is in solution *and* there is another unpaired an active arm in solution, say b' , then the arms b and b' are bound together, i.e. a new edge in the graph is created.

We also proceed in such a way that the process is càdlàg.

We obviously need some assumptions on $\alpha(N)$ and $\mu^{(N)}$. For $\mu^{(N)}$, classical results on random graphs, as explained in Section 3.2.2, require that there exists a probability μ such that the sequence $(\mu^{(N)})$ converges to μ weakly and $\langle \mu^{(N)}, k^2 \rangle \rightarrow \langle \mu, k^2 \rangle < +\infty$. This is also the assumption we will make. For technical reasons, we also require that μ has a third moment, i.e.

$$\langle \mu, k^3 \rangle < +\infty,$$

though we believe that a finite second moment should be enough.

Concerning $\alpha(N)$, it is clear from Fournier and Laurençot's work [19] that we should take $1 \ll \alpha(N) \ll N$. Actually, for technical reasons, we need

$$\frac{\alpha(N)}{N} \xrightarrow{N \rightarrow \infty} 0 \quad \text{and} \quad \frac{\alpha(N)}{N^{1/3}} \xrightarrow{N \rightarrow \infty} +\infty,$$

though we also believe that having $\alpha(N) \rightarrow +\infty$ should be enough.

6.2 Post-gelation result

At any time $t \geq 0$, this model provides a good microscopic model for (1.3), in that the techniques of [19] can be adapted to show that the concentration of clusters with a available arms and mass m converge to a solution of a *time-changed* Smoluchowski's equation (1.3). This time-change comes from the fact that the model is slightly different from the one explained in Section 3.3, where clocks are set on *couples* of arms, whereas here, clocks are set on arms. However, this time-change is explicit and is a diffeomorphism of \mathbb{R}^+ , so it hardly makes any difference. In particular, the limiting quantities as $t \rightarrow +\infty$ are the same.

Now, our main interest is to study two relevant quantities, namely

- the proportion of particles in solution at time t , denoted $n_t^{(N)}$,
- the empirical distribution $\pi_t^{(N)}$ of activated arms of the particles in solution (that is, for each k , $\pi_t^{(N)}(k)$ is the proportion of particles in solution with k activated arms).

Our main result is the convergence of these two quantities in the space $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+ \times \mathcal{M}_1)$ of càdlàg processes with values in $\mathbb{R}^+ \times \mathcal{M}_1$, where \mathcal{M}_1 is the space of probabilities on \mathbb{N} .

Theorem 6.1. *The sequence $(n^{(N)}, \pi^{(N)})$ converges in $D(\mathbb{R}^+, \mathbb{R}^+ \times \mathcal{M}_1)$ as $N \rightarrow +\infty$ to some continuous deterministic (n, π) . Moreover, for $t \geq T_{\text{gel}}$, π_t is critical, in the sense that $\langle \hat{\pi}_t, k \rangle = 1$.*

We can also provide explicit formulas for n_t and for the generating function of π_t , so we can compute basically any quantity we wish.

6.3 Remarks

This result actually describes precisely the state in solution at any time t . Indeed, we will see in Chapter 4 that, at time t , the state in solution is approximately a configuration model $CM(n_t N, \pi_t)$. Hence, the same description of a typical cluster in solution as in Section 3.3.2 can be carried out. In particular, it means that a particle picked uniformly at random in solution is the root of a $\mathbb{GW}_{\pi_t, \hat{\pi}_t}$ -tree.

Probably the most interesting part of this result is the *post-gelation Self-Organized-Criticality* (SOC), i.e. in symbols $\langle \hat{\pi}_t, k \rangle = 1$ for $t \geq T_{\text{gel}}$. The fact that we let all the big clusters fall into the gel indeed allows for the model to remain at criticality, after it has reached it at time T_{gel} . In comparison, this is not the case for Flory's model, which is subcritical before T_{gel} , critical at T_{gel} and becomes subcritical again just afterwards. Informally, the criticality is achieved because the two phenomena of activation of arms and falls into the gel compete to reach an equilibrium. This SOC phenomenon is well-known in the Physics literature, but quite seldom proved mathematically. It is however worth mentioning Ráth and Tóth beautiful paper [47], where it is proved, under similar heuristics (growth of trees vs. forest fires), that SOC occurs.

In our case, the post-gelation SOC means that the typical clusters in solution are *critical* Galton-Watson tree, which are thus a.s. finite but quite large, whereas in Flory's model, the typical clusters in solution are *subcritical* trees, and so are significantly smaller.

6.4 Final state

Let us conclude this introduction and come full circle by explaining Formula (1.5). So consider the final state of the system. Recall that η is defined as the unique solution to Equation (1.9).

Corollary 6.2. *The mass converges to a limit*

$$n_\infty = G_\mu(\eta) \tag{1.13}$$

and π_t converges to a critical probability π_∞ with generating function

$$G_{\pi_\infty}(x) = \frac{G_\mu(\eta x)}{G_\mu(\eta)}, \quad x \in [0, 1]. \tag{1.14}$$

The first obvious remark is that Formulas (1.12) and (1.13) agree. Now, as already noted above, a typical particle in solution is the root of a $\mathbb{GW}_{\pi_\infty, \hat{\pi}_\infty}$ -tree. As in Section 3.3.2, we may then say that $c_\infty(m)$, the concentration, in a unit volume, of clusters of mass m and with no arms in the final state, can be written as

$$c_\infty(m) = \frac{1}{m} \mathbb{P}(T(\pi_\infty, \hat{\pi}_\infty) = m).$$

But Dwass' formula [9] yields

$$\mathbb{P}(T(\pi_\infty, \hat{\pi}_\infty) = m) = (m-1)^{-1} \beta^{m-1} \nu^{*m} (m-2)$$

and we have therefore recovered (1.5).

Chapter 2

A model for coagulation with mating

The material of this chapter is taken from an article [43] published in the *Journal of Statistical Physics*.

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1 Introduction

1.1 Model

In this chapter, we study a “sexed” model of coagulation of clusters, generalizing the original model (1.1) of Smoluchowski [57], and Bertoin’s model (1.3) introduced in [3]. The model is sexed in that we consider particles which are initially given a certain number of male and female arms. These arms are used to perform the coagulations: two clusters coagulate when a male arm of one and a female arm of another bind. Introducing this kind of model is natural when we think of Chemistry. For instance, consider male particles (which have only male arms), and female particles. Then a coagulation between a male and a female particle can be thought of as an ionic bond between a cation and an anion. This kind of models has also been investigated in the Physics literature. For instance, in [48, 49], the authors study coalescing monomers with two types, A and B, with bonding only allowed between A and B, hence forming alternating linear polymers. In this work, this corresponds to giving to each particle exactly one male arm and one female arm.

When (elementary) particles coalesce, clusters are created. In our model, a cluster is characterized by a triple (a, b, m) , $a \in \mathbb{N}$ being its number of male arms, $b \in \mathbb{N}$ its number of female arms, and $m \in \mathbb{N}^*$ its mass. Two clusters may coagulate when one has a female arm and the other has a male arm, and when a coagulation occurs, the used arms merge and create an edge, so they cannot be used again. Hence, we may only observe the transition

$$\{(a, b, m), (a', b', m')\} \rightarrow (a + a' - 1, b + b' - 1, m + m').$$

We will assume that this transition occurs with a rate given by the number of pairs formed of a female arm and of a male arm, that is $a'b + ab'$. We wish to study how the concentration of each type of cluster evolves when time passes. The precise mathematical formulation of this problem is given in Section 2.

1.2 Bertoin’s models

Our model can be seen as a variation of the two models of Bertoin introduced in [3]. In this work, he considers coalescing clusters characterized by a number a of arms and a mass m , and studies two models. We already discussed in the introduction the “symmetric” model (1.3) (in the terminology of [3]), where *each pair of arms* is activated independently. The concentration $c_t(a, m)$ of (a, m) -clusters is then governed by the equation

$$\begin{aligned} \frac{d}{dt} c_t(a, m) = & \frac{1}{2} \sum_{a'=1}^{a+1} \sum_{m'=1}^{m-1} a'(a+2-a') c_t(a', m') c_t(a+2-a', m-m') \\ & - \sum_{a' \geq 1} \sum_{m' \geq 1} a a' c_t(a, m) c_t(a', m'). \end{aligned} \tag{2.1}$$

In that case, there is a critical time such that there is a unique solution up to this time. Moreover, for monodisperse initial conditions, explicit solutions are given, which can be related to Galton-Watson processes at the limit, whenever the critical time is infinite.

The second model (actually the first in [3]) is an “oriented” model, where *each arm* is activated independently, and then chooses to bind to another cluster chosen uniformly at random. Hence, only one arm is consumed when a coagulation occurs, and we only observe the transitions

$$\{(a, m), (a', m')\} \rightarrow (a + a' - 1, m + m').$$

The evolution of the concentrations is then given by the equation

$$\begin{aligned} \frac{d}{dt}c_t(a, m) = & \frac{1}{2} \sum_{a'=0}^{a+1} \sum_{m'=1}^{m-1} (a+1)c_t(a', m')c_t(a+1-a', m-m') \\ & - \sum_{a' \geq 0} \sum_{m' \geq 1} (a+a')c_t(a, m)c_t(a', m'). \end{aligned} \quad (2.2)$$

The main result of [3] in that case is that there is a unique solution on \mathbb{R}^+ , for which explicit formulas are given.

We will see that our sexed model contains Bertoin’s: the oriented model corresponds indeed to ours if each particle is given precisely one female arm, and the symmetric model corresponds to the sexed one if the arms are given a gender uniformly at random.

1.3 Results and plan of the paper

This paper is divided in two parts. In the first one (Sections 2 to 6), we shall study the sexed Smoluchowski’s equation. We first (Section 2) introduce the problem and state our main result, viz. the well-posedness of the sexed Smoluchowski equation up to some critical time. In Section 3, we provide some preliminary results and prove some physically intuitive facts. Then (Section 4), we prove our main result. The tools used are analogous to those in [3], but since we are dealing with a two-dimensional problem, several technical issues need to be addressed. The outline of the proof is as follows. First, we transform the system into a PDE by considering the generating functions of the concentrations. This PDE is not quasilinear, but it may however be solved by the method of characteristics. This method requires the inversion of a two-dimensional mapping, and this can be done precisely up to the critical time.

Unfortunately, even for monodisperse initial conditions, the inversion is not explicit (one could use the two-variable Lagrange inversion formula, but in general, the expression it provides is too cumbersome). Nonetheless, in some specific cases (Section 5), the Lagrange Inversion Formula yields explicit results. In particular, we recover the solutions obtained in [3]. Finally, we show (Section 6) that there exist limiting concentrations when $t \rightarrow +\infty$, and that they are related to the distribution of the total progeny of some two-type Galton-Watson process.

In the second part (Section 7), we study a microscopic model. Given a finite number of particles, we let them coagulate and observe the evolution of the concentrations of the different types of clusters. This is a Marcus-Lushnikov process, and we show that it converges, before the critical time, to a process solving Smoluchowski’s equation (2.3). As pointed out in the introduction, this kind of convergence had already been proved by Norris ([45], see as well [27]). The difference here is that we consider a model with male or female arms. Moreover, the proof is made much easier by the fact that the rate of coagulation is explicit. In particular, we will appeal to the PDE obtained in the first part. This discrete model provides a justification to the sexed Smoluchowski’s equation (2.3).

Finally, note that our construction can also provide a model for random oriented graphs, since a coagulation can be seen as the creation of an oriented edge between two

vertices in a graph, whose orientation is given e.g. from the male arm to the female arm. Hence, we can consider a large number n of particles and let them coagulate. When all the coagulations are performed, we obtain a set of oriented graphs. When $n \rightarrow +\infty$, we may wonder what the distribution of their sizes is, what a typical graph looks like, etc. A heuristic answer, motivated by the works [4, 5], and by the results obtained in this paper (Section 6), is that a typical graph would be a two-type Galton-Watson tree, provided there are few arms (in the notations of this chapter, this means that $T_{\text{gel}} = +\infty$ and μ is not degenerate).

2 Setting and results

2.1 Notations

Let us first introduce some notations as well as the sexed Smoluchowski equation, and state our main result.

- $\mathbb{N} = \{0, 1, 2, \dots\}$ and $\mathbb{N}^* = \{1, 2, \dots\}$.
- $S = \mathbb{N} \times \mathbb{N} \times \mathbb{N}^*$ is the set of the different types of clusters. A generic element of S will be denoted by p , and if $p = (a, b, m)$, we will call a p -cluster a cluster with a male arms, b female arms, and mass m .
- For $p = (a, b, m) \in S$ and $p' = (a', b', m') \in S$, we will denote

$$p.p' = a'b + ab'$$

the *rate* of coagulation and

$$p \circ p' = (a + a' - 1, b + b' - 1, m + m')$$

the type of the cluster resulting from such a coagulation. We say that $p' \preceq p$ if $a' \leq a + 1$, $b' \leq b + 1$ and $m' \leq m - 1$. When $p' \preceq p$, we write

$$p \setminus p' = (a + 1 - a', b + 1 - b', m - m')$$

the type of cluster such that $p' \circ (p \setminus p') = p$.

- For two functions $c, f : S \rightarrow \mathbb{R}$, we will denote, when the series converge absolutely,

$$\langle c, f \rangle := \sum_{p \in S} c(p)f(p).$$

When using this notation, we will write, with a slight abuse of notation, a for the function $(a, b, m) \mapsto a$, b for $(a, b, m) \mapsto b$, etc.

Let us recall our goal. We are interested in a system of coagulating clusters with male and female arms. We assume that each couple formed of a p -cluster and of a p' -cluster coagulates at rate $p.p'$, to form a $p \circ p'$ -cluster. This means that if we denote $c_t(p)$ the concentration of p -clusters, then $(c_t(p), p \in S)$ solves the following infinite system of nonlinear differential equations

$$\frac{d}{dt}c_t(p) = \frac{1}{2} \sum_{p' \preceq p} p'.(p \setminus p')c_t(p')c_t(p \setminus p') - \sum_{p' \in S} p.p'c_t(p)c_t(p'). \quad (2.3)$$

The first term accounts for the creation of p -clusters by coagulation of p' - and $p \setminus p'$ -clusters (the factor $1/2$ stems from an obvious symmetry). The second accounts for the disappearance of p -clusters by coagulation with other clusters.

To see the resemblance with Bertoin's equations (2.1) and (2.2), let us once write down this formula in full: for all $(a, b, m) \in S$, the concentration of (a, b, m) -clusters verifies

$$\begin{aligned} \frac{d}{dt} c_t(a, b, m) = & \frac{1}{2} \sum_{m'=1}^{m-1} \sum_{a'=0}^{a+1} \sum_{b'=0}^{b+1} (a'(b+1-b') + b'(a+1-a')) \times \\ & c_t(a', b', m') c_t(a+1-a', b+1-b', m-m') \\ & - \sum_{m' \geq 1} \sum_{a' \geq 0} \sum_{b' \geq 0} (ab' + a'b) c_t(a, b, m) c_t(a', b', m'). \end{aligned}$$

Let us now define what we call a *solution* to the sexed Smoluchowski equation.

Definition 2.1. We call a family $(c_t(p), p \in S, t \in [0, T])$ of differentiable functions a solution of Smoluchowski's equation (2.3), if

1. for every $t \in [0, T]$, $\langle |c_t|, a + b \rangle < +\infty$,
2. the family $(c_t(p))$ solves the system (2.3) for $t \in [0, T]$.

Remark 2.2. • We will always assume that at time 0, $\langle c_0, a + b + 1 \rangle < +\infty$, and that the mean number of male arms $\langle c_0, a \rangle$ and the mean number of female arms $\langle c_0, b \rangle$ are equal. Physically, it is then obvious that they will remain equal as time passes. This shall be proven later on, in Lemma 3.5.

- It is easy to see that if $(c_t)_{t \in [0, T]}$ is a solution to (2.3) with initial conditions c_0 , and $\lambda > 0$, then $(\lambda c_{t/\lambda^2})_{t \in [0, T]}$ is a solution to (2.3) with initial conditions λc_0 . Hence, it is enough to assume that $\langle c_0, a \rangle = \langle c_0, b \rangle = 1$, what will always be the case from now on.

2.2 Main result

Our main result is existence and uniqueness of a solution to (2.3) up to a critical time. In all the statements and proofs, we are given nonnegative initial concentrations c_0 such that $\langle c_0, 1 \rangle < +\infty$, $\langle c_0, a \rangle = \langle c_0, b \rangle = 1$ and $\langle c_0, a^2 + b^2 \rangle < +\infty$. We can then define the critical time T_{gel} .

Definition 2.3. Let

$$M = \langle c_0, ab \rangle + \sqrt{\langle c_0, a(a-1) \rangle \langle c_0, b(b-1) \rangle}$$

and define the gelation time as

$$T_{\text{gel}} = \begin{cases} +\infty & \text{if } M \leq 1 \\ \frac{1}{M-1} & \text{if } M > 1. \end{cases} \quad (2.4)$$

Remark 2.4. As explained in the introduction of this thesis, this gelation time is a priori just a technical device: after this time, the equations become much harder to solve, since, as the following theorem will show, this is actually the time when the second moment $\langle c_t, a^2 + b^2 \rangle$ explodes. However, we will not give a proof that a solution to (2.3) defined on a larger interval than $[0, T_{\text{gel}})$ sees its mass decrease, which would show that T_{gel} really

is the gelation time. That being said, we firmly believe this to be true, since this holds for Bertoin's equation (2.1) as we will show in the next chapter. The techniques of that chapter could probably be applied to the present case, though doing so would surely be really tedious.

We will also constantly use the generating function of (c_0)

$$g_0(x, y, z) := \langle c_0, x^a y^b z^m \rangle = \sum_{(a,b,m) \in \mathcal{S}} c_0(a, b, m) x^a y^b z^m.$$

Since $\langle c_0, 1 \rangle < +\infty$, g_0 is well-defined on $[0, 1]^3$. Thanks to the assumption $\langle c_0, a + b \rangle < +\infty$, its partial derivatives with respect to x and y are well-defined and continuous on $[0, 1]^3$ and they remain in $[0, 1]$. We may now state our main result.

Theorem 2.5. (i) *Smoluchowski's equation (2.3) with initial conditions c_0 has a unique solution (c_t) defined on $[0, T_{\text{gel}})$.*

(ii) *For $t \in [0, T_{\text{gel}})$, $\langle c_t, a^2 + b^2 \rangle < +\infty$, and $\langle c_t, a^2 + b^2 \rangle \rightarrow +\infty$ when $t \rightarrow T_{\text{gel}}$.*

(iii) *The total mass $\langle c_t, m \rangle$ is constant on $[0, T_{\text{gel}})$.*

(iv) *For $t \in [0, T_{\text{gel}})$ and $z \in [0, 1]$, the mapping $\phi_t(\cdot, \cdot, z)$, given for $(x, y) \in [0, 1]^2$ by*

$$\phi_t(x, y, z) = \left((1+t)x - t \frac{\partial g_0}{\partial y}(x, y, z), (1+t)y - t \frac{\partial g_0}{\partial x}(x, y, z) \right),$$

has a right-inverse $h_t = (h_t^{(1)}, h_t^{(2)})$ which is well-defined and analytic on $(0, 1)^2$. Then the generating function g_t of (c_t) is given by

$$g_t(x, y, z) = \frac{1}{1+t} \left(\tilde{H}_t^{(2)}(x, y, z) + \tilde{H}_t^{(1)}(0, y, z) \right) + G_t(z) \quad (2.5)$$

where, for $t > 0$,

$$\tilde{h}_t^{(1)} = \frac{1+t}{t} h_t^{(1)}(x, y, z) - \frac{x}{t}, \quad \tilde{h}_t^{(2)} := \frac{1+t}{t} h_t^{(2)}(x, y, z) - \frac{y}{t} \quad (2.6)$$

and

- $\tilde{H}_t^{(1)}$ *is the antiderivative of $\tilde{h}_t^{(1)}$ with respect to y , vanishing at $y = 0$,*
- $\tilde{H}_t^{(2)}$ *is the antiderivative of $\tilde{h}_t^{(2)}$ with respect to x , vanishing at $x = 0$,*
- $G_t(z)$ *is the antiderivative of*

$$\frac{\partial g_0}{\partial z} \left(h_t^{(1)}(0, 0, z), h_t^{(2)}(0, 0, z), z \right) \quad (2.7)$$

with respect to z , vanishing at 0.

The relation with Bertoin's Theorem 3.1 is obvious. The two differences are the conservation of the mass, which is implicit in [3], and the representation formula, which is given for monodisperse or polydisperse initial conditions. Bertoin gives explicit formulas in the monodisperse case, what we cannot do here, except in some special cases (see Section 5). This representation formula is however at the heart of the proof, and allows to prove the properties of the solution given in the statement.

3 Preliminary results

In this section, we give some physically intuitive results, and deduce the “weak” form of the equation.

3.1 Nonnegativeness of the solutions

Let us start with the following lemma. Recall that $c_t(p)$ is meant to model a concentration, and that $c_0(p) \geq 0$ for any $p \in S$.

Lemma 3.1. *Any solution to the sexed Smoluchowski equation remains nonnegative, that is, if $(c_t)_{t \in [0, T]}$ is a solution to (2.3) with initial conditions (c_0) , then, for all $t \in [0, T]$ and $p \in S$, $c_t(p) \geq 0$.*

Proof. Take some $t \in [0, T]$. System (2.3) gives

$$\frac{d}{dt}c_t(a, b, 1) = -c_t(a, b, 1) \sum_{m' \geq 1} \sum_{a' \geq 0} \sum_{b' \geq 0} (ab' + a'b)c_t(a', b', m') := -\gamma(t)c_t(a, b, 1).$$

Let $G(t) = \int_0^t \gamma(s) ds$. Then $c_t(a, b, 1) = c_0(a, b, 1)e^{-G(t)}$, so it remains nonnegative. Let now $m \geq 1$, and suppose that the $c_t(a, b, m')$ are nonnegative for $a, b \geq 0$ and $1 \leq m' \leq m$. For some $p = (a, b, m + 1)$, we have

$$\begin{aligned} \frac{d}{dt}c_t(p) &= \frac{1}{2} \sum_{p' \preceq p} p' \cdot (p \setminus p') c_t(p') c_t(p \setminus p') - c_t(p) \sum_{p' \in S} p \cdot p' c_t(p') \\ &= \beta(t) - c_t(p) \gamma(t). \end{aligned}$$

So we may write

$$c_t(p) = \left(c_0(p) + \int_0^t \beta(s) e^{G(s)} ds \right) e^{-G(t)}.$$

But $\beta(t) \geq 0$ since it is a linear combination with nonnegative coefficients of the $c_t(a, b, m')$ for $a, b \geq 0$ and $1 \leq m' \leq m$. So $c_t(a, b, m + 1)$ is nonnegative, what gives the result by induction. \square

3.2 Weak form of the equation

The following result is a straightforward generalization of Lemma 1 in [3]. Note however that the monotone convergence used in the proof requires that the coefficients (c_t) be nonnegative.

Lemma 3.2. (i) *If (c_t) is a solution to Smoluchowski’s equation (2.3), then $t \mapsto \langle c_t, 1 \rangle$, $t \mapsto \langle c_t, a \rangle$ and $t \mapsto \langle c_t, b \rangle$ are decreasing.*

(ii) *A family (c_t) is a solution to (2.3) if and only if it solves*

$$\frac{d}{dt} \langle c_t, f \rangle = \frac{1}{2} \sum_{p, p' \in S} p \cdot p' c_t(p) c_t(p') (f(p \circ p') - f(p) - f(p')) \quad (2.8)$$

for every bounded $f : S \rightarrow \mathbb{R}$.

Remark 3.3. • The derivative in this lemma has to be understood in the weak sense, i.e. the formula actually holds in the integral form. But if $f(a, b, m) \rightarrow 0$ when $(a, b, m) \rightarrow \infty$, then it is easy to check that the formula holds in the strong sense. The distinction is implicit in the proofs (i.e. we only write derivatives) and should not stop the reader.

- Consider in particular, the generating function of c_t , $g_t(x, y, z) = \langle c_t, x^a y^b z^m \rangle$. Then g is regular, in the sense of Definition 3.4 below.

Definition 3.4. We say that a function $(t, x, y) \mapsto g_t(x, y)$ defined on $[0, T) \times (0, 1)^2$ is regular if

- $t \mapsto g_t(x, y)$ is C^1 and $(x, y) \mapsto \frac{\partial g_t}{\partial t}(x, y)$ are C^1 ,
- $(x, y) \mapsto g_t(x, y)$ is C^2 , $t \mapsto \frac{\partial g_t}{\partial x}(x, y)$ and $t \mapsto \frac{\partial g_t}{\partial y}(x, y)$ are C^1 .

3.3 Number of arms

This result is also a generalization of Lemma 2 in [3], and stems directly from the weak form of the equation.

Lemma 3.5. Let (c_t) be a solution to Smoluchowski's equation (2.3), and let

$$\Gamma_r = \inf\{t \geq 0, \langle c_t, a^2 + b^2 \rangle > r\}, \quad \Gamma_\infty = \sup_{r>0} \Gamma_r.$$

Consider the mean numbers of male and female arms $A_t = \langle c_t, a \rangle$ and $B_t = \langle c_t, b \rangle$, and assume $A_0 = B_0 = 1$. Then

$$A_t = B_t = \frac{1}{1+t} \tag{2.9}$$

for all $t \in [0, T \wedge \Gamma_\infty)$.

3.4 Boundedness of the second moment

Recall that $\langle c_0, a^2 + b^2 \rangle < +\infty$. The next result ensures that actually $\langle c_t, a^2 + b^2 \rangle$ remains bounded on a small time-interval after 0. This ensures that Γ_∞ , as defined above, is positive, so that, informally, we will be able to write the PDE at least on that interval. This fact was actually taken into account in the definition of a solution in [43], as well as in [44], but, as we shall now see, this turns out to be an unnecessary assumption.

Lemma 3.6. Let (c_t) be a solution to Smoluchowski's equation (2.3) with initial conditions (c_0) such that $\langle c_0, a^2 + b^2 \rangle < +\infty$. Then $\langle c_t, a^2 + b^2 \rangle$ is bounded in a neighborhood of 0.

Proof. Take $f^{a,M}(a, b) = (a \wedge M)^2 \mathbb{1}_{\{b \leq M\}}$ and $f^{b,M}(a, b) = (b \wedge M)^2 \mathbb{1}_{\{a \leq M\}}$ for some $M \geq 1$. Then it is easy to see that, for $p = (a, b, m)$ and $p' = (a', b', m')$,

$$f^{a,M}(p \circ p') - f^{a,M}(p) - f^{a,M}(p') \leq 2aa' \mathbb{1}_{\{a, b \leq M\}} \mathbb{1}_{\{a', b' \leq M\}}.$$

From (2.8) and Cauchy-Schwarz inequality, we then get

$$\begin{aligned} \frac{d}{dt} \langle c_t, f^{a,M} \rangle &\leq \frac{1}{2} \sum_{(a,b,m), (a',b',m') \in \mathcal{S}} 2aa'(ab' + a'b)c_t(a, b, m)c_t(a', b', m') \mathbb{1}_{\{a, b \leq M\}} \mathbb{1}_{\{a', b' \leq M\}} \\ &\leq 2 \left(\langle c_t, f^{a,M} \rangle \right)^{3/2} \left(\langle c_t, f^{b,M} \rangle \right)^{1/2}. \end{aligned}$$

Taking now $f = f^{b,M}$ in (2.8) also shows that

$$\frac{d}{dt} \langle c_t, f^{b,M} \rangle \leq 2 \left(\langle c_t, f^{a,M} \rangle \right)^{1/2} \left(\langle c_t, f^{b,M} \rangle \right)^{3/2}.$$

Recalling that these two quantities are nonnegative, we then get

$$\frac{d}{dt}(\langle c_t, f^{a,M} \rangle \langle c_t, f^{b,M} \rangle) \leq 2 \left(\langle c_t, f^{a,M} \rangle \langle c_t, f^{b,M} \rangle \right)^{3/2}$$

which readily shows that

$$\langle c_t, f^{a,M} \rangle \langle c_t, f^{b,M} \rangle \leq \left(\frac{1}{\sqrt{\langle c_0, f^{a,M} \rangle \langle c_0, f^{b,M} \rangle}} - 2t \right)^{-2}$$

as long as the RHS remains bounded. Having $M \rightarrow +\infty$ and using monotone convergence, we thus get that

$$\langle c_t, a^2 \rangle \langle c_t, b^2 \rangle \leq \left(\frac{1}{\sqrt{\langle c_0, a^2 \rangle \langle c_0, b^2 \rangle}} - 2t \right)^{-2}$$

as long as the RHS is bounded, what shows the result. \square

This gives an a priori lower bound on Γ_∞ , and thus, as we shall see, of the time of existence and uniqueness of a solution to (2.3). However, this bound is not interesting for us since we will be able to easily get a better one.

4 Proof of the theorem

4.1 Overview of the method

In this section, we give a sketch of the proof which contains all the important ideas. The rigorous proof however requires some care, and it is given in detail afterwards. So, consider a solution $(c_t)_{t \in [0, T]}$ to Smoluchowski's equation (2.3), and

$$g_t(x, y, z) = \langle c_t, x^a y^b z^m \rangle = \sum_{a \geq 0} \sum_{b \geq 0} \sum_{m \geq 1} c_t(a, b, m) x^a y^b z^m.$$

Using (2.8) and Lemma 3.5, it is easy to see that g_t solves the following PDE

$$\frac{\partial g_t}{\partial t} = \frac{\partial g_t}{\partial x} \frac{\partial g_t}{\partial y} - \frac{1}{1+t} \left(x \frac{\partial g_t}{\partial x} + y \frac{\partial g_t}{\partial y} \right). \quad (2.10)$$

Now, we can solve this PDE using the method of characteristics: we want to find a trajectory $(x(t), y(t))$ starting from some $(x, y) \in [0, 1]^2$ such that $g_t(x(t), y(t), z)$ is easy to compute. So let

$$(p_1(t), p_2(t)) = \left(\frac{\partial g_t}{\partial x}(x(t), y(t), z), \frac{\partial g_t}{\partial y}(x(t), y(t), z) \right).$$

An easy calculation shows that

$$\dot{p}_1(t) = \frac{\partial^2 g_t}{\partial x^2} \left(\dot{x}(t) + p_2(t) - \frac{x(t)}{1+t} \right) + \frac{\partial^2 g_t}{\partial x \partial y} \left(\dot{y}(t) + p_1(t) - \frac{y(t)}{1+t} \right) - \frac{p_1(t)}{1+t}, \quad (2.11)$$

and a similar formula for \dot{p}_2 . Now, if we require

$$\dot{x}(t) + p_2(t) - \frac{x(t)}{1+t} = \dot{y}(t) + p_1(t) - \frac{y(t)}{1+t} = 0,$$

then

$$\dot{p}_i(t) = -\frac{p_i(t)}{1+t}, \quad i = 1, 2.$$

These ODE's are readily solved, with $p_1(0) = \frac{\partial g_0}{\partial x}(x, y)$ and $p_2(0) = \frac{\partial g_0}{\partial y}(x, y)$, and we obtain

$$p_i(t) = \frac{p_i(0)}{1+t} \quad (2.12)$$

and

$$x(t) = x + (x - p_2(0))t \quad ; \quad y(t) = y + (y - p_1(0))t.$$

Using the PDE, we now see that

$$\frac{d}{dt}g_t(x(t), y(t), z) = -\frac{p_1(0)p_2(0)}{(1+t)^2}, \quad (2.13)$$

so by integrating

$$g_t(x(t), y(t), z) = g_t(\phi_t(x, y, z), z) = g_0(x, y, z) - \frac{t}{1+t} \frac{\partial g_0}{\partial x}(x, y, z) \frac{\partial g_0}{\partial y}(x, y, z). \quad (2.14)$$

To obtain g_t , it only remains to invert ϕ_t , for, if $\phi_t(h_t) = \text{Id}$, then

$$g_t(x, y, z) = g_0(h_t(x, y, z), z) - \frac{t}{1+t} \frac{\partial g_0}{\partial x}(h_t(x, y, z), z) \frac{\partial g_0}{\partial y}(h_t(x, y, z), z).$$

We may now start a rigorous proof, which consists mainly of 3 steps: study the map ϕ_t , then solve the PDE (2.17), and show that the generating function of a family (c_t) solves (2.17) if and only if (c_t) solves Smoluchowski's equation (2.3). The conclusion is then easy to obtain.

4.2 Inversion of the mapping

In this section, we study the map ϕ_t , which is useful both for solving theoretically the PDE, and for obtaining explicit solutions. We will need two preliminary lemmas.

Lemma 4.1. *Let $\alpha > 0$, $\beta, \gamma \geq 0$ and $K = [0, \alpha] \times [0, \beta] \times [0, \gamma]$. For $(r, s, t) \in K$, denote*

$$A(r, s, t) := \begin{pmatrix} r & s \\ t & r \end{pmatrix}.$$

Then for every $\varepsilon > 0$, there is a norm $\|\cdot\|$ on \mathbb{R}^2 such that

$$\max_{(r,s,t) \in K} \|A(r, s, t)\| \leq \alpha + \sqrt{\beta\gamma} + \varepsilon,$$

where we also denote by $\|\cdot\|$ the induced norm on the 2×2 matrices.

Remark 4.2. This is a uniform version of the well-known result (see e.g. [51]) which states that

- for every (square) matrix A and norm $\|\cdot\|$, one has $\|A\| \geq \rho(A)$, where $\rho(A)$ is the spectral radius of A ;
- for every matrix A and $\varepsilon > 0$, there is a norm $\|\cdot\|$ such that $\|A\| \leq \rho(A) + \varepsilon$.

Note indeed that $\alpha + \sqrt{\beta\gamma}$ is the spectral radius of $A(\alpha, \beta, \gamma)$.

Proof. 1. First assume that β and γ are positive. We can diagonalize $A := A(\alpha, \beta, \gamma)$.

If we let $a := \alpha$, $b := \sqrt{\beta}$ and $c := \sqrt{\gamma}$ then

$$A = P \begin{pmatrix} a + bc & 0 \\ 0 & a - bc \end{pmatrix} P^{-1},$$

where

$$P = \begin{pmatrix} b & -b \\ c & c \end{pmatrix}, \quad P^{-1} = \frac{1}{2bc} \begin{pmatrix} c & b \\ -c & b \end{pmatrix}.$$

Now, consider the following norm: for $x \in \mathbb{R}^2$, let $\|x\| = \|P^{-1}x\|_\infty$, where $\|\cdot\|_\infty$ is the sup norm: $\|(x_1, x_2)\|_\infty = \max(|x_1|, |x_2|)$. Then for any 2×2 matrix M ,

$$\|M\| = \max_{x \neq 0} \frac{\|Mx\|}{\|x\|} = \max_{x \neq 0} \frac{\|P^{-1}Mx\|_\infty}{\|P^{-1}x\|_\infty} = \max_{y \neq 0} \frac{\|P^{-1}MPy\|_\infty}{\|y\|_\infty} = \|P^{-1}MP\|_\infty.$$

An easy computation shows that for $(r, s, t) \in K$,

$$P^{-1}A(r, s, t)P = \begin{pmatrix} r + \frac{bt}{2c} + \frac{cs}{2b} & -\frac{bt}{2c} + \frac{cs}{2b} \\ \frac{bt}{2c} - \frac{cs}{2b} & r - \frac{bt}{2c} - \frac{cs}{2b} \end{pmatrix}.$$

Recall that for a matrix M ,

$$\|M\|_\infty = \max_i \sum_j |M_{i,j}|,$$

so that, since $r \geq 0$,

$$\|P^{-1}A(r, s, t)P\|_\infty = r + \frac{bt}{2c} + \frac{cs}{2b} + \left| \frac{bt}{2c} - \frac{cs}{2b} \right| := F(r, s, t).$$

It remains to find the maximum of F on K . First, note that for $(r, s, t) \in K$,

$$0 \leq F(r, s, t) \leq F(\alpha, s, t).$$

Then, for every $(s, t) \in [0, \beta] \times [0, \gamma]$, we can write $t = ps$, $p \geq 0$. If $p \leq c^2/b^2$, then $cs/(2b) \geq bt/(2c)$, so that $F(\alpha, s, t) = \alpha + cs/b$. But $s \leq b^2$, so $F(\alpha, s, t) \leq \alpha + bc = \alpha + \sqrt{\beta\gamma}$. And if $p > c^2/b^2$, then $cs/(2b) \leq bt/(2c)$, so that $F(\alpha, s, t) = \alpha + bt/c$. But $t \leq c^2$, so $F(\alpha, s, t) \leq \alpha + bc = \alpha + \sqrt{\beta\gamma}$. Finally, the maximum of F on K , i.e. the maximum of $\|A(r, s, t)\|$ on K , is $\alpha + \sqrt{\beta\gamma}$.

2. Assume now that β or γ is zero, say e.g. $\gamma = 0$. Take $\varepsilon > 0$, and $M > 0$ such that $\beta/M < \varepsilon$. Consider the norm $\|x\| = \|Px\|_\infty$, where P is a diagonal matrix with diagonal $(1, M)$. For $(r, s, 0) \in K$, we have as before

$$\|A(r, s, 0)\| = \|PA(r, s, 0)P^{-1}\|_\infty = \left\| \begin{pmatrix} r & s/M \\ 0 & r \end{pmatrix} \right\|_\infty.$$

Since $s \leq \beta$, this shows that $\|A(r, s, 0)\| \leq \alpha + \varepsilon$. □

We will deal often with real-analytic functions in the remaining of the proofs. For the definitions and results on this topic, we refer to [31]. We will show the following result.

Proposition 4.3. For $t \in [0, T_{\text{gel}})$ and $z \in [0, 1]$, define $\phi_t(\cdot, \cdot, z) : [0, 1]^2 \rightarrow \mathbb{R}^2$ by

$$\phi_t(x, y, z) = \left((1+t)x - t \frac{\partial g_0}{\partial y}(x, y, z), (1+t)y - t \frac{\partial g_0}{\partial x}(x, y, z) \right), \quad (2.15)$$

and let $K_t(z)$ be the closed subset of $[0, 1]^2$: $K_t(z) = \phi_t(\cdot, \cdot, z)^{-1}([0, 1]^2)$. Then

- (i) $\phi_t(\cdot, \cdot, z) : K_t(z) \rightarrow [0, 1]^2$ is a homeomorphism. Denote $h_t(\cdot, \cdot, z)$ its inverse and write $h_t(\cdot, \cdot, z) = (h_t^{(1)}(\cdot, \cdot, z), h_t^{(2)}(\cdot, \cdot, z))$.
- (ii) For $i = 1, 2$, $(x, y, z, t) \mapsto h_t^{(i)}(x, y, z)$ is an analytic function on $(0, 1)^3 \times (0, T_{\text{gel}})$.

Proof. (i) Fix some $z \in [0, 1]$ and some $t \in (0, T_{\text{gel}})$, and keep the notations of the statement. For notational simplicity, we omit the parameter z . Let $0 \leq t < T_{\text{gel}}$. We first want to show that $\phi_t : K_t \rightarrow [0, 1]^2$ is one-to-one and onto. Fix $(u, v) \in [0, 1]^2$ and let us check that there is a unique couple $(x, y) \in [0, 1]^2$ such that $\phi_t(x, y) = (u, v)$. This requirement is equivalent to finding a unique fixed point to

$$F_t(x, y) = \left(\frac{u}{1+t} + \frac{t}{1+t} \frac{\partial g_0}{\partial y}(x, y, z), \frac{v}{1+t} + \frac{t}{1+t} \frac{\partial g_0}{\partial x}(x, y, z) \right).$$

Because of the remark above, F_t is a mapping from $[0, 1]^2$ to $[0, 1]^2$. It remains to check that it is contracting. Its differential is

$$DF_t(x, y) = \frac{t}{1+t} \begin{pmatrix} \frac{\partial^2 g_0}{\partial x \partial y} & \frac{\partial^2 g_0}{\partial y^2} \\ \frac{\partial^2 g_0}{\partial x^2} & \frac{\partial^2 g_0}{\partial x \partial y} \end{pmatrix} := \frac{t}{1+t} \begin{pmatrix} \alpha(x, y, z) & \beta(x, y, z) \\ \gamma(x, y, z) & \alpha(x, y, z) \end{pmatrix}. \quad (2.16)$$

Let $\alpha = \alpha(1, 1, 1) = \langle c_0, ab \rangle$, $\beta = \beta(1, 1, 1) = \langle c_0, b(b-1) \rangle$ and $\gamma = \gamma(1, 1, 1) = \langle c_0, a(a-1) \rangle$. Since $t < T_{\text{gel}}$, then $\frac{t}{1+t}(\alpha + \sqrt{\beta\gamma} + \varepsilon) < 1$ for some small enough $\varepsilon > 0$. Hence, by Lemma 4.1, there is a norm $\|\cdot\|$ such that

$$\max_{(x,y) \in [0,1]^2} \|DF_t(x, y)\| \leq \frac{t}{1+t}(\alpha + \sqrt{\beta\gamma} + \varepsilon) < 1,$$

so that F_t is contracting. Hence it has a unique fixed point. As a consequence, there is a unique couple $(x, y) \in [0, 1]^2$ such that $\phi_t(x, y) = (u, v)$. Moreover, since F_t is continuous with respect to (u, v) and uniformly contracting in (u, v) , then the mapping $(u, v) \mapsto (x, y)$ is continuous, that is $h_t : [0, 1]^2 \rightarrow K_t$ is a homeomorphism.

- (ii) For $t_0 \in (0, T_{\text{gel}})$, $z_0 \in (0, 1)$ and $(x_0, y_0) \in U_{t_0}$, the matrix $D\phi_{t_0}(x_0, y_0, z_0)$ is invertible. Then Theorem 2.5.3 in [31] shows that the inverse mapping of ϕ_t has real-analytic coefficients, i.e. $h_t^{(i)}$ are real-analytic functions on $(0, 1)^3 \times (0, T_{\text{gel}})$. \square

4.3 Study of the PDE

The following (non-quasilinear) PDE is a central feature of our discussion

$$\frac{\partial g_t}{\partial t} = \frac{\partial g_t}{\partial x} \frac{\partial g_t}{\partial y} - \frac{1}{1+t} \left(x \frac{\partial g_t}{\partial x} + y \frac{\partial g_t}{\partial y} \right). \quad (2.17)$$

A preliminary result to the proof is the following. Its proof is exactly the same as the one of Lemma 3.1.

Lemma 4.4. *Let $(c_t)_{t \in [0, T]}$ be a solution to the system*

$$\frac{d}{dt}c_t(p) = \frac{1}{2} \sum_{p' \preceq p} p' \cdot (p \setminus p') c_t(p') c_t(p \setminus p') - \frac{a+b}{1+t} c_t(p) \quad (2.18)$$

for $p = (a, b, m) \in S$, with nonnegative initial conditions. Then for all $t \in [0, T]$ and $p \in S$, $c_t(p) \geq 0$.

Recall also that the regularity of a mapping is to be understood in the sense of Definition 3.4.

Proposition 4.5. (i) *For every $z \in [0, 1]$, the PDE (2.17) with initial conditions $g_0 = g_0(\cdot, \cdot, z)$ has a unique regular solution $(t, x, y) \mapsto g_t(x, y, z)$ defined on $[0, T_{\text{gel}}) \times (0, 1)^2$.*

(ii) *The solution of the PDE is given by*

$$g_t(x, y, z) = g_0(h_t(x, y, z), z) - \frac{t}{1+t} \frac{\partial g_0}{\partial x}(h_t(x, y, z), z) \frac{\partial g_0}{\partial y}(h_t(x, y, z), z), \quad (2.19)$$

where h_t is defined in Proposition 4.3.

(iii) *We have the alternative expression*

$$g_t(x, y, z) = \frac{1}{1+t} \left(\tilde{H}_t^{(2)}(x, y, z) + \tilde{H}_t^{(1)}(0, y, z) \right) + G_t(z) \quad (2.20)$$

in the notations of Theorem 2.5.

(iv) *For every $t \in [0, T_{\text{gel}})$, g_t has an analytic expansion*

$$g_t(x, y, z) = \sum_{(a, b, m) \in S} c_t(a, b, m) x^a y^b z^m \quad (2.21)$$

for $(x, y, z) \in [0, 1]^3$, where $c_t(a, b, m) \geq 0$.

Remark 4.6. Formula (2.20) will be useful to compute explicit solutions, since with it, it is enough to have the analytic expansion of h_t around 0 to obtain the one of g_t (whose coefficients are precisely the solution to (2.3)). Note however that G_t may be tedious to compute in general, but since it is a function of z only, it is relevant only when we wish to compute the concentrations of clusters with no arms. Rather than computing G_t , a better way to obtain those concentrations is to use Equation (2.3), since

$$\frac{d}{dt}c_t(0, 0, m) = \sum_{m'=1}^{m-1} c_t(1, 0, m') c_t(0, 1, m - m'). \quad (2.22)$$

Proof. We will prove the statement in four steps. First we will show that a solution has to be written as in (2.19). Next that this formula does provide a solution. Proving formula (2.20) is then an easy matter. In all the proof, some $z \in [0, 1]$ is fixed.

1. Let $U_t = \phi_t(\cdot, \cdot, z)^{-1}((0, 1)^2)$, and consider g_t a regular solution of (2.17) on $[0, T) \times (0, 1)^2$. Fix $t_0 \in (0, T)$ and $(x, y) \in U_{t_0}$, and let

$$(p_1(t), p_2(t)) := \left(\frac{\partial g_t}{\partial x}(\phi_t(x, y, z), z), \frac{\partial g_t}{\partial y}(\phi_t(x, y, z), z) \right).$$

It is easy to see that U_t decreases with t , so for $t \leq t_0$, this definition makes sense and we can differentiate p_i . The regularity assumptions on g_t are just those needed to allow the use of Schwarz's theorem, and an easy computation shows that, on $[0, t_0]$, (p_1, p_2) solves a linear differential system with continuous coefficients, whose solution is given by (2.12). Hence

$$\frac{\partial g_t}{\partial x}(\phi_t(x, y, z), z) = \frac{\partial g_0}{\partial x}(x, y, z)/(1+t), \quad \frac{\partial g_t}{\partial y}(\phi_t(x, y, z), z) = \frac{\partial g_0}{\partial y}(x, y, z)/(1+t) \quad (2.23)$$

for all $(x, y) \in U_t$. Then, it is easy to check that for all $(x, y) \in U_t$, (2.13) and (2.14) hold. Replacing (x, y) by $h_t(x, y, z)$ (recall $h_t : (0, 1)^2 \rightarrow U_t$ is the right-inverse of ϕ_t), we finally obtain (2.19). This shows that the PDE has at most one solution.

2. The existence of a solution is now straightforward. Let g_t be defined as in (2.19). Because of the regularity of h_t and of g_0 , g has the required regularity properties. It then suffices to show that it is actually a solution. To this end, let us first compute

$$(p_1(t), p_2(t)) := \left(\frac{\partial g_t}{\partial x}(\phi_t), \frac{\partial g_t}{\partial y}(\phi_t) \right)$$

for some fixed $t \in [0, T_{\text{gel}})$ and $(x, y) \in U_t$. By differentiating $g_t(\phi_t)$ with respect to x and y , it is easy to see that it solves a linear system, which has, before T_{gel} , a unique solution, given by equation (2.23). To conclude, we may differentiate $g_t(\phi_t)$ in two different ways: one using (2.19) and (2.23). The other with the chain rule. Compounding by h_t in the obtained equality readily shows that g_t solves the PDE (2.17) for $t \in [0, T_{\text{gel}})$, $(x, y) \in (0, 1)^2$.

3. The formula (2.20) is easy to obtain, by differentiating $g_0(h_t(x, y, z), z)$ with respect to x , y and z , and using the fact that

$$\frac{\partial g_0}{\partial x}(h_t, z) = \tilde{h}_t^{(2)}, \quad \frac{\partial g_0}{\partial y}(h_t, z) = \tilde{h}_t^{(1)}.$$

in the notations of Theorem 2.5.

4. To prove the last point, consider $t_0 \in [0, T_{\text{gel}})$. ϕ_t is well-defined and analytic (in (t, x, y, z)) in a neighborhood of $(t_0, 0, 0, 0)$, and $D\phi_{t_0}(0, 0, 0)$ is invertible. So, by theorem 2.5. in [31], h_t is analytic near $(t_0, 0, 0, 0)$, hence so is $g_t = g_0(h_t)$. So we may write

$$g_t(x) = \sum_{(a,b,m) \in S} c_t(a, b, m) x^a y^b z^m \quad (2.24)$$

for (t, x, y, z) in a neighborhood of $(t_0, 0, 0, 0)$ and analytic c_t . By analytic continuation, the c_t are uniquely defined, so we can let

$$E = \{t \in [0, T_{\text{gel}}), \forall p \in S \ c_t(p) \geq 0\}.$$

By continuity, E is a closed set containing 0. On the other hand, (2.24) holds for (t, x, y, z) in a neighbourhood of $(t_0, 0, 0, 0)$, so for $t_0 \in E$, there is a $\varepsilon > 0$ such that (2.24) holds for $t \in (t_0 - \varepsilon, t_0 + \varepsilon)$ and $(x, y, z) \in (-\varepsilon, \varepsilon)^3$. In particular, since g_t solves the PDE (2.17), it is easy to see, using a Cauchy product and identifying the coefficients, that c_t solves (2.18) for $t \in (t_0 - \varepsilon, t_0 + \varepsilon)$. So by Lemma (4.4), $(t_0 - \varepsilon, t_0 + \varepsilon) \subset E$. So E is open and $E = [0, T_{\text{gel}})$. Finally, recall from Proposition 4.3 that h_t , and so g_t , are analytic on $[0, 1]^3$. But we have just shown that g_t has an analytic expansion around 0 with nonnegative coefficients. So (see e.g. the proof of Bernstein's theorem in [31]), this expression actually holds on $[0, 1]^3$.

□

4.4 Equivalence between the system and the PDE

Smoluchowski's equation (2.3) is solved thanks to PDE (2.17).

Proposition 4.7. (i) *Let $(c_t)_{t \in [0, T]}$ be a solution to Smoluchowski's equation (2.3), and let g_t be its generating function, defined by $g_t(x, y, z) := \langle c_t, x^a y^b z^m \rangle$. Then for all $z \in [0, 1]$, $(t, x, y) \mapsto g_t(x, y, z)$ is a regular solution to the PDE (2.17) on $[0, T \wedge \Gamma_\infty) \times (0, 1)^2$, with initial conditions $g_0(\cdot, \cdot, z)$.*

(ii) *Conversely, let $(c_t(p))_{p \in S}$, $t \in [0, T)$, be a family of differentiable functions. Let $g_t(x, y, z)$ be its generating function and assume it is defined for $t \in [0, T)$, $(x, y) \in (0, 1)^2$ and $z \in [0, 1]$. Assume that for every $z \in [0, 1]$, $g_t(\cdot, \cdot, z)$ is a regular solution to the PDE (2.17) with initial conditions $g_0(\cdot, \cdot, z)$. Then*

- for all $p \in S$ and $t \in [0, T)$, $c_t(p) \geq 0$;
- (c_t) is a solution to Smoluchowski's equation (2.3) for $t \in [T \wedge T_{\text{gel}})$, with initial conditions c_0 .

Remark 4.8. An important feature of this result is that the PDE (2.17) and the system (2.3) are equivalent only before the critical time (T_{gel} or Γ_∞). This fact is crucial when we study the microscopic model. We indeed obtain a family of coefficients whose generating function solves the PDE (on \mathbb{R}^+), but we cannot ensure that they solve Smoluchowski's equation (2.3) after T_{gel} . Actually, there is every chance that they do not, as we already discussed in Section 2.4 of the introduction of this thesis.

Proof of Proposition 4.7. (i) First note that g is regular according to Remark 3.3. If one takes $f(a, b, m) = x^a y^b z^m$ in (2.8), for some fixed $(x, y, z) \in (0, 1)^2 \times [0, 1]$, then one gets

$$\frac{d}{dt} g_t(x, y, z) = \frac{\partial g_t}{\partial x} \frac{\partial g_t}{\partial y} - A_t y \frac{\partial g_t}{\partial y} - B_t x \frac{\partial g_t}{\partial x}.$$

Recall from Lemma 3.5 that when $t < \Gamma_\infty$, $A_t = B_t = 1/(1+t)$. Replacing in the equation above shows that g_t solves (2.17) for $(x, y) \in (0, 1)^2$ and $0 \leq t < T \wedge \Gamma_\infty$.

(ii) As in the fourth part of the proof of Proposition 4.5, we see that the $(c_t(p))$ solve (2.18), and hence that they are nonnegative. By uniqueness of a solution to the PDE (2.17), for $t \in [0, T \wedge T_{\text{gel}})$, these coefficients are those obtained in (2.21). Now, let $t < T_{\text{gel}} \wedge T$, $U_t = \phi_t^{-1}(\cdot, \cdot, 1)((0, 1)^2)$, $K_t = K_t(1) = \phi_t^{-1}(\cdot, \cdot, 1)([0, 1]^2)$, and recall from (2.23) that since g is a regular solution to (2.17), then for all $(x, y) \in U_t$

$$\frac{\partial g_t}{\partial x}(\phi_t(x, y, 1), 1) = \frac{\partial g_0}{\partial x}(x, y, 1) \frac{1}{1+t},$$

what we can write

$$\sum_{(a, b, m) \in S} a c_t(a, b, m) \phi_t^{(1)}(x, y, 1)^a \phi_t^{(2)}(x, y, 1)^b = \sum_{(a, b, m) \in S} a c_0(a, b, m) x^a y^b \times \frac{1}{1+t}. \quad (2.25)$$

Note now that since $t < T_{\text{gel}}$, then $\phi_t(\cdot, \cdot, 1) : K_t \rightarrow [0, 1]^2$ is a homeomorphism, so $\bar{U}_t = K_t$. Since $(1, 1) \in K_t$, we can pass to the limit in the equality above when $(x, y) \rightarrow (1, 1)$. Using monotone convergence and the continuity of ϕ_t , we obtain

$$\langle c_t, a \rangle = \langle c_0, a \rangle \times \frac{1}{1+t} = \frac{1}{1+t}.$$

The same reasoning shows that $\langle c_t, b \rangle = 1/(1+t)$ for $t < T_{\text{gel}}$. Hence, we may re-write (2.18) before T_{gel} by substituting

$$\frac{a}{1+t} = a\langle c_t, b \rangle, \quad \frac{b}{1+t} = b\langle c_t, a \rangle,$$

which shows that (c_t) solves Smoluchowski's equation (2.3) before T_{gel} . \square

4.5 Existence and uniqueness of a solution

With these results, proving Theorem 2.5 is now an easy matter.

Proof of theorem 2.5. 1. Let us first prove that $\langle c_t, a^2 + b^2 \rangle$ is finite before T_{gel} and tends to $+\infty$ when $t \rightarrow T_{\text{gel}}$. So take $(c_t(a, b, m))_{t \in [0, T]}$ a solution to the system (2.3), and g_t its generating function. Since $\langle c_t, a^2 + b^2 \rangle < +\infty$ in a neighborhood of 0 by Lemma 3.6, we have

$$\frac{\partial^2 g_t}{\partial x^2}(1, 1, 1) = \langle c_t, a^2 - a \rangle,$$

as long as $\langle c_t, a^2 + b^2 \rangle < +\infty$. Note that by Lemma 3.5 $\langle c_t, a \rangle$ is bounded by 1, so $\langle c_t, a^2 \rangle$ explodes if and only if $\frac{\partial^2 g_t}{\partial x^2}(1, 1, 1)$ explodes. Let us compute the latter. Differentiating (2.23) with respect to x and y and having (x, y) tend to $(1, 1)$, we obtain that

$$\begin{pmatrix} 1+t-t\alpha & -t\gamma \\ -t\beta & 1+t-t\alpha \end{pmatrix} \begin{pmatrix} \delta \\ \eta \end{pmatrix} = \frac{1}{1+t} \begin{pmatrix} \gamma \\ \alpha \end{pmatrix},$$

where

$$\alpha = \frac{\partial^2 g_0}{\partial x \partial y} = \langle c_0, ab \rangle, \quad \beta = \frac{\partial^2 g_0}{\partial y^2} = \langle c_0, b^2 - b \rangle, \quad \gamma = \frac{\partial^2 g_0}{\partial x^2} = \langle c_0, a^2 - a \rangle.$$

and

$$\delta = \frac{\partial^2 g_t}{\partial x^2}(1, 1, 1), \quad \eta = \frac{\partial^2 g_t}{\partial x \partial y}(1, 1, 1).$$

Hence

$$\delta = \frac{\partial^2 g_t}{\partial x^2}(1, 1, 1) = \langle c_t, a(a-1) \rangle = \frac{\gamma}{(1+t-t\alpha)^2 - t^2\gamma\beta}.$$

This expression is valid as long as $t < T_{\text{gel}}$, since the determinant of the matrix is then nonzero. In the same way, we also have

$$\eta = \frac{\partial^2 g_t}{\partial y^2}(1, 1, 1) = \langle c_t, b(b-1) \rangle = \frac{\beta}{(1+t-t\alpha)^2 - t^2\gamma\beta}.$$

If γ or β is nonzero, then $\langle c_t, a^2 + b^2 \rangle \rightarrow +\infty$ when $t \rightarrow T_{\text{gel}}$. If $\gamma = \beta = 0$, then $\langle c_t, a^2 + b^2 \rangle$ remains finite, but this condition also imposes that $M = 1$, and so $T_{\text{gel}} = +\infty$.

2. Uniqueness is now easy to obtain: assume $(c_t^{(1)})$ and $(c_t^{(2)})$ solve the system 2.3 on $[0, T)$, $T \leq T_{\text{gel}}$, with initial conditions c_0 . Let $g_t^{(1)}$ and $g_t^{(2)}$ be their generating functions. Since $\Gamma_\infty = T_{\text{gel}}$ and $T \leq T_{\text{gel}}$, then by Proposition 4.7, for every $z \in [0, 1]$, they are regular solutions to the PDE (2.17) on $[0, T) \times (0, 1)^2$, with initial conditions $g_0(\cdot, \cdot, z)$. But by Proposition 4.5 there is a unique regular solution to the PDE on $[0, T_{\text{gel}})$, so $g_t^{(1)} = g_t^{(2)}$ on $[0, T)$, so that $(c_t^{(1)}) = (c_t^{(2)})$.

3. The existence is given by point (iv) of Proposition 4.5, and point (ii) of Proposition 4.7.
4. Let us finally prove that the total mass is conserved. Let $\psi_t(x, y, z) = (\phi_t(x, y, z), z)$ and consider $U'_t = \psi_t^{-1}((0, 1)^3)$ and $K'_t = \psi_t^{-1}([0, 1]^3)$. For $(x, y, z) \in U'_t$, we can differentiate $g_t(\psi_t(x, y, z))$ with respect to z , and using (2.23), we obtain

$$\frac{\partial g_t}{\partial z}(\psi_t(x, y, z)) = \frac{\partial g_0}{\partial z}(\psi_t(x, y, z)).$$

Now, ψ_t is a homeomorphism from K'_t to $[0, 1]^3$, so $\overline{U'_t} = K'_t$. But $(1, 1, 1) \in K'_t$, so we may pass to the limit when $(x, y, z) \rightarrow (1, 1, 1)$ in the equality above, to obtain

$$\frac{\partial g_t}{\partial z}(1, 1, 1) = \frac{\partial g_0}{\partial z}(1, 1, 1),$$

what precisely means $\langle c_t, m \rangle = \langle c_0, m \rangle$. □

5 Explicit formulas

We give in this section some explicit solutions to Equation (2.3). We will consider only monodisperse initial conditions, i.e. assume that at time 0, there are only particles of size 1 in the medium. So given a (finite) measure μ on $\mathbb{N} \times \mathbb{N}$, we assume

$$c_0(a, b, m) = \mu(a, b) \mathbb{1}_{\{m=1\}}$$

and as usual $\langle c_0, a \rangle = \langle c_0, b \rangle = 1$. To obtain the solutions, we need to invert ϕ_t , what can be done using the (two-variable) Lagrange inversion formula (a statement is given by Good [21]). But it is much more involved than the one-dimensional formula, and the expressions it would provide can hardly be called explicit. Let us however study three easy cases. Only the last one requires the two-variable formula.

5.1 Particles with one female arm

The first case is when each particle has exactly one female arm, and a number of male arms distributed according to a measure μ_1 . So take

$$\mu(a, b) = \begin{cases} 0 & \text{if } b \neq 1 \\ \mu_1(a) & \text{if } b = 1 \end{cases}$$

and we will assume that $A_0 = B_0 = 1$, i.e. μ_1 is a probability measure with unit mean. In this case, we obtain, for every $a, b \geq 0$ and $m \geq 1$,

$$c_t(a, b, m) = \begin{cases} 0 & \text{if } b \neq 1 \\ \frac{t^{m-1}}{(1+t)^{m+a}} \frac{1}{m} \binom{m+a-1}{a} \mu_1^{*m}(m+a-1) & \text{if } b = 1. \end{cases}$$

In particular there exists only clusters with one female arm, what is physically obvious. Moreover, the concentration $c_t(a, 1, m)$ is exactly the concentration of clusters with a arms and mass m obtained in the ‘‘oriented model’’ (2.2) of [3], with initial distribution μ_1 . This is also natural, since in this case, $(a, 1, m)$ - and $(a', 1, m')$ -clusters coagulate at rate $a + a'$, which is the rate of the oriented model. Note also that $T_{\text{gel}} = +\infty$, as in the oriented model.

5.2 Arms with uniform random genders

In this model, the total number of arms of a particle is chosen according to a measure μ_1 , then each arm is given a gender independently, with probability $1/2$. That is, we let

$$\mu(a, b) = \mu_1(a + b) \binom{a + b}{b} \frac{1}{2^{a+b}}.$$

We will assume that μ_1 has mean 2, so that $A_0 = B_0 = 1$. Let $\nu_1(j) = (j + 1)\mu(j + 1)$. Then we obtain, for $(a, b) \neq (0, 0)$,

$$c_t(a, b, m) = \frac{1}{2} \frac{t^{m-1}}{(1+t)^{a+b+m-1}} \frac{(m+a+b-2)!}{m!a!b!} \left(\frac{\nu_1}{2}\right)^{*m} (m+a+b-2)$$

and

$$c_t(0, 0, m) = \frac{1}{2} \frac{1}{m(m-1)} \frac{1}{(1+1/t)^{m-1}} \left(\frac{\nu_1}{2}\right)^{*m} (m-2)$$

provided $\nu_1(0) > 0$. If $T_{\text{gel}} = +\infty$, this condition means that $\nu_1 \neq \delta_1$. In particular, one easily checks that

$$\sum_{a+b=k} c_t(a, b, m) = 2c_t^{\text{sym}}(k, m)$$

where $c_t^{\text{sym}}(k, m)$ is the concentration of clusters with k arms and mass m in the symmetric model (2.2) of [3], with initial arm distribution $\mu_1/2$. The factor is irrelevant since it 2 comes from the normalization: in our model, the total concentration of arms in the medium is 2, when it is 1 in the symmetric model.

It is also worth stressing the stronger fact that for $a, b \geq 0$, we have

$$c_t(a, b, m) = \frac{1}{2} \binom{a+b}{b} \frac{1}{2^{a+b}} c_t(a+b, m).$$

Hence, at any given time, the distribution of the number of male (or female) arms is still binomial. So, if at some time we chose to reassign to each arm a gender uniformly and independently, and let the system evolve on from this state, no difference would be observed. Or we could forget about the genders at some time t and observe the state at time t of Bertoin's model (2.2). Or, finally, we could watch a system evolve like the symmetric model (2.2) starting from an arm distribution $\mu_1/2$, and then at some time give the arms a gender uniformly at random and independently. The evolution afterwards will be the evolution of the sexed model with initial arm distribution μ . Note as before that the critical time is the same than in the symmetric model with initial distribution $\mu_1/2$.

More generally, consider initial concentrations such that for all $a, b \geq 0$ $\mu(a, b) = \mu(b, a)$, and the solution $(c_t)_{t \in [0, T_{\text{gel}}]}$ to Smoluchowski's equation (2.3). Then it is easy to check (by uniqueness) that, for all $t \in [0, T_{\text{gel}})$, $c_t(a, b, m) = c_t(b, a, m)$, and that, if we denote

$$k_t(l, m) := \sum_{a+b=l} c_t(a, b, m),$$

then (k_t) is governed (up to a factor $1/2$) by the symmetric Smoluchowski equation of [3]. Hence, in this case too, $k_t(l, m) = 2c_t^{\text{sym}}(l, m)$.

5.3 Particles with one gender

Let us finally consider the more intricate case where at time 0, the arms of each particle have all the same gender. This is motivated by the idea of ionic bonds: a particle with only male (resp. female) arms can be considered as a cation (resp. an anion), and cations can only bond with anions. Hence, consider, for $i = 1, 2$, μ_i two measures with mean 1 such that $\mu_1(0) = \mu_2(0)$, and take

$$\mu(a, b) = \begin{cases} \mu_1(a) & \text{if } b = 0 \\ \mu_2(b) & \text{if } a = 0 \\ 0 & \text{else} \end{cases}$$

and $\nu_i(j) = (j+1)\mu_i(j+1)$. The two-variable Lagrange inversion formula gives, for $(a, b) \neq (0, 0)$,

$$c_t(a, b, m) = \frac{t^{m-1}}{(1+t)^{m+a+b-1}} \sum_{k=0}^m \frac{(m-k+b-1)!(k+a-1)!}{(m-k)!k!a!b!} \nu_1^{*(m-k)}(k+a-1) \nu_2^{*k}(m-k-1+b).$$

If we also let

$$\nu_1 \diamond \nu_2(m) = (m-1) \sum_{k=1}^{m-1} \frac{1}{k} \nu_1^{*(m-k)}(k-1) \frac{1}{m-k} \nu_2^{*k}(m-k-1)$$

then, for $m \geq 2$,

$$c_t(0, 0, m) = \frac{1}{m-1} \frac{1}{(1+1/t)^{m-1}} \nu_1 \diamond \nu_2(m),$$

provided $\nu_1(0)\nu_2(0) > 0$ (which means, if $T_{\text{gel}} = +\infty$, that ν_1 and ν_2 are not δ_1). In particular, we see that if $T_{\text{gel}} = +\infty$ and $\nu_1, \nu_2 \neq \delta_1$, then for $m \geq 2$,

$$c_t(a, b, m) \rightarrow \begin{cases} 0 & \text{if } (a, b) \neq (0, 0) \\ \frac{1}{m-1} \nu_1 \diamond \nu_2(m) & \text{if } a = b = 0. \end{cases}$$

Hence all the arms are used to coagulate. Chemically, this means that there are no more ions in the medium. The limiting distribution of the sizes is given by $(m-1)^{-1} \nu_1 \diamond \nu_2(m)$. We will generalize this fact in the following section, and give a probabilistic interpretation of the measure $\nu_1 \diamond \nu_2$. Also, if M_i is the mean of ν_i , then, in the notations of Definition 2.3, $M = \sqrt{M_1 M_2}$ and $T_{\text{gel}} = 1/(M-1)$, or $+\infty$ if $M \leq 1$. If $\mu_1 = \mu_2 = \mu$, then the critical time is the same as in the symmetric model with initial distribution μ .

6 Limiting concentrations and Galton-Watson processes

6.1 Convergence of the concentrations

In this section, we will study the limiting concentrations. Similarly to what happens in the oriented and symmetric model of [3], we expect the concentrations to converge when the time tends to $+\infty$, whenever gelation does not occur. Physically, this would mean that the system converges to a terminal state where all arms have been used (otherwise, further coagulations “should” occur). This is actually true, and this is an easy consequence of the preceding results.

Corollary 6.1. *Assume $T_{\text{gel}} = +\infty$, and let $(c_t)_{t \geq 0}$ be the solution to Smoluchowski's equation (2.3).*

(i) When $t \rightarrow +\infty$, there exists limiting concentrations $c_\infty(m)$ such that

$$c_t(a, b, m) \rightarrow c_\infty(m) \mathbb{1}_{\{a=b=0\}}$$

in $\ell^1(S)$.

(ii) For $z \in [0, 1)$, the generating function $g_\infty(z)$ of $(c_\infty(m))_{m \geq 1}$ is the antiderivative vanishing at 0 of

$$\frac{\partial g_0}{\partial z} (h_\infty^{(1)}(z), h_\infty^{(2)}(z), z), \quad (2.26)$$

where $(h_\infty^{(1)}, h_\infty^{(2)})$ is characterized by

$$\begin{cases} h_\infty^{(1)}(z) &= \frac{\partial g_0}{\partial y} (h_\infty^{(1)}(z), h_\infty^{(2)}(z), z) \\ h_\infty^{(2)}(z) &= \frac{\partial g_0}{\partial x} (h_\infty^{(1)}(z), h_\infty^{(2)}(z), z). \end{cases} \quad (2.27)$$

Proof. (i) Since $\Gamma_\infty = T_{\text{gel}} = +\infty$, then (2.9) holds for all $t \geq 0$, so

$$\sum_{(a,b) \neq (0,0)} c_t(a, b, m) \xrightarrow{t \rightarrow +\infty} 0.$$

Then, using (2.22), we get, for all $t \geq 0$,

$$c_t(0, 0, m) = c_0(0, 0, m) + \sum_{m'=1}^{m-1} \int_0^t c_s(1, 0, m') c_s(0, 1, m - m') ds.$$

But the integrand is bounded by $A_s B_s = 1/(1+s)^2$. Hence the integral has a finite limit when $t \rightarrow +\infty$, and so does $c_t(0, 0, m)$. Finally, $\langle c_t, m \rangle$ is bounded by Theorem 2.5, and $\langle c_\infty, m \rangle < +\infty$ by Fatou's lemma, so Cauchy-Schwarz inequality shows that

$$\sum_{m \geq 1} |c_t(0, 0, m) - c_\infty(m)| \xrightarrow{t \rightarrow +\infty} 0$$

and the result follows.

(ii) By ℓ^1 -convergence, we have

$$g_\infty(z) = \lim_{t \rightarrow +\infty} g_t(0, 0, z),$$

so, using (2.5) and the fact that $\tilde{H}_t^{(1)}$ and $\tilde{H}_t^{(2)}$ are bounded by 1,

$$g_\infty(z) = \lim_{t \rightarrow +\infty} G_t(z).$$

It just remains to check that $h_t^{(1)}(0, 0, z)$ and $h_t^{(2)}(0, 0, z)$ do have a limit when $t \rightarrow +\infty$. From their definition (2.6), they have the same limit (if any) as $k_t^{(1)}(z) := \tilde{h}_t^{(1)}(0, 0, z)$ and $k_t^{(2)}(z) := \tilde{h}_t^{(2)}(0, 0, z)$. Now fix $Z \in [0, 1)$, and consider $k_t^{(1)}$ and $k_t^{(2)}$ as (continuous) maps on $[0, Z]$. But h_t is the right-inverse of ϕ_t , so

$$\frac{\partial g_0}{\partial x} (k_t^{(1)}(z), k_t^{(2)}(z), z) = k_t^{(2)}(z), \quad \frac{\partial g_0}{\partial y} (k_t^{(1)}(z), k_t^{(2)}(z), z) = k_t^{(1)}(z) \quad (2.28)$$

and, since $\langle c_0, a^2 + b^2 \rangle < +\infty$, then $\frac{\partial g_0}{\partial x}(x, y, z)$ has a bounded differential on $[0, 1]^2 \times [0, Z]$. Hence $k_t^{(2)}$, and for the same reason $k_t^{(1)}$, are Lipschitz-continuous on $[0, Z]$, with a constant independent of t . Ascoli's theorem thus shows that the families $(k_t^{(1)})$ and $(k_t^{(2)})$, $t \geq 0$, lie in a compact set (for the uniform topology on $[0, Z]$). So the family $(k_t^{(1)}, k_t^{(2)})$ lies in a compact set, and passing to the limit in (2.28) shows that any of its limit points solves (2.27). But since $T_{\text{gel}} = +\infty$, the application

$$(x, y) \mapsto \left(\frac{\partial g_0}{\partial y}(x, y, z), \frac{\partial g_0}{\partial x}(x, y, z) \right)$$

is contracting for every $z \in [0, Z]$. So there is a unique solution to (2.27), and $(k_t^{(1)}, k_t^{(2)})$ converges to this solution. \square

6.2 Connection with two-type Galton-Watson processes

In [3], Bertoin shows that for monodisperse initial conditions and when gelation does not occur, the limiting concentrations can be described in terms of Galton-Watson processes. The same kind of analogy is observed in our case. Precisely, we start from initial conditions

$$c_0(a, b, m) = \mu(a, b) \mathbb{1}_{\{m=1\}}$$

for a measure μ on $\mathbb{N} \times \mathbb{N}$ with $\langle \mu, a \rangle = \langle \mu, b \rangle = 1$. We may then define the probability measures

- $\mu_i(a, b) = \mu(a, b) / \langle \mu, 1 \rangle$, with generating function $\phi_i(x, y) := g_0(x, y, 1) / g_0(1, 1, 1)$.
- $\mu_m(a, b) = (b + 1) \mu(a, b + 1)$, with generating function $\phi_m(x, y) := \frac{\partial g_0}{\partial y}(x, y, 1)$,
- $\mu_f(a, b) = (a + 1) \mu(a + 1, b)$, with generating function $\phi_f(x, y) := \frac{\partial g_0}{\partial x}(x, y, 1)$.

Now, consider a Galton-Watson tree with two genders, constructed as follows. We start from an ancestor, who gives birth to a number a of male children, and a number b of female children, where (a, b) is distributed according to μ_i . Then each child gives birth to a certain number of children, distributed according to μ_m for the males, and to μ_f for the females, and so on. Consider $T(\mu_i, \mu_m, \mu_f)$ the total population of such a Galton-Watson process and let, for $r \in [0, 1]$,

$$g_i(r) = \mathbb{E}(r^{T(\mu_i, \mu_m, \mu_f)}), \quad g_m(r) = \mathbb{E}(r^{T(\mu_m, \mu_m, \mu_f)}), \quad g_f(r) = \mathbb{E}(r^{T(\mu_f, \mu_m, \mu_f)}).$$

It is classical (see e.g. [24]) that they solve the following system:

$$\begin{cases} g_i(r) &= r \phi_i(g_m(r), g_f(r)) \\ g_m(r) &= r \phi_m(g_m(r), g_f(r)) \\ g_f(r) &= r \phi_f(g_m(r), g_f(r)). \end{cases} \quad (2.29)$$

In particular, Equation (2.27) shows that (g_m, g_f) and $(h_\infty^{(1)}, h_\infty^{(2)})$ solve the same equation, which has a unique solution by Corollary 6.1, so $h_\infty^{(1)} = g_m$, $h_\infty^{(2)} = g_f$. If we define

$[z^m]h(z)$ the coefficient of z^m in the expansion around 0 of an analytic function h , we may then compute

$$\begin{aligned} [z^m]g_\infty(z) &= \frac{1}{m}[z^{m-1}]g_0(h_\infty^{(1)}(z), h_\infty^{(2)}(z), 1) = \frac{1}{m}g_0(1, 1, 1)[z^{m-1}]\phi_i(g_m(z), g_f(z)) \\ &= \frac{1}{m}\langle \mu, 1 \rangle [z^m]g_i(z) = \frac{1}{m}\langle \mu, 1 \rangle \mathbb{P}(T(\mu_i, \mu_m, \mu_f) = m). \end{aligned}$$

Finally, let us call a measure μ on $\mathbb{N} \times \mathbb{N}$ degenerate if $\mu = \delta_{(1,1)}$ or $\mu = \frac{1}{2}(\delta_{(2,0)} + \delta_{(0,2)})$, or $\mu(a, b) = 0$ for $a \neq 1$, or $\mu(a, b) = 0$ for $b \neq 1$. We let the reader check (using e.g. Theorem 10.1 in [24]) that under the assumptions $T_{\text{gel}} = +\infty$, and ruling out the degenerate cases, $T(\mu_i, \mu_m, \mu_f)$ is finite a.s. We have thus proved the following.

Corollary 6.2. *The limiting concentrations verify for $m \geq 2$*

$$c_\infty(m) = \frac{1}{m}\langle \mu, 1 \rangle \mathbb{P}(T(\mu_i, \mu_m, \mu_f) = m).$$

Moreover, if μ is not degenerate, then $T(\mu_i, \mu_m, \mu_f) < +\infty$ a.s.

As is explained in Section 4.2.3 of the introduction, and similarly to Bertoin's case [3, 4], we may interpret this formula by saying that a typical cluster in solution is a Galton-Watson $\mathbb{G}\mathbb{W}_{\mu_i, \mu_m, \mu_f}$.

This also provides an interpretation for the law \diamond of Section 5.3. This shows indeed that $\nu_1 \diamond \nu_2$ is the law of the total population of a two-type Galton Watson process started from one male and one female ancestors, where the males give birth to females according to the law ν_2 , and the females give birth to males according to the law ν_1 . In particular, if $\nu_1 = \nu_2 = \nu$, then $\nu \diamond \nu$ is the distribution of the size of a Galton-Watson tree with reproduction law ν and starting from two ancestors and we get (what is not obvious from the formula for \diamond), that for $m \geq 2$

$$\nu \diamond \nu(m) = \frac{2}{m}\nu^{*m}(m-2).$$

6.3 Mass at infinity

This corollary answers another question about gelation. By Theorem 2.5, the total mass $\langle c_t, m \rangle$ is conserved as time passes, so gelation does not occur before T_{gel} . But, if $T_{\text{gel}} = +\infty$, it may occur at infinity: some mass may be lost then. For monodisperse initial conditions, Corollary 6.2 proves that this cannot happen, except in the degenerate cases. Whenever μ is not degenerated, Corollary 6.2 indeed shows that

$$\langle c_\infty, m \rangle = \langle \mu, 1 \rangle \mathbb{P}(T(\mu_i, \mu_m, \mu_f) < +\infty) = \langle \mu, 1 \rangle = \langle c_0, 1 \rangle = \langle c_0, m \rangle$$

In the degenerate cases, it is easy to get explicit expressions for the concentrations, and they show that the mass at infinity is 0.

7 Microscopic model

7.1 Notations and preliminary results

The goal of this section is to construct a sequence of random processes modeling the coagulation of clusters with male and female arms. We will start with n particles (and then let $n \rightarrow +\infty$). Let us first set some notations.

- Recall that $S = \mathbb{N} \times \mathbb{N} \times \mathbb{N}^*$.
- $\llbracket 0, n \rrbracket = \{0, \dots, n\}$.
- $M > 0$ is a fixed real number. The number of arms and the total mass are assumed to grow at most like Mn (see the definition of E_n).
- The sequence of the number of p -clusters is an element of

$$E_n = \left\{ \mathbf{N} \in \llbracket 0, n \rrbracket^S, \langle \mathbf{N}, a + b + m \rangle \leq Mn \right\}$$

which is a finite set.

- $\frac{1}{n}E_n$ is a subset of

$$E = \left\{ C \in [0, 1]^S, \langle C, a + b + m \rangle \leq M \right\}.$$

An element of E represents the sequence of concentrations of p -clusters. E is a metric space endowed with the ℓ^1 distance

$$d(C^{(1)}, C^{(2)}) = \sum_{p \in S} |C^{(1)}(p) - C^{(2)}(p)|.$$

- We will call C-convergence the compact convergence (i.e. uniform convergence on every compact set) for functions from \mathbb{R}^+ to E .
- $\mathbb{D}(\mathbb{R}^+, H)$ is the space of càdlàg functions from \mathbb{R}^+ to a metric space (H, d) , endowed with the Skorokhod distance. We will call S-convergence the convergence for Skorokhod's distance. For the basic facts about Skorokhod distance for functions with value in a (complete separable) metric space, see [16].

So as to use standard theorems, the following easy result will be useful.

Lemma 7.1. *(E, d) is a compact metric space. In particular, it is a Polish space.*

7.2 Model

Let us now introduce the model. Informally, we consider a finite number n of particles with integer mass, and assume that at time 0, the total mass of the system plus the total number of arms is less than Mn . Then, each pair formed of a p -cluster and of a p' -cluster may coagulate at rate $\frac{1}{2}p.p'$, independently of the other pairs, to form a $p \circ p'$ -cluster, that is, the time one has to wait to see them coagulate is exponential with parameter $\frac{1}{2}p.p'$.

In other words, assume that the system is in the state η at a given time, that is $\eta \in E_n$ and $\eta(p)$ is the number of p -clusters. There are $\eta(p)\eta(p')$ (or $\eta(p)(\eta(p) - 1)$ if $p = p'$) pairs formed of a p -cluster and of a p' -cluster. Let

$$\lambda_\eta(p, p') = \begin{cases} \frac{1}{2}p.p'\eta(p)\eta(p') & \text{if } p \neq p' \\ \frac{1}{2}p.p\eta(p)(\eta(p) - 1) & \text{if } p = p'. \end{cases}$$

We set independently on each couple (p, p') an exponential clock with parameter $\lambda_\eta(p, p')$ (an exponential random variable with parameter 0 is assumed to be a.s. infinite). There

is a.s. one and only clock which rings first. If it is the clock on the couple (p, p') , then the system jumps to the state $\eta + \Delta_{p,p'}$ where

$$\begin{cases} \Delta_{p,p'}(p) = \Delta_{p,p'}(p') = -1 & \text{if } p \neq p' \\ \Delta_{p,p'}(p) = -2 & \text{if } p = p' \end{cases}, \quad \Delta_{p,p'}(p \circ p') = +1$$

and $\Delta_{p,p'} = 0$ otherwise. Then restart the construction afresh from the new state. Note that only finitely many $\eta(p)$ are nonzero, so the first jump occurs after an exponential time with parameter

$$\lambda_\eta = \sum_{p,p' \in S} \lambda_\eta(p, p') < +\infty.$$

We will consider the Markov chain constructed according to this rule. That is, we fix the following, for every $n \geq 1$.

- An element $\mathbf{X}_0^{(n)}$ of E_n , which is the initial number of particles.
- A pure-jump Markov process $\mathbf{X}^{(n)}$ on E_n , defined on a probability space $(\Omega^n, \mathcal{A}^n, \mathbb{P}^n)$, starting from $\mathbf{X}_0^{(n)}$, and with generator

$$Gf(\eta) = \sum_{(p,p') \in S^2} (f(\eta + \Delta_{p,p'}) - f(\eta)) \lambda_\eta(p, p')$$

for every bounded function $f : E_n \rightarrow \mathbb{R}$ and $\eta \in E_n$. The construction of such a process is obvious since E_n is finite.

- The rescaled and time-changed process

$$C_t^{(n)} = \frac{1}{n} \mathbf{X}_{t/n}^{(n)}.$$

Note that $C^{(n)}$ is a pure-jump Markov process on $\frac{1}{n}E_n \subset E$, starting from $C_0^{(n)} = \mathbf{X}_0^{(n)}/n$, and with generator

$$G^{(n)}f(\eta) = \sum_{(p,p') \in S^2} \left(f\left(\eta + \frac{1}{n}\Delta_{p,p'}\right) - f(\eta) \right) \lambda_\eta^{(n)}(p, p')$$

where

$$\lambda_\eta^{(n)}(p, p') = \frac{1}{n} \lambda_{n\eta}(p, p').$$

We shall prove the following result.

Theorem 7.2. *Assume that for every $p \in S$, $C_0^{(n)}(p) \rightarrow c_0(p)$ for some $c_0(p) \geq 0$, that*

$$\langle C_0^{(n)}, a \rangle \rightarrow \langle c_0, a \rangle = 1, \quad \langle C_0^{(n)}, b \rangle \rightarrow \langle c_0, b \rangle = 1$$

and that

$$\langle c_0, a^2 + b^2 \rangle < +\infty.$$

Let T_{gel} be defined as in Definition 2.4. Then $(C_t^{(n)})_{t \in [0, T_{\text{gel}}]}$ converges (in distribution) to the unique solution of Smoluchowski's equation (2.3) with initial conditions (c_0) .

Remark 7.3. Obviously, convergence has to be understood with respect to Skorokhod's topology on $[0, T_{\text{gel}}]$ (which is the trace topology of Skorokhod's topology on $[0, +\infty)$).

Note that the law P_n of the process $C^{(n)}$ is a probability measure on $\mathbb{D}(\mathbb{R}^+, E)$. To prove Theorem 7.2, we will show that the sequence (P_n) is tight, and that for every limit point P , and almost every process (C_t) with law P , (C_t) solves some system, which is Smoluchowski's equation (2.3) before the critical time. Because of the uniqueness of such a solution, this will show that (P_n) itself converges to the solution of Smoluchowski's equation (2.3) before the critical time.

7.3 Tightness

The proof of tightness is analogous to the one in [27], up to some slight modifications.

Lemma 7.4. *The sequence $(P_n)_{n \geq 0}$ is tight.*

Proof. We will use the classical tightness criterion stated in [28, p. 34] or in [16, Th 7.2]. For $t \geq 0$, let $P_t^{(n)}$ be the law of $C_t^{(n)}$, which is a probability measure on E . Since E is compact by Lemma 7.1, the tightness of the sequence $(P_t^{(n)})_{n \geq 0}$ is obvious.

Now, $C^{(n)}$ is a pure-jump process on $\frac{1}{n}E_n \subset \mathbb{E}$, with generator $G^{(n)}$. Hence, when the process is in the state η , then the time before the next jump is exponential with parameter

$$\lambda_\eta^{(n)} := \sum_{p, p' \in S} \lambda_\eta^{(n)}(p, p') = \frac{1}{2} \left(\sum_{p, p' \in S} np \cdot p' \eta(p) \eta(p') - \sum_{p \in S} p \cdot p \eta(p) \right)$$

and, since $\eta \in E$, $\lambda_\eta^{(n)} \leq M^2 n := cn$. Now take $N > 0$, $\beta > 0$, $\varepsilon > 0$, and let $\delta > 0$ such that $N = \delta l$ for some $l \in \mathbb{N}^*$, and $3c\delta\varepsilon/\beta < 1$. Define now

$$w^N(Y, \delta) := \inf_{\pi \in \Pi_\delta} \max_{t_i \in \pi} \sup_{t_i \leq s < t < t_{i+1}} d(Y_t, Y_s),$$

Π_δ being the set of all subdivisions $0 = t_0 < t_1 < \dots < t_n = N$ of $[0, N]$ such that $t_{i+1} - t_i \geq \delta$ for all i . Consider the partition $t_0 = 0 < t_1 = \delta < \dots < t_l = \lambda = N$ of $[0, N]$. Let $Z_i := \sup_{t_i \leq s < t < t_{i+1}} d(C_s^{(n)}, C_t^{(n)})$ for $0 \leq i \leq l-1$. Then

$$\mathbb{P}_n(w^N(C^{(n)}, \delta) > \beta) \leq \mathbb{P}_n \left(\max_{0 \leq i \leq l-1} Z_i > \beta \right) \leq l \max_{0 \leq i \leq l-1} \mathbb{P}_n(Z_i > \beta).$$

But the size of a jump, that is $d(C_{t-}^{(n)}, C_t^{(n)})$, is $3/n$. Hence, if $Z_i > \beta$, then the process has jumped more than $k := \lceil \beta n / 3 \rceil := \lceil c'n \rceil$ times between t_i and t_{i+1} (where $\lceil x \rceil$ is the first integer strictly greater than x). If S_k is the time of the k -th jump, the Markov property tell us that

$$\mathbb{P}_n(Z_i > \beta) \leq \mathbb{P}_n(S_k \leq \delta).$$

But S_k is the sum of k independent exponential random variables, with parameter smaller than cn . So, if S'_k is the sum of k independent exponential random variables with parameter cn (on a probability space $(\Omega, \mathcal{A}, \mathbb{P})$), then S_k is stochastically dominated by S'_k , that is

$$\mathbb{P}_n(S_k \leq \delta) \leq \mathbb{P}(S'_k \leq \delta).$$

To conclude, note that the last term is the probability that a Poisson process with parameter cn jumps more than k times on $[0, \delta]$, and Stirling's formula shows that this tends to zero for $(c\delta\varepsilon/c') < 1$. \square

7.4 Convergence

In this section, we prove the convergence of $C^{(n)}$ to a process solving the system (2.18), and deduce that it solves Smoluchowski's equation (2.3) before T_{gel} .

Proposition 7.5. *Assume that for every $p \in S$, $C_0^{(n)}(p) \rightarrow c_0(p)$ for some $c_0(p) \geq 0$ and that*

$$\langle C_0^{(n)}, a \rangle \rightarrow \langle c_0, a \rangle = 1, \quad \langle C_0^{(n)}, b \rangle \rightarrow \langle c_0, b \rangle = 1.$$

Let P be a limit point of (P_n) , and let (c_t) be a process with law P . Then a.s. (c_t) solves Equation (2.18), with initial conditions (c_0) .

In the following proofs, we take a subsequence of $(C^{(n)})$ which converges in law to some possibly random $c \in \mathbb{D}(\mathbb{R}^+, E)$. For notational simplicity, we will assume that $(C^{(n)})$ itself tends to c . Since E is compact, it is separable, and hence, so is $\mathbb{D}(\mathbb{R}^+, E)$. Skorokhod's representation theorem (cf e.g. [16]) now allows us to assume that the $C^{(n)}$ are defined on the same probability space $(\Omega, \mathcal{F}, \mathbb{P})$, that $C^{(n)} \rightarrow c$ a.s. (that is, for almost every $\omega \in \Omega$, the function $C^{(n)}(\omega)$ tends to $c(\omega)$ for Skorokhod's distance), and that in the statement, there is a.s. convergence. We will also constantly use the fact that for every bounded Borel function $f : E \rightarrow \mathbb{R}$, the processes

$$f(C_t^{(n)}) - f(C_0^{(n)}) - \int_0^t G^{(n)} f(C_s^{(n)}) \, ds := M_t^{(n)} \quad (2.30)$$

and

$$\left(M_t^{(n)}\right)^2 - \int_0^t \left(G^{(n)}(f^2)(C_s^{(n)}) - 2f(C_s^{(n)})G^{(n)}f(C_s^{(n)})\right) \, ds \quad (2.31)$$

are martingales. Note also that if $f : E \rightarrow \mathbb{R}$ is linear, then, for all $\eta \in E$,

$$G^{(n)}f(\eta) = \frac{1}{2} \sum_{p,p' \in S} f(\Delta_{p,p'}) p \cdot p' \eta(p) \eta(p') - \frac{1}{2n} \sum_{p \in S} f(\Delta_{p,p}) \eta(p) \quad (2.32)$$

and

$$\left(G^{(n)}(f^2) - 2fG^{(n)}f\right)(\eta) = \frac{1}{2n} \left(\sum_{p,p' \in S} f(\Delta_{p,p'})^2 \eta(p) \eta(p') p \cdot p' - \frac{1}{n} \sum_{p \in S} f(\Delta_{p,p})^2 \eta(p) p \cdot p \right) \quad (2.33)$$

We will also need the following convergence result.

Lemma 7.6. *Let*

$$A_s^{(n)} = \langle C_s^{(n)}, a \rangle, \quad B_s^{(n)} = \langle C_s^{(n)}, b \rangle.$$

Then $(A^{(n)})$ and $(B^{(n)})$ C-converge a.s. to $t \mapsto 1/(1+t)$.

Proof. Obviously, we cannot pass to the limit immediately in these expressions. So consider the maps from E to \mathbb{R} : $C \mapsto \langle a, C \rangle$ and $C \mapsto \langle b, C \rangle$, which are measurable and bounded (by M). By (2.30) and (2.32), there are martingales $M^{A,(n)}$ and $M^{B,(n)}$ such that

$$A_t^{(n)} = A_0^{(n)} - \int_0^t A_s^{(n)} B_s^{(n)} \, ds + \varepsilon_n + M_t^{A,(n)}, \quad B_t^{(n)} = B_0^{(n)} - \int_0^t A_s^{(n)} B_s^{(n)} \, ds + \varepsilon_n + M_t^{B,(n)} \quad (2.34)$$

where $\varepsilon_n \leq M/n$. Now, (2.31) and (2.33) show that the quadratic variation of $M^{A,(n)}$ verifies

$$\langle M^{A,(n)} \rangle_t \leq \frac{1}{n} \int_0^t A_s B_s \, ds \leq \frac{M^2 t}{n}.$$

By Doob's inequality,

$$\mathbb{E} \left(\left(\sup_{0 \leq t \leq T} M_t^{A,(n)} \right)^2 \right) \rightarrow 0$$

for all $T > 0$. Hence there is a subsequence of $(M^{A,(n)})$ which C-converges a.s. to 0. In particular, it S-converges. For notational simplicity, we will assume that $(M^{A,(n)})$ itself converges. For the same reason, we may assume that $(M^{B,(n)})$ also S-converges a.s. to 0.

Now, note that the proof of Lemma 7.4 still works for $(A^{(n)})$, since the size of its jumps is bounded by $1/n$. So $(A^{(n)})$ is tight, and by Prokhorov's theorem¹, this means that for almost every $\omega \in \Omega$, $(A^{(n)}(\omega))_{n \geq 0}$ lies in a compact of $\mathbb{D}(\mathbb{R}^+, E)$. The same works for $(B^{(n)})$, so we can find $\Omega' \subset \Omega$ with $\mathbb{P}(\Omega') = 1$, such that for every $\omega \in \Omega'$, $C_n(\omega) \rightarrow c(\omega)$ (for Skorokhod's distance), $M^{A^{(n)}}(\omega) \rightarrow 0$ and $M^{B^{(n)}}(\omega) \rightarrow 0$ (compactly), and $(A^{(n)}(\omega))_{n \geq 0}$ and $(B^{(n)}(\omega))_{n \geq 0}$ lie in a compact.

Next, fix $\omega \in \Omega'$, and let us find the limit of $(A^{(n)}(\omega), B^{(n)}(\omega))$ (for the product topology — which is not Skorokhod's topology on $\mathbb{D}(\mathbb{R}^+, E^2)$). Since it lies in a compact set, it is enough to show that it has only one limit point. So assume $(A^{(n)}(\omega), B^{(n)}(\omega))$ converges to some (A, B) . Then $(A_t^{(n)}(\omega))$ converges to A_t for every $t \in K$, the set of continuity points of A . But A is càdlàg, so it has only countably many points of discontinuity, and hence K^c has Lebesgue-measure 0. Hence $(A^{(n)}(\omega))$ converges to A Lebesgue-a.s., and ditto for $(B^{(n)}(\omega))$. Also, $(A^{(n)}(\omega))$ and $(B^{(n)}(\omega))$ are bounded by M , so using dominated convergence in (2.34) and recalling that $A_0^{(n)}(\omega)$ and $B_0^{(n)}(\omega) \rightarrow 1$ by assumption, we obtain

$$A_t = 1 - \int_0^t A_s B_s \, ds, \quad B_t = 1 - \int_0^t A_s B_s \, ds.$$

Hence

$$A_t = B_t = \frac{1}{1+t}.$$

Finally there is only one limit point to $(A^{(n)}(\omega), B^{(n)}(\omega))$. So $(A^{(n)}(\omega))$ and $(B^{(n)}(\omega))$ both S-converge to $t \mapsto 1/(1+t)$, and, since this function is continuous, they C-converge. \square

Remark 7.7. As pointed out in the proof, the convergence of $A^{(n)}$ and $B^{(n)}$ to the actual number of arms

$$A_t = \langle c_t, a \rangle, \quad B_t = \langle c_t, b \rangle$$

is not obvious. There is no such problem for a strictly sublinear coagulation rate (as in Jeon's proof [27]). In our (linear) case, we prove below that this convergence holds before the critical time (we also refer to Norris [45] for general sublinear rates in a model with no arms). In fact, if there is a solution (c_t) to (2.3) defined after T_{gel} , we believe, for the reasons explained in Section 2.4 of the introduction, that $A_t^{(n)}$ and $B_t^{(n)}$ do not converge to A_t and B_t after T_{gel} (and that this number of arms is then strictly lesser than $1/(1+t)$).

Proof of Proposition 7.5. 1. Take some $p_0 = (a_0, b_0, m_0) \in S$, and define, for $C \in \mathbb{D}(\mathbb{R}^+, E)$, $f(C) = C(p_0)$. According to (2.30),

$$C_t^{(n)}(p_0) - C_0^{(n)}(p_0) - \int_0^t G^{(n)} f(C_s^{(n)}) \, ds := M_t^{p_0, (n)} \quad (2.35)$$

is a martingale. Note also that for $p, p' \in S$, $f(\Delta_{p, p'})$ is 0, except if p or p' or $p \circ p'$ is p_0 . Hence, it is easy to check using (2.30) that

$$\begin{aligned} G^{(n)} f(C_s^{(n)}) &= - \sum_{p \in S} C_s^{(n)}(p) C_s^{(n)}(p_0) p_0 \cdot p + \frac{1}{2} \sum_{p \leq p_0} p \cdot (p_0 \setminus p) C_s^{(n)}(p) C_s^{(n)}(p_0 \setminus p) \\ &\quad - \frac{1}{n} \sum_{p \in S} f(\Delta_{p, p}) C_s^{(n)}(p). \end{aligned} \quad (2.36)$$

¹Actually, we do not need this implication of Prokhorov's theorem: this is just a consequence of the proof of Lemma 7.4

The last term is due to the difference between $\lambda_\eta(p, p')$ when $p \neq p'$ and when $p = p'$. In any case, it tends to 0 uniformly on \mathbb{R}^+ and uniformly in p_0 .

2. Let us now study the martingale term. By Doob's inequality, we have for every $T > 0$

$$\mathbb{E} \left(\left(\sup_{0 \leq t \leq T} M_t^{p_0, (n)} \right)^2 \right) \leq 4\mathbb{E} \left(\left(M_T^{p_0, (n)} \right)^2 \right)$$

and by (2.31), this last term is

$$\mathbb{E} \left(\int_0^T (G^{(n)} f^2 - 2fG^{(n)} f)(C_s^{(n)}) ds \right).$$

But by (2.33), and since $f(\Delta_{p, p'}) \leq M$ for all $p, p' \in S$, then

$$(G^{(n)} f^2 - 2fG^{(n)} f)(C_s^{(n)}) \leq M^4/n,$$

so that

$$\mathbb{E} \left(\left(\sup_{0 \leq t \leq T} M_t^{p_0, (n)} \right)^2 \right) \rightarrow 0.$$

Hence, there is a subsequence of $(M^{p_0, (n)})$ which a.s. converges to 0 uniformly on \mathbb{R}^+ . For notational simplicity, we will now assume that $(M^{p_0, (n)})$ itself C-converges to 0. Using the diagonal method, we may as well assume that $(M^{p_0, (n)})$ C-converges to 0 for every $p_0 \in S$.

3. We have already seen in the proof of Lemma 7.4 that $d(C_t^{(n)}, C_{t^-}^{(n)}) \leq 3/n$ a.s. By continuity of $X \mapsto \sup_{s \in [0, t]} d(X_{s^-}, X_s)$ (cf [16]), this ensures that c is almost surely continuous, so $C^{(n)}$ actually C-converges to c . From the definition of d , it is also obvious that $C^{(n)}(p)$ C-converges to $c(p)$ for every $p \in S$.
4. With these results, we may now pass to the limit in (2.35) and (2.36). Write (2.36) in the form

$$G^{(n)} f(C_s^{(n)}) = -\alpha_n(s) + \frac{1}{2}\beta_n(s) + \varepsilon_n(s).$$

Equation (2.35) shows that

$$C_t^{(n)}(p_0) = C_0^{(n)}(p_0) - \int_0^t \alpha_n(s) ds + \frac{1}{2} \int_0^t \beta_n(s) ds + \int_0^t \varepsilon_n(s) ds + M_t^{(n)}. \quad (2.37)$$

By Point 3, $C_t^{(n)}(p_0)$ C-converges a.s. to $c(p_0)$. $C_0^{(n)}(p_0)$ tends to $c_0(p_0)$ by assumption. $\beta_n(t)$ is a finite sum, so

$$\beta_n(t) \rightarrow \sum_{p \preceq p_0} p \cdot (p_0 \setminus p) C_s^{(n)}(p) C_s^{(n)}(p_0 \setminus p)$$

compactly. Finally, note that

$$\begin{aligned} \alpha_n(s) &= C_s^{(n)}(p_0) \left(a_0 \langle C_s^{(n)}, b \rangle + b_0 \langle C_s^{(n)}, a \rangle \right) \\ &= C_s^{(n)}(p_0) (a_0 B_s^{(n)} + b_0 A_s^{(n)}). \end{aligned}$$

By Lemma 7.6, $A_t^{(n)}$ and $B_t^{(n)}$ converge compactly to $t \mapsto 1/(1+t)$, so

$$\lim_{n \rightarrow +\infty} \alpha_n(s) = \frac{a+b}{1+t} c_s(p_0) \quad \text{a.s.}$$

compactly. Since these are all compact convergences, we can pass to the limit in (2.37), for all $p_0 \in S$. This readily shows that $(c(p))$ solves (2.18), with initial conditions (c_0) . □

7.5 End of the proof

We can now end the proof of Theorem 7.2. Hence, we assume now that $\langle c_0, a^2 + b^2 \rangle < +\infty$. Let Q_n be the law of $(C_t^{(n)})_{t \in [0, T_{\text{gel}}]}$. The sequence (Q_n) is obviously tight. Let Q one of its limit points, and let c a process with law Q . By Proposition 7.5 above, c solves a.s. the system (2.18), with initial conditions (c_0) .

Now, let $g_t(x, y, z)$ the (a priori random) generating function of c . It is easy to see that g is well defined for $(t, x, y, z) \in [0, T_{\text{gel}}] \times (0, 1)^2 \times [0, 1]$, and that $g_t(\cdot, \cdot, z)$ is regular for every $z \in [0, 1]$. Moreover, we see as in the proof of Proposition 4.7 that for every $z \in [0, 1]$, $g_t(\cdot, \cdot, z)$ solves the PDE (2.17) with initial conditions $(x, y) \mapsto g_0(x, y, z) = \sum c_0(a, b, m) x^a y^b z^m$. Hence by Proposition 4.7, (c_t) solves Smoluchowski's equation 2.3 until T_{gel} . But by Theorem 2.5, there is a unique solution to this equation on $[0, T_{\text{gel}}]$. Hence there is a unique limit point to (Q_n) , so the sequence itself converges to the solution of Smoluchowski's equation (2.3) on $[0, T_{\text{gel}}]$.

Chapter 3

Uniqueness of post-gelation solutions of coagulation equations

This chapter is taken from a joint work [44] with Lorenzo Zambotti, published in the *Annales de l'Institut Henri Poincaré, Analyse non-linéaire*.

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1 Introduction

1.1 Coagulation models

In this chapter, we deal with the problem of uniqueness of post-gelation solutions of several models of coagulations, namely Smoluchowski's and Flory's classical models, Bertoin's models with limited aggregations and the corresponding Flory equation. Let us recall that Smoluchowski's equation concerns the evolution of the concentration of $c_t(m)$ of clusters of mass $m \in \mathbb{N}^*$ and reads

$$\frac{d}{dt}c_t(m) = \frac{1}{2} \sum_{m'=1}^{m-1} \kappa(m, m') c_t(m') c_t(m - m') - \sum_{m' \geq 1} \kappa(m, m') c_t(m) c_t(m') \quad (3.1)$$

with $m \in \mathbb{N}^*$, for a symmetric kernel $\kappa(m, m')$, and that Bertoin's coagulation with limited aggregations deals with the evolution of the concentration $c_t(a, m)$ of clusters with a arms and mass m , and is given by

$$\begin{aligned} \frac{d}{dt}c_t(a, m) &= \frac{1}{2} \sum_{a'=1}^{a+1} \sum_{m'=1}^{m-1} a'(a+2-a')c_t(a', m')c_t(a+2-a', m-m') \\ &\quad - \sum_{a' \geq 1} \sum_{m' \geq 1} aa'c_t(a, m)c_t(a', m') \end{aligned} \quad (3.2)$$

for $(a, m) \in \mathbb{N} \times \mathbb{N}^*$.

As we already discussed in the introduction of this thesis, in general, when solving these equations, a critical time, known as the gelation time, appears. In [46], Norris considers far more general models of cluster coagulation, where the rate of coalescence does not depend only on the mass of the clusters but also on other parameters. In that case again, most results (existence, uniqueness) are obtained before some critical time.

1.2 The gelation phase transition

The reason why Bertoin's equation (3.2) can be solved, is that it can be transformed (see [3]) into a solvable PDE involving the generating function of (c_t) . For the standard Smoluchowski equation (3.1), this transformation is also possible for several particular choices of the kernel $\kappa(m, m')$, namely when κ is constant, additive or multiplicative: see e.g. [7]. However, the mass is a parameter of this PDE, so it is easy to solve only when the mass is known. But if large clusters can coagulate sufficiently fast, then one may observe in finite time the gelation phenomenon, which is interpreted as the formation of clusters of infinite mass, the gel. From the gelation time, the mass starts to decrease.

Existence and uniqueness of solutions of (3.1) are thus easy up to gelation, since in this regime the total mass

$$M_t := \sum_{m \geq 1} mc_t(m),$$

which appears in (3.1) and in the associated PDE, is constant.

It is well-known that the occurrence of gelation depends heavily on the choice of the coagulation rate $\kappa(m, m')$, and that in the multiplicative case, gelation always occurs [14, 18, 35]. After gelation, the mass is not known, so M_t itself becomes an unknown of the equation, and well-posedness of the equation is then much less trivial.

The multiplicative kernel is therefore particularly interesting, since it exhibits a non-trivial behavior but can also be studied in detail by means of explicit computations.

For (3.2), before gelation the mean number of arms A_t is explicitly known and equal to

$$A_t := \sum_{a,m \geq 1} ac_t(a,m) = \frac{A_0}{1 + tA_0}.$$

As we saw, the same phenomenon of gelation occurs in (3.2) for a class of initial concentrations c_0 . After that time, A_t is lesser than $A_0/(1 + tA_0)$, and the equation becomes much harder to solve.

1.3 Main result

The main results of this chapter are the well-posedness of the standard (3.1) Smoluchowski equation with a multiplicative kernel, and of the modified (3.2) Smoluchowski equation, for general initial conditions.

We start off in Section 2 by considering existence, uniqueness and representation formulae for global solutions of the version of (3.1) with continuous masses and a multiplicative kernel, introducing and exploiting all main techniques which are needed afterwards to tackle the same issues in the case of Bertoin's equation (3.2). We prove that for the most general initial conditions $\mu_0(dm)$, a positive measure on $(0, +\infty)$, Smoluchowski's equation with a multiplicative kernel has a unique solution before and after gelation. We can in particular consider initial concentrations with infinite total mass, i.e. such that

$$M_0 := \int_{(0,+\infty)} m \mu_0(dm) = +\infty.$$

This can be applied for instance to initial conditions of Gamma type

$$m c_0(dm) = C_p m^{p-2} e^{-m}$$

with $p \in (0, 1]$.

We also show existence and uniqueness for the modified version of Smoluchowski's model, namely Flory's equation (1.2), in Section 3. This was already proved by Norris [46] for much more general models, but the techniques in that particular case are much simpler and allow to compute many interesting quantities.

A probably more interesting case is Bertoin's model with limited aggregations (3.2) and we prove analogous results as for (3.1) in Section 4. We can in particular consider initial conditions $(c_0(a, m), a \in \mathbb{N}, m \in \mathbb{N}^*)$ with an initial infinite number of arms, that is such that

$$A_0 := \sum_{a,m \geq 1} ac_0(a, m)$$

is infinite, and show that there is a unique solution "coming down from infinity sufficiently fast", i.e. such that, for positive t ,

$$\int_0^t A_s^2 ds < +\infty.$$

Note however that this is no technical condition, but a mere assumption to ensure that the equation is well-defined. The same techniques as for the standard Smoluchowski equation of Section 2 are used, so we shall not give all the details in that case. We can also consider the corresponding Flory model with limited aggregations, which is dealt with in Section 5.

A most striking feature of the result about (3.2) is the expression for the limiting concentrations which we compute in Section 6, which are not trivial, in comparison with

the standard Smoluchowski and Flory cases (for which they are always zero). In Flory's case with limited aggregations, the formula obtained extends to all possible initial concentrations the computations done in [3] when there is no gelation. In Smoluchowski's model with limited aggregations, a slight modification appears, which calls for a probabilistic interpretation which is the motivation for the study of a microscopic model for (3.2) undertaken in the fourth chapter.

The technique used, as in [3], is to transform the equation into a PDE. However, since the mass (in the case of (3.1)) or the mean number of arms (in the case (3.2)) is not a priori known, this PDE is non local, unlike the one obtained in the regime before gelation, and this is the main difficulty we have to deal with. We may though use a modification of the classical characteristics method to show the uniqueness of a solution to this PDE, and hence to (3.2).

This seems to be the first case of a cluster coagulation model for which global well-posedness in presence of gelation can be proven. Another setting to which these techniques could be applied is the coagulation model with mating [43] introduced in the second chapter of this thesis.

1.4 Bibliographical comments

Smoluchowski's equation (3.1) has been extensively studied; we refer to the reviews [1, 33, 45]. There are conditions about the kernel κ to know if gelation occurs or not, but this requires a precise definition of gelation (see e.g. [15], or [27] in a probabilistic setting). Essentially, before gelation, Smoluchowski's solution has a unique solution [45, 8, 18, 33]. However, in the multiplicative case, gelation always occurs [14, 18, 35]. For monodisperse initial conditions $c_0(m) = \mathbb{1}_{\{m=1\}}$, the first proof of existence and uniqueness to (3.1) before gelation is given in [39], and a proof of the global existence and uniqueness in [29], essentially because in this case, explicit solutions can be obtained. The case of general nonzero initial conditions has been considered by several papers in the Physics literature [12, 13, 36, 53, 59], and by at least one mathematical paper [55], which however treats in full details only the regime before gelation (see Remark 2.9 below). The same authors also provide in [56] an exact formula for the post-gelation mass of (3.1), but with no rigorous proof. Thus, a clear statement about well-posedness of (3.1) for the most general initial conditions still seems to be missing.

The use of generating functions (or Laplace transforms) is classical [7, 3], and it makes the proof of uniqueness before gelation for a multiplicative kernel quite easy, see [40].

We will not be interested in the existence of solutions, since it has been obtained in a much more general setting by analytic [34, 35, 46] or probabilistic [19, 27] means. However, the case of an infinite initial mass seems to have been considered only in [34] in the discrete case, so we refer to Section 2.6 below for a proof.

2 Smoluchowski's equation

2.1 Notation and results

In this section we develop our method in the case of the continuous version of the standard Smoluchowski equation (3.1), proving existence, uniqueness and representation formulae for global solutions. Let us first fix some notations.

- \mathcal{M}_f^+ is the set of all non-negative finite measures on $(0, +\infty)$.
- \mathcal{M}_c^+ is the set of all non-negative Radon measures on $(0, +\infty)$.

- For $\mu \in \mathcal{M}_c^+$ and $f \in L^1(\mu)$ or $f \geq 0$,

$$\langle \mu, f \rangle = \int_{(0, +\infty)} f(m) \mu(dm).$$

We will write m for the function $m \mapsto m$, m^2 for $m \mapsto m^2$, etc.

- For $\phi : (0, +\infty) \rightarrow \mathbb{R}$ and $m, m' > 0$, $\Delta\phi(m, m') = \phi(m + m') - \phi(m) - \phi(m')$.
- $C_c(\mathbb{R}^+)$ is the space of continuous functions on $(0, +\infty)$ with compact support.
- For a function $(t, x) \mapsto \phi_t(x)$, $\phi'_t(x)$ is the partial derivative of ϕ with respect to x .
- $\frac{\partial^+}{\partial t}$ or $\frac{d^+}{dt}$ denotes the right partial derivative with respect to t .

Let us now define our equation and we call a solution. Note that the first requirement in the following definition is only present for the equation to make sense.

Definition 2.1. Let $\mu_0 \in \mathcal{M}_c^+$. We say that a family $(\mu_t)_{t \geq 0} \subset \mathcal{M}_c^+$ solves Smoluchowski's equation if

- for every $t > 0$, $\int_0^t \langle \mu_s(dm), m \rangle^2 ds < +\infty$,
- for all $\phi \in C_c(\mathbb{R}^+)$ and $t > 0$

$$\langle \mu_t, \phi \rangle = \langle \mu_0, \phi \rangle + \frac{1}{2} \int_0^t \langle \mu_s(dm) \mu_s(dm'), mm' \Delta\phi(m, m') \rangle ds. \quad (3.3)$$

The global behavior of this equation has been studied first for monodisperse initial conditions (i.e. $\mu_0 = \delta_1$), in which case it can be proven that there is a unique solution $(\mu_t)_{t \geq 0}$ on \mathbb{R}^+ , which is also explicit, see [39, 29]. This solution clearly exhibits the gelation phase transition. Up to the gelation time $T_{\text{gel}} = 1$, the total mass $\langle \mu_t, m \rangle$ is constant and equal to 1, and then it decreases: $\langle \mu_t, m \rangle = 1/t$ for $t \geq 1$. Moreover, the second moment $\langle \mu_t, m^2 \rangle$ is finite before time 1, and then infinite on $[1, +\infty)$. It is also known in the literature that for any nonzero initial conditions, there is a gelation time $0 < T_{\text{gel}} < +\infty$, such that there is a unique solution to (3.3) on $[0, T_{\text{gel}})$, and $\langle \mu_t, m^2 \rangle \rightarrow +\infty$ when $t \rightarrow T_{\text{gel}}^-$: see e.g. [18].

Our initial condition will be a non-null measure $\mu_0 \in \mathcal{M}_c^+$ such that

$$\langle \mu_0, m \wedge 1 \rangle = \int_{(0, +\infty)} (m \wedge 1) \mu_0(dm) < +\infty. \quad (3.4)$$

We can then define the generating function

$$g_0(x) := \langle \mu_0, mx^m \rangle = \int_{(0, +\infty)} mx^m \mu_0(dm), \quad x \in [0, 1], \quad (3.5)$$

as well as the gelation time.

Definition 2.2. For

$$M_0 := \langle \mu_0, m \rangle \in (0, +\infty], \quad K := \langle \mu_0, m^2 \rangle \in (0, +\infty],$$

the gelation time is defined as

$$T_{\text{gel}} := 1/K \in [0, +\infty). \quad (3.6)$$

Our main result on (3.3) is then the following.

Theorem 2.3. *Smoluchowski's equation (3.3) has a unique solution on \mathbb{R}^+ . It has the following properties.*

1. *The total mass $M_t = \langle \mu_t, m \rangle$ is given by $M_t = g_0(\ell_t)$, where $\ell_1 = 1$ for $t \leq T_{\text{gel}}$ and, for $t > T_{\text{gel}}$, ℓ_t is uniquely defined by*

$$\ell_t g_0'(\ell_t) = \frac{1}{t}.$$

Therefore, $t \mapsto M_t$ is continuous on $[0, +\infty)$, constant on $[0, T_{\text{gel}}]$, strictly decreasing on $[T_{\text{gel}}, +\infty)$ and analytic on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$.

2. *The function*

$$\phi_t(x) = x \exp\left(\int_0^t g_0(\ell_s) ds\right) e^{-t g_0(x)}, \quad x \in [0, 1], t > 0,$$

has a right-inverse $h_t : [0, 1] \rightarrow [0, \ell_t]$. The generating function $g_t(x) := \langle \mu_t, m x^m \rangle$ is given for $t \geq 0$ by

$$g_t(x) = g_0(h_t(x)).$$

3. *If the following limit exists*

$$\nu := - \lim_{x \rightarrow 1^-} \frac{(g_0'(x))^3}{g_0'(x) + x g_0''(x)} \in [-\infty, 0],$$

then the right derivative $\dot{M}_{T_{\text{gel}}}$ of M at $t = T_{\text{gel}}$ is equal to ν .

4. *Let $m_0 = \inf \text{supp } \mu_0 \in [0, +\infty)$. When $t \rightarrow +\infty$,*

$$\frac{1}{t M_t} \rightarrow m_0.$$

5. *The second moment $\langle \mu_t, m^2 \rangle$ is finite for $t \in [0, T_{\text{gel}})$ and infinite for $t \in [T_{\text{gel}}, +\infty)$.*

Remark 2.4. • This result allows to recover the pre- and post-gelation formulas obtained with no rigorous proof in some earlier papers [13, 12, 29, 36, 55, 56, 53]. The decrease of the mass in $1/t$ when $m_0 > 0$ was also observed in these papers. Also, some upper bounds in $1/t$ for the mass were proven in [15, 35].

- If there exists *dust*, in the classical terminology [2] of coagulation-fragmentation, i.e. if $m_0 = 0$, the mass tends to 0 more slowly than $1/t$: small clusters need to coagulate before any big cluster can appear, and they coagulate really slowly. For instance, a straightforward computation shows that if $\mu_0(dm) = e^{-m} dm$, then $M_t \sim t^{-2/3}$. More generally, the explicit formula in Proposition (2.8) allows to compute M_t for any initial conditions.
- With this formula, it is easy to check that $\dot{M}_{T_{\text{gel}}+}$ can be anything from $-\infty$ to 0. For instance, $\dot{M}_{T_{\text{gel}}+} = 0$ for $g_0(x) = (1-x) \log(1-x) + x$, $\dot{M}_0 = -\infty$ for $g_0(x) = \sqrt{1-x} \log(1-x) + x$, and for $0 < \alpha < +\infty$, $\dot{M}_0 = -\alpha$ for $g_0(x) = 1 - \sqrt{1-x^{2\alpha}}$. In particular, M need not be convex on $[T_{\text{gel}}, +\infty)$.

2.2 Preliminaries

Let μ_0 be defined as in the previous statement. We shall prove that, starting from μ_0 , there is a unique solution to (3.3) on \mathbb{R}^+ , and give a representation formula for this solution. This allows to study the behavior of the moments. Let us start with some easy lemmas. So take a solution $(\mu_t)_{t \geq 0}$ to (3.3) and set

$$M_t = \langle \mu_t, m \rangle. \quad (3.7)$$

The two following lemmas are easy to prove, using monotone and dominated convergence.

Lemma 2.5. *$(M_t)_{t \geq 0}$ is monotone non-increasing and right-continuous. Moreover, $M_t < +\infty$ for all $t > 0$.*

Proof. Take $\phi^K(m) = m$ for $m \in [0, K]$, $\phi^K(m) = 2K - m$ for $m \in [K, 2K]$, and $\phi^K(m) = 0$ for $m \geq 2K$, so that $\phi^K \in C_c$. Plugging ϕ^K in Smoluchowski's equation (3.3), letting $K \rightarrow +\infty$ and using Fatou's lemma readily shows that $(M_t)_{t \geq 0}$ is monotone non-increasing. Note also that $t \mapsto M_t = \sup_K \langle \mu_t, \phi^K \rangle$ is the supremum of a sequence of continuous functions and so is lower semi-continuous, which implies, for a monotone non-increasing function, right-continuity. Finiteness of M_t is now obvious since $s \mapsto M_s^2$, and hence $s \mapsto M_s$, are integrable by Definition 2.1. \square

Lemma 2.6. *Assume that $\langle \mu_0, m^2 \rangle < +\infty$. Then $t \mapsto \langle \mu_t, m^2 \rangle$ is bounded in a neighborhood $[0, T_0]$, $T_0 > 0$, of 0. Moreover $M_t = M_0$ for $t \in [0, T_0]$.*

Proof. The first part of the statement is analogous to Lemma 3.6 of Chapter 2. More precisely, first note that (3.3) can be extended to any bounded $\phi : (0, +\infty) \rightarrow \mathbb{R}^+$ by a standard approximation procedure, so we may take take, for $M \in \mathbb{N}$,

$$\phi(m) = \phi^M(m) := (m \wedge M)^2.$$

It is easy to check that $\Delta\phi(m, m') \leq 2(m \wedge M)(m' \wedge M)$, so plugging this in (3.3), we get

$$\langle \mu_t, \phi^M \rangle \leq \langle \mu_0, \phi^M \rangle + \int_0^t \langle \mu_s, dm \mu_s, dm' \rangle, (m \wedge M)^2 (m' \wedge M)^2 \rangle ds = \int_0^t (\langle \mu_s, \phi^M \rangle)^2 ds.$$

This readily shows that $\langle \mu_t, \phi^M \rangle \leq (1/\langle \mu_0, \phi^M \rangle - t)^{-1}$ as long as the RHS remains finite, so by monotone convergence, having $M \rightarrow +\infty$, we get

$$\langle \mu_t, m^2 \rangle \leq \frac{1}{1/\langle \mu_0, m^2 \rangle - t}$$

for $t < 1/\langle \mu_0, m^2 \rangle$, whence the result follows. The second part is classical and is just obtained by dominated convergence. \square

Remark 2.7. The first part of this result was actually absent from the original paper [44], what led to an awkward definition of a solution where we required the second moment to be bounded in a neighborhood of 0. As we just saw, this is an unnecessary assumption.

2.3 PDE

By Lemma 2.5, $\langle \mu_t, m \rangle < +\infty$ for $t > 0$, so that we can define

$$g_t(x) = \langle \mu_t, mx^m \rangle = \int_{(0,+\infty)} mx^m \mu_t(dm), \quad x \in [0, 1], \quad t > 0, \quad (3.8)$$

which is the generating function of $m \mu_t(dm)$. Then, using a standard approximation procedure, it is easy to see that g satisfies

$$\begin{cases} g_t(x) = g_0(x) + \int_0^t x(g_s(x) - M_s) \frac{\partial^+ g_s}{\partial x}(x) ds, & t \geq 0, \quad x \in (0, 1) \\ g_t(1) = M_t, & t \geq 0. \end{cases} \quad (3.9)$$

It is well-known, and will be proven again below, that $M_t = M_0$ for all $t \leq T_{\text{gel}}$, since then, the PDE (3.29) can be solved by the method of characteristics: the function $\phi_t(\cdot) : [0, 1] \rightarrow [0, 1]$ such that

$$\phi_t(x) = xe^{t(M_0 - g_0(x))}, \quad x \in [0, 1], \quad t \leq T_{\text{gel}}$$

is one-to-one and onto, has an inverse $h_t : [0, 1] \rightarrow [0, 1]$ and we find

$$g_t(x) = g_0(h_t(x)), \quad x \in [0, 1], \quad t \leq T_{\text{gel}}.$$

However M_t is not necessary constant for $t > T_{\text{gel}}$ and the form of ϕ_t has to be modified. We thus define

$$\phi_t(x) = x\alpha_t e^{-tg_0(x)}, \quad x \in [0, 1], \quad t > 0 \quad (3.10)$$

where

$$\alpha_t := \exp\left(\int_0^t M_s ds\right), \quad t \geq 0. \quad (3.11)$$

For $t > T_{\text{gel}}$, M_t is possibly less than M_0 and ϕ_t , which depends explicitly on $(M_s)_{s \in [0, t]}$, is possibly neither injective nor surjective. We shall prove that it is indeed possible to find $\ell_t \in (0, 1)$ such that $\phi_t(x) : [0, 1] \mapsto [0, \ell_t]$ is one-to-one and ℓ_t is uniquely determined by g_0 .

2.4 Uniqueness of solutions

Using an adaptation of the method of characteristics, we are going to prove the following result. Note that in [55], this properties are claimed to be true but a proof seems to lack. We will use the same techniques in the proof of Theorem 4.3 for the model with limited aggregations, but they are easier to understand in the present case.

Proposition 2.8. *Let $(\mu_t)_{t \geq 0}$ be a solution of Smoluchowski's equation (3.3).*

1. *For all $t \in [0, T_{\text{gel}}]$, $M_t = M_0 = g_0(\ell_t)$, where $\ell_t := 1$. For all $t > T_{\text{gel}}$, $M_t = g_0(\ell_t)$ where $\ell_t \in (0, 1)$ is uniquely defined by*

$$\ell_t g_0'(\ell_t) = \frac{1}{t}. \quad (3.12)$$

Moreover ℓ_t and $\phi_t(\cdot)$ satisfy

$$\phi_t'(\ell_t) = 0, \quad \phi_t(\ell_t) = 1 > \phi_t(x), \quad \forall x \in (0, 1). \quad (3.13)$$

2. For all $t > 0$, the function $\phi_t(\cdot)$ defined in (3.10) has a right inverse

$$h_t : [0, 1] \mapsto [0, \ell_t], \quad \phi_t(h_t(x)) = x, \quad x \in [0, 1], \quad (3.14)$$

and

$$g_t(x) = g_0(h_t(x)), \quad t > 0, \quad x \in [0, 1]. \quad (3.15)$$

3. The functions $(\ell_t)_{t \geq 0}$ and $(M_t)_{t \geq 0}$ are continuous.

4. (μ_t) is uniquely defined by μ_0 .

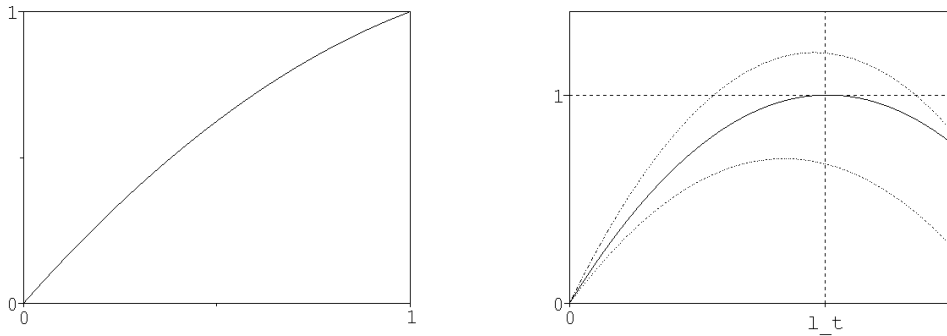


Figure 3.1: ϕ_t before and after gelation. The dotted lines represent what ϕ_t may look like. The solid one is the actual ϕ_t .

Remark 2.9. • For all $t \leq T_{\text{gel}}$, $M_t = M_0$, $\ell_t = 1$ and $\phi_t : [0, 1] \mapsto [0, 1]$ is one-to-one and onto. The first thing one needs to prove is that for all $t > T_{\text{gel}}$, $\ell_t < 1$, i.e. there is indeed $x \in [0, 1]$ such that $\phi_t(x) = 1$, see Lemma 2.11; the second one, is that $\ell_t = m_t$, i.e. $\phi_t(\cdot)$ has an absolute maximum at ℓ_t , see Lemma 2.12. In other words, one has to exclude the dotted lines as possible profiles of $\phi_t(\cdot)$ in Figure 2.4. These properties are not obvious, since ϕ_t depends on $(M_s)_{s \in [0, t]}$ which is, at this point, unknown. All other properties are derived from these two.

- In [55, Section 6] one finds a discussion of post-gelation solutions, in particular of the results of our Proposition 2.8. However this discussion falls short of a complete proof, since the two above-mentioned properties are not proven. In particular, no precise statement about what initial conditions can be considered is given.

The following lemma is a list of obvious but useful properties satisfied by g and ϕ .

Lemma 2.10. *The function g defined in (3.8) satisfies the following properties.*

- (a1) $(t, x) \mapsto g_t(x)$ is finite and continuous on $[0, +\infty) \times [0, 1)$;
- (a2) For all $x \in [0, 1)$, $t \mapsto g_t(x)$ is right differentiable on $(0, +\infty)$;
- (a3) For all $t \geq 0$, $x \mapsto g_t(x)$ is analytic on $(0, 1)$ and monotone non-decreasing;
- (a4) For all $t > 0$, $x \mapsto g_t(x) \in [0, +\infty]$ is continuous on $[0, 1]$.

The function ϕ defined in (3.10) satisfies the following properties.

- (b1) ϕ_t is continuous on $[0, 1]$ and analytic on $(0, 1)$;

(b2) $\phi_t(0) = 0$, $\phi_t(1) = e^{-\int_0^t (M_0 - M_s) ds} \in [0, 1]$;

(b3) $\phi'_t(x) = \alpha_t e^{-tg_0(x)}(1 - txg'_0(x))$ for $x \in (0, 1)$;

(b4) For $t \leq T_{\text{gel}}$, ϕ_t is increasing. For $t > T_{\text{gel}}$, $x \mapsto xg'_0(x)$ is increasing, $\phi'_t(0) > 0$ and $\phi'_t(1) < 0$. In particular, for $t > T_{\text{gel}}$, there is precisely one point $m_t \in (0, 1)$ such that

$$\phi'_t(m_t) = 0. \quad (3.16)$$

(b5) For $t > T_{\text{gel}}$, ϕ_t is increasing on $[0, m_t]$ and decreasing on $[m_t, 1]$.

Moreover,

(c1) The map $(t, x) \mapsto \phi_t(x)$ is continuous on $\mathbb{R}^+ \times [0, 1]$;

(c2) The map $(t, x) \mapsto \phi'_t(x)$ is continuous on $\mathbb{R}^+ \times (0, 1)$;

(c3) For every $x \in [0, 1]$, $t \mapsto \phi_t(x)$ is right differentiable and

$$\frac{\partial^+ \phi_t(x)}{\partial t} = \phi_t(x)(M_t - g_0(x)) \quad x \in [0, 1], t \geq 0. \quad (3.17)$$

Property (b5) implies that there are at most two points in $(0, 1)$ where ϕ_t equals 1. Take ℓ_t to be the smallest, if any, i.e.

$$\ell_t = \inf\{x \geq 0 : \phi_t(x) = 1\} \quad (\inf \emptyset := 1). \quad (3.18)$$

Lemma 2.11. 1. For every $t \geq 0$ and every $x \in [0, \ell_t]$

$$g_t(\phi_t(x)) = g_0(x). \quad (3.19)$$

2. For all $t \in [0, T_{\text{gel}}]$, $\ell_t = 1$, and for $t > T_{\text{gel}}$, $0 < \ell_t < 1$. In particular, for all $t > 0$, $\phi_t(\ell_t) = 1$ and

$$g_0(\ell_t) = g_t(1) = M_t. \quad (3.20)$$

3. Finally, $t \mapsto \ell_t$ is monotone non-increasing and continuous on \mathbb{R}^+ .

Proof. 1. Let us first prove that there exists $\tau > 0$ such that (3.19) holds for $t \in [0, \tau]$. Fix $0 < a < b < 1$. Since $0 < \min_{[a,b]} \phi_0 < \max_{[a,b]} \phi_0 < 1$, then by property (c1) there is $\tau > 0$ such that

$$0 < \min_{[a,b]} \phi_t < \max_{[a,b]} \phi_t < 1, \quad \forall t \in [0, \tau].$$

So, for a fixed $x \in [a, b]$, the function

$$u_t := g_t(\phi_t(x)) - g_0(x)$$

is well-defined and using (3.29) and (3.17), we see that

$$u_t = \int_0^t \left(\frac{\partial^+ g_s}{\partial s}(\phi_s(x)) + \frac{\partial g_s}{\partial x}(\phi_s(x)) \frac{\partial^+ \phi_s}{\partial s}(x) \right) ds = \int_0^t \gamma_s u_s ds$$

where

$$\gamma_t := \frac{\partial g_t}{\partial x}(\phi_t(x)) \phi_t(x), \quad t > 0.$$

Since $x \in [0, 1]$, $\sup_{t \in [0, \tau]} |\gamma_t| < +\infty$ and therefore $u_t \equiv 0$. Hence (3.19) holds for $x \in [a, b]$ and $t \in [0, \tau]$. Since both terms of (3.19) are analytic functions of x on $(0, \ell_t)$, by analytic continuation, (3.19) actually holds on $(0, \ell_t)$, and hence on $[0, \ell_t]$ by continuity.

2. Let us now extend this formula to $t \in \mathbb{R}^+$. Let

$$T = \sup\{t > 0 : \forall s \in [0, t], \forall x \in [0, \ell_s], g_s(\phi_s(x)) = g_0(x)\} \geq \tau > 0,$$

assume $T < +\infty$, and denote by ℓ the left limit of $(\ell_t)_{t \geq 0}$ at T . First, ℓ cannot be 0, since otherwise we would get, when $s \rightarrow T^-$,

$$1 = \phi_s(\ell_s) = \ell_s \alpha_s e^{-s g_0(\ell_s)} \rightarrow 0.$$

For every $t < T$, $0 < \ell \leq \ell_t$, so for every $x \in (0, \ell)$, $g_t(\phi_t(x)) = g_0(x)$ and $\phi_t(x) < 1$. Using the continuity property (c1) and passing to the limit when $t \rightarrow T^-$ in this equality, we get

$$g_T(\phi_T(x)) = g_0(x), \quad \forall x \in (0, \ell).$$

By the same reasoning as in point (i), we obtain a $T' > T$ such that $g_t(\phi_t(x)) = g_0(x)$ for all $t \in [T, T')$ and x in a non-empty open subset of $(0, \ell)$. By analyticity and continuity, the formula $g_t(\phi_t(x)) = g_0(x)$ holds for every $t \in [T, T')$ and $x \in [0, \ell_t]$. This contradicts the definition of T , and so $T = +\infty$. This concludes the proof of point (1) of the lemma.

3. For the statement (2) of the lemma, let us show first that $\langle \mu_t, m^2 \rangle$ is bounded on $[0, T_0)$, for every $T_0 \in [0, T_{\text{gel}})$. Let T' , the smallest time when this fails (provided of course that $T_{\text{gel}} > 0$). By Lemma 2.6, $T' > 0$. Differentiating (3.19) with respect to x and having x tend to $\ell_t = 1$ gives, for $t < T'$,

$$g'_t(1) = \langle \mu_t, m^2 \rangle = \frac{1}{1 - tK}.$$

This quantity explodes only when $t = T_{\text{gel}} = 1/K$, so $T' = T_{\text{gel}}$.

4. The boundedness of $(\langle \mu_t, m^2 \rangle)_{t \in [0, T_0)}$ just proved for all $T_0 \in [0, T_{\text{gel}})$ and Lemma 2.6 imply that for $t \in [0, T_{\text{gel}})$, $M_t = M_0$. By the definition (3.10) of ϕ_t , it follows that $\phi_t(1) = 1$ for $t \in [0, T_{\text{gel}})$. But ϕ_t is increasing, so $\ell_t = 1$ for $t \in [0, T_{\text{gel}})$. Assume now that for some $t > T_{\text{gel}}$, $\ell_t = 1$. Then (3.19) holds on $[0, 1]$, and this is impossible since the right term is an increasing function of x , whereas the left one decreases in a left neighborhood of 1 since $\phi'_t(1) < 0$. The fact that $\phi_t(\ell_t) = 1$ follows then directly from the definition of ℓ_t and the continuity of $\phi_t(\cdot)$. Finally, the inequality $\ell_t > 0$ is obvious since $\phi_t(0) = 0$, and computing (3.19) at $x = \ell_t$ gives (3.20). This concludes the proof of (2).

5. We know that $\ell_t = 1$ and $M_t = M_0$ for all $t < T_{\text{gel}}$. Now, g_0 is strictly increasing and continuous. Since $(M_t)_{t \geq 0}$ is monotone non-increasing and right-continuous by Lemma 2.5, so is $(\ell_t)_{t \geq 0}$ by (3.20). To get left-continuity of $(\ell_t)_{t > T_{\text{gel}}}$, consider $t > T_{\text{gel}}$, and let ℓ be the left limit of ℓ_s at t . We have $\ell \leq \ell_{t+(t-T_{\text{gel}})/2} < 1$, so by the continuity property (c1) above,

$$1 = \phi_s(\ell_s) \xrightarrow{s \rightarrow t^-} \phi_t(\ell).$$

Hence $\phi_t(\ell) = 1$. Assume $\ell > \ell_t$ (that is, ℓ is the second point where ϕ_t reaches 1). Take $x \in (\ell_t, \ell)$. By property (b5), $\phi_t(x) > 1$. But on the other hand, $x < \ell \leq \ell_s$ for $s < t$, so $\phi_s(x) \leq 1$, and so $\phi_t(x) \leq 1$, and this is a contradiction. So $\ell = \ell_t$ and $(\ell_t)_{t \geq 0}$ is indeed continuous. This concludes the proof of (3) and of the Lemma. \square

Finally, we will see that for $t > T_{\text{gel}}$, $\ell_t = m_t$, so that ϕ_t increases from 0 to 1, which is its maximum, and then decreases. To this end, recall that $(\ell_t)_{t \geq 0}$ is monotone non-increasing and that $(\ell_t)_{t \geq 0}$ and $(\phi_t)_{t \geq 0}$ are continuous, so the chain rule for Stieltjes integrals and (3.17) give

$$\begin{aligned} 1 = \phi_t(\ell_t) &= \phi_0(\ell_0) + \int_0^t \phi'_s(\ell_s) d\ell_s + \int_0^t \frac{\partial^+ \phi_s}{\partial s}(\ell_s) ds \\ &= 1 + \int_0^t \phi'_s(\ell_s) d\ell_s + \int_0^t \phi_s(\ell_s)(M_s - g_0(\ell_s)) ds \end{aligned}$$

that is, with (3.20),

$$\phi'_t(\ell_t) d\ell_t = 0. \quad (3.21)$$

Hence, $d\ell_t$ -a.e. $\phi'_t(\ell_t) = 0$, i.e. $\ell_t = m_t$. This is actually true for all $t > T_{\text{gel}}$, as we shall now prove. This result also has its counterpart in the model with limited aggregations, namely Part 3 of the proof of Theorem 4.3.

Lemma 2.12. *For every $t > T_{\text{gel}}$, $\phi'_t(\ell_t) = 0$, i.e. $\ell_t = m_t$, the point where ϕ_t attains its maximum. In particular,*

$$\ell_t g'_0(\ell_t) = \frac{1}{t}, \quad \forall t > T_{\text{gel}}. \quad (3.22)$$

Proof. First, recall that ϕ_t is increasing on $[0, \ell_t]$, so that $\phi'_t(\ell_t) \geq 0$, that is

$$\ell_t g'_0(\ell_t) \leq \frac{1}{t}. \quad (3.23)$$

Assume now that there is a $t > T_{\text{gel}}$ such that $\phi'_t(\ell_t) > 0$, and consider

$$s = \sup\{r \in (T_{\text{gel}}, t) : \phi'_r(\ell_r) = 0\}.$$

As noted before, $t \mapsto \ell_t$ is strictly decreasing for $t > T_{\text{gel}}$ for any $t > T_{\text{gel}}$, so $d\ell_t([T_{\text{gel}}, T_{\text{gel}} + \varepsilon]) > 0$ for all $\varepsilon > 0$. Hence there are points $r < t$ where $\phi'_r(\ell_r) = 0$, and thus the definition of s does make sense.

Take now (r_n) a sequence of points such that $T < r_n < t$, $\phi'_{r_n}(\ell_{r_n}) = 0$ and (r_n) converges to s . Since $0 < \ell_s < 1$, by property (c2) above, we get

$$0 = \phi'_{r_n}(\ell_{r_n}) \rightarrow \phi'_s(\ell_s)$$

so that $\phi'_s(\ell_s) = 0$. This shows that $s < t$, and that for $r \in (s, t)$, $\phi'_r(\ell_r) > 0$. Hence, by continuity of $(\ell_r)_{r \geq 0}$ and by (3.21), $(\ell_r)_{r \in [s, t]}$ is constant. This gives

$$\frac{1}{s} = \ell_s g'_0(\ell_s) = \ell_t g'_0(\ell_t) \leq \frac{1}{t}$$

which is a contradiction since $s < t$. In particular, $\phi'_t(\ell_t) = 0$ implies (3.22). \square

We can now end the proof of Proposition 2.8. By Lemma 2.12, necessarily $M_t = M_0$ on $[0, T_{\text{gel}}]$ and for $t > T_{\text{gel}}$, $M_t := g_t(1) = g_0(\ell_t)$, where

$$\ell_t g'_0(\ell_t) = \frac{1}{t}. \quad (3.24)$$

Since $x \mapsto x g'_0(x)$ is strictly increasing from $[0, 1]$ to $[0, K]$, where $K = \langle \mu_0, m^2 \rangle = 1/T_{\text{gel}}$, this equation has a unique solution for $t > T_{\text{gel}}$. Hence M_t is uniquely defined. Therefore α_t and ϕ_t are uniquely determined by g_0 , so we can define ϕ_t as in (3.10), and Lemma

2.11 shows that $g_t(\phi_t(x)) = g_0(x)$ for $x \in [0, \ell_t]$, and that ϕ_t is a bijection from $[0, \ell_t]$ to $[0, 1]$. So it has a right inverse h_t , and compounding by h_t in the previous formula gives

$$g_t(x) = g_0(h_t(x)) \quad (3.25)$$

for all $x \in [0, 1]$, $t \geq 0$. Thus g_t can be expressed by a formula involving only g_0 and in particular, (μ_t) depends only on μ_0 . This shows the uniqueness of a solution to Smoluchowski's equation (3.3).

2.5 Behavior of the moments

In this paragraph, we will study the behavior of the first and second moment of $(\mu_t)_{t \geq 0}$ as time passes, showing how to prove rigorously and recover the results of [13]. For more general coagulation rates, one can obtain upper bounds of the same nature, see [35].

First consider the mass $M_t = \langle \mu_t, m \rangle$. We will always assume that $T_{\text{gel}} < +\infty$. Let us start with the following lemma.

Lemma 2.13. *Let $\nu \in \mathcal{M}_c^+$ be a measure which integrates $x \mapsto y^x$ for small enough $y > 0$. Let m_0 be the infimum of its support. Then*

$$\lim_{y \rightarrow 0^+} \frac{\langle \nu, xy^x \rangle}{\langle \nu, y^x \rangle} = m_0.$$

Proof. First, note that $xy^x \geq m_0 y^x$ ν -a.e. so

$$\liminf_{y \rightarrow 0} \frac{\langle \nu, xy^x \rangle}{\langle \nu, y^x \rangle} \geq m_0.$$

Let us prove now that

$$\limsup_{y \rightarrow 0} \frac{\langle \nu, xy^x \rangle}{\langle \nu, y^x \rangle} \leq m_0.$$

Assume this is not true. Then, up to extraction of a subsequence, we may assume that there exists $\alpha > 0$ such that for arbitrary small $y \in (0, 1)$, $\langle \nu, xy^x \rangle \geq (m_0 + \alpha) \langle \nu, y^x \rangle$. Hence $\langle \nu, (x - m_0 - \alpha)y^x \rangle \geq 0$, so

$$\langle \nu, (x - m_0 - \alpha)y^x \mathbf{1}_{\{x > m_0 + \alpha\}} \rangle \geq \langle \nu, (m_0 + \alpha - x)y^x \mathbf{1}_{\{m_0 \leq x \leq m_0 + \alpha\}} \rangle. \quad (3.26)$$

But

$$\begin{aligned} & \langle \nu, (m_0 + \alpha - x)y^x \mathbf{1}_{\{m_0 \leq x \leq m_0 + \alpha\}} \rangle \\ & \geq \langle \nu, (m_0 + \alpha - x)y^x \mathbf{1}_{\{m_0 \leq x \leq m_0 + \alpha/2\}} \rangle \\ & \geq \langle \nu, (m_0 + \alpha - x) \mathbf{1}_{\{m_0 \leq x \leq m_0 + \alpha/2\}} \rangle y^{m_0 + \alpha/2} \end{aligned}$$

and

$$\langle \nu, (x - m_0 - \alpha)y^x \mathbf{1}_{\{x > m_0 + \alpha\}} \rangle \leq \langle \nu, (x - m_0 - \alpha) \mathbf{1}_{\{x > m_0 + \alpha\}} \rangle y^{m_0 + \alpha}.$$

With (3.26), this shows that

$$\langle \nu, (x - m_0 - \alpha) \mathbf{1}_{\{x > m_0 + \alpha\}} \rangle y^{\alpha/2} \geq \langle \nu, (m_0 + \alpha - x) \mathbf{1}_{\{m_0 \leq x \leq m_0 + \alpha/2\}} \rangle$$

and having y tend to zero gives

$$0 \geq \langle \nu, (m_0 + \alpha - x) \mathbf{1}_{\{m_0 \leq x \leq m_0 + \alpha/2\}} \rangle$$

which is a contradiction since $\nu([m_0, m_0 + \alpha/2]) > 0$. \square

Corollary 2.14. *The mass of the system is continuous and positive. It is strictly decreasing on $[T_{\text{gel}}, +\infty)$. Moreover, denote $m_0 = \inf \text{supp } \mu_0$. Then*

$$\lim_{t \rightarrow +\infty} \frac{1}{tM_t} = m_0.$$

Proof. Recall that $M_t = g_0(\ell_t)$ so the first properties follow from Lemma 2.11. Denote now $\nu(dm) = m\mu_0(dm)$. For $t > T_{\text{gel}}$, $\ell_t g'_0(\ell_t) = 1/t$, so

$$\frac{1}{tM_t} = \frac{\langle \nu, x\ell_t^x \rangle}{\langle \nu, \ell_t^x \rangle}$$

and since $\ell_t \rightarrow 0$ when $t \rightarrow +\infty$, this tends to m_0 by Lemma 2.13. \square

We can also study the behavior of the mass for small times. Recall that before gelation, the mass is constant at 1. We have seen that it is continuous at the gelation time. We may then wonder if its derivative is continuous, that is if $\dot{M}_{T_{\text{gel}}+}$ is zero or not.

Lemma 2.15. *The right derivative of M at T_{gel} is given by*

$$\dot{M}_{T_{\text{gel}}+} = - \lim_{x \rightarrow 1^-} \frac{g'_0(x)^3}{g'_0(x) + xg''_0(x)} \in [-\infty, 0]$$

provided the limit exists.

Proof of Lemma 2.15. For $t > T_{\text{gel}}$, $f(\ell_t) = 1/t$ with $f(x) = xg'_0(x)$, and $0 < \ell_t < 1$. But $f'(\ell_t) \neq 0$, so by the inverse mapping theorem, $(\ell_t)_{t \geq 0}$ is differentiable and

$$\dot{\ell}_t = -\frac{1}{t^2 f'(\ell_t)}.$$

Using the fact that $M_t = g_0(\ell_t)$, it is then easy to see that

$$\dot{M}_t = -\ell_t^2 \frac{g'_0(\ell_t)^3}{g'_0(\ell_t) + \ell_t g''_0(\ell_t)}.$$

Since $(\ell_t)_{t \geq 0}$ is continuous at T_{gel} and $\ell_{T_{\text{gel}}} = 1$, the result follows. \square

Recall that the gelation time is precisely the first time when the second moment $\langle \mu_t, m^2 \rangle$ of $(\mu_t)_{t \geq 0}$ becomes infinite. It actually remains infinite afterwards.

Corollary 2.16. *For all $t \geq T_{\text{gel}}$, $\langle \mu_t, m^2 \rangle = +\infty$.*

Proof. Note that

$$\langle \mu_t, m^2 \rangle = g'_t(1),$$

this formula being understood as a monotone limit. By (3.19), for $x < \ell_t$

$$\phi'_t(x) g'_t(\phi_t(x)) = g'_0(x).$$

When $x \rightarrow \ell_t^-$, $\phi'_t(x) \rightarrow 0$ by Lemma 2.12, and $g'_0(x) \rightarrow g'_0(\ell_t) \neq 0$ since $\ell_t > 0$. So

$$g'_t(\phi_t(\ell_t)) = g'_t(1) = +\infty.$$

\square

2.6 Existence of solutions

Existence of solutions of (3.3) is a well-known topic, see e.g. [19]. However, the case $M_0 = +\infty$ is apparently new, so that we give a short proof for the general case based on previous papers, mainly [55].

Let now $\mu_0 \in \mathcal{M}_f^+$ be as in the statement of Theorem 2.3 and let us set g_0 as in (3.5), ℓ_t and M_t as in point (1) of Proposition 2.8, α_t and ϕ_t as in (3.11) and (3.10). Then it is easy to see that ϕ_t admits a right inverse h_t satisfying (3.14), and we can thus define

$$g_t(x) := g_0(h_t(x)), \quad t \geq 0, \quad x \in [0, 1].$$

It is an easy but tedious task to check that g_t satisfies (3.29) and all properties (a1)-(a4) above. In particular, if $g_0(1) = +\infty$ then $h_t(1) < 1$ and therefore $g_t(1) < +\infty$ for all $t > 0$. Following [55], we can now prove the following.

Proposition 2.17. *For all $t > 0$ there exists $\mu_t \in \mathcal{M}_f^+$ such that*

$$g_t(x) = \langle \mu_t, mx^m \rangle = \int_{(0, +\infty)} mx^m \mu_t(dm), \quad x \in [0, 1].$$

Proof. Let $t > 0$ be fixed. We set for all $y \geq 0$

$$\Phi(y) := g_0(e^{-y}), \quad \Gamma(y) := tg_0(e^{-y}), \quad G(y) := \Gamma(y) + y - \log \alpha_t = -\log \phi_t(e^{-y}).$$

We recall that $f : [0, +\infty) \mapsto [0, +\infty)$ is completely monotone if f is continuous on $[0, +\infty)$, infinitely many times differentiable on $(0, +\infty)$ and

$$(-1)^k \frac{d^k f}{dy^k}(y) \geq 0, \quad \forall k \geq 0, \quad y \in (0, +\infty).$$

It is easy to see that Φ and Γ are completely monotone. Moreover, G has a right inverse

$$G^{-1} : [0, +\infty) \mapsto [\log(1/\ell_t), +\infty), \quad G^{-1}(y) = -\log h_t(e^{-y}), \quad y \geq 0,$$

and therefore by the definitions

$$g_0(h_t(e^{-y})) = \Phi(G^{-1}(y)), \quad y \geq 0.$$

By [55, Thm. 3.2], $\Phi \circ G^{-1}$ is completely monotone and therefore, by Bernstein's Theorem, there exists a unique $\nu_t \in \mathcal{M}_f^+$ such that

$$g_t(e^{-y}) = g_0(h_t(e^{-y})) = \Phi(G^{-1}(y)) = \int_{(0, +\infty)} e^{-ym} \nu_t(dm), \quad y \geq 0.$$

Since $g_t(1) < +\infty$ for all $t > 0$, we obtain that $\langle \nu_t, m \rangle < +\infty$, so that we can set $\mu_t(dm) := m \nu_t(dm)$, and we have found that there is a unique $\mu_t \in \mathcal{M}_f^+$ such that

$$g_t(x) = g_0(h_t(x)) = \int_{(0, +\infty)} x^m m \mu_t(dm), \quad x \in (0, 1].$$

□

In order to show that $(\mu_t)_{t \geq 0}$ is a solution of Smoluchowski's equation in the sense of Definition 2.1, we have to check that $\int_0^\varepsilon M_t^2 dt < +\infty$ for all $\varepsilon > 0$. This is the content of the next result.

Lemma 2.18. *If $(\mu_t)_{t \geq 0}$ is the family constructed in Proposition 2.17, then for all $\varepsilon > 0$, $\int_0^\varepsilon \langle \mu_s, m \rangle^2 ds < +\infty$.*

Proof. If $M_0 < +\infty$ then there is nothing to prove, since $(M_t)_{t \geq 0}$ is monotone non-increasing, so let us consider the case $M_0 = +\infty$ and thus $T_{\text{gel}} = 0$. Since $M_t = g_0(\ell_t)$ is bounded and continuous for $t \in [\delta, \varepsilon]$ for all $\delta \in (0, \varepsilon)$, we have, by (3.12) and (3.5),

$$\begin{aligned} \int_\delta^\varepsilon M_t^2 dt &= \int_\delta^\varepsilon g_0^2(\ell_t) dt = \varepsilon g_0^2(\ell_\varepsilon) - \delta g_0^2(\ell_\delta) - \int_\delta^\varepsilon 2tg_0(\ell_t)g_0'(\ell_t) d\ell_t \\ &\leq \varepsilon g_0^2(\ell_\varepsilon) - \int_\delta^\varepsilon 2g_0(\ell_t) \frac{d\ell_t}{\ell_t} = \varepsilon g_0(\ell_\varepsilon) + 2 \int_{\ell_\varepsilon}^{\ell_\delta} g_0(y) \frac{dy}{y} \\ &\leq \varepsilon g_0(\ell_\varepsilon) + \frac{2}{\ell_\varepsilon} \langle \mu_0, \frac{m}{1+m} \rangle \leq \varepsilon g_0(\ell_\varepsilon) + \frac{2}{\ell_\varepsilon} \langle \mu_0, m \wedge 1 \rangle. \end{aligned}$$

Letting $\delta \downarrow 0$, by (3.4) we obtain the desired result. \square

We now finish the proof of existence of a solution by showing that $(\mu_t)_{t \geq 0}$ indeed solves (3.3). By choosing $x = e^{-y}$, $y \geq 0$, in (3.29), we find an equality between Laplace transforms. Since the Laplace transform is one-to-one, then we obtain (3.3).

Remark 2.19. In the proof of uniqueness, we may only require that $\langle \mu_0, my^m \rangle < +\infty$ for every $y \in [0, 1)$. However, the same kind of computation as in Lemma 2.18 shows that if this the case, but $\langle \mu_0, m \wedge 1 \rangle = +\infty$, then $\int_0^t M_s^2 ds = +\infty$ for all $t > 0$, in contradiction with Definition 2.1 of a solution.

3 Flory's equation

3.1 Introduction and results

We will now consider the modified version of Smoluchowski's equation, also known as *Flory's equation*, with a multiplicative kernel.

Definition 3.1. Let $\mu_0 \in \mathcal{M}_c^+$. We say that a family $(\mu_t)_{t \geq 0} \subset \mathcal{M}_c^+$ solves Flory's equation (3.3) if

- for every $t > 0$, $\int_0^t \langle \mu_s(dm), m \rangle^2 ds < +\infty$,
- for all $\phi \in C_c(\mathbb{R}^+)$ and $t > 0$

$$\begin{aligned} \langle \mu_t, \phi \rangle &= \langle \mu_0, \phi \rangle + \frac{1}{2} \int_0^t \langle \mu_s(dm) \mu_s(dm'), mm' \Delta \phi(m, m') \rangle ds \\ &\quad - \int_0^t \langle \mu_s, \phi \rangle \langle \mu_0(dm) - \mu_s(dm), m \rangle ds. \end{aligned} \tag{3.27}$$

In equation (3.27), the mass that vanishes in the gel interacts with the other clusters. It is a modified Smoluchowski's equation, where a term has been added, representing the interaction of the clusters of mass m with the gel, whose mass is

$$\langle \mu_0 - \mu_s, m \rangle$$

i.e. precisely the missing mass of the system. Notice that in this case the equation makes sense only if $\langle \mu_0, m \rangle < +\infty$.

The mass is expected to decrease faster in this case than for (3.3). This is actually true, as we can see in the following result. We take here as initial condition a non-null measure $\mu_0 \in \mathcal{M}_c^+$ such that $\langle \mu_0, m \rangle < +\infty$, and define $T_{\text{gel}} \in [0, +\infty)$ as in Definition 2.2.

Theorem 3.2. *Flory's equation (3.27) has a unique solution (μ_t) on \mathbb{R}^+ . It has the following properties.*

1. *The mass is given by $M_t = g_0(l_t)$, where $l_t = 1$ for $t \leq T_{\text{gel}}$ and, for $t > T_{\text{gel}}$, l_t is uniquely defined by*

$$l_t = e^{-t(M_0 - g_0(l_t))}, \quad l_t \in [0, 1].$$

In particular, $t \mapsto M_t$ is continuous on $[0, +\infty)$, constant on $[0, T_{\text{gel}}]$, strictly decreasing on $[T_{\text{gel}}, +\infty)$ and analytic on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$.

2. *The function $\phi_t(x) = xe^{t(M_0 - g_0(x))}$ has a right inverse $h_t : [0, 1] \rightarrow [0, l_t]$. The generating function $g_t(x) = \langle \mu_t, mx^m \rangle$ is given for $t \geq 0$ by*

$$g_t(x) = g_0(h_t(x)). \quad (3.28)$$

3. *Let $m_0 = \inf \text{supp } \mu_0 \geq 0$. Then, when $t \rightarrow +\infty$,*

$$M_t e^{m_0 t} \rightarrow m_0 \mu_0(\{m_0\})$$

and for every $\varepsilon > 0$

$$M_t e^{(m_0 + \varepsilon)t} \rightarrow +\infty.$$

4. *The second moment $\langle \mu_t, m^2 \rangle$ is finite on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$ and infinite at T_{gel} .*

Remark 3.3. • Norris [46, Thm 2.8] has a proof of global uniqueness of Flory's equation (3.27) for slightly less general initial conditions (μ_0 such that $\langle \mu_0, 1 + m \rangle < +\infty$), but for a much more general model.

- When $m_0 > 0$, it was already observed (Proposition 5.3 in [14]) that the mass decays (at least) exponentially fast (see also [12, 53, 59]).
- The mass in Flory's equation may decrease slower if $\inf \text{supp } \mu_0 = 0$. For instance, if $\mu_0(dm) = e^{-m} dm$, then $M_t \sim t^{-2}$.

Corollary 3.4. *Let $\mu_0 \in \mathcal{M}_c^+$ such that $\langle \mu_0, m \rangle < +\infty$ and let $(\mu_t^S)_{t \geq 0}$ and $(\mu_t^F)_{t \geq 0}$ the solutions of (3.3), respectively, (3.27). Then*

- $\mu_t^S \equiv \mu_t^F$ for all $t \leq T_{\text{gel}} := 1/\langle \mu_0, m^2 \rangle$;
- $\langle \mu_t^F, m \rangle < \langle \mu_t^S, m \rangle$ for all $t > T_{\text{gel}}$.

As anticipated, the mass decreases faster in Flory's case than for Smoluchowski's equation. In particular, in Flory's case $\langle \mu_t, m^2 \rangle$ becomes finite immediately after gelation, the mass remaining however continuous (we can think that the big clusters, which have the biggest influence on this second moment, disappear into the gel). Moreover, if $\inf \text{supp } \mu_0 > 0$ then the mass decays exponentially fast, which is to be compared with the slower decrease in $1/t$ in Smoluchowski's equation.

3.2 Proof of Theorem 3.2

The proof is very similar to (and actually easier than) that of Theorem 2.3.

1. Arguing as in the proof of Lemma 2.5, we obtain easily that $(M_t)_{t \geq 0}$ is monotone non-increasing and right-continuous. As in Lemma 2.6, if $\langle \mu_0, m^2 \rangle < +\infty$, then $t \mapsto \langle \mu_t, m^2 \rangle$ is bounded on some interval $[0, T_0]$ and $M_t = M_0$ for $t \in [0, T_0]$. Therefore (μ_t) is a solution of Smoluchowski's equation (3.3) on $[0, T_0]$.

2. Consider initial concentrations μ_0 as in the statement, a solution $(\mu_t)_{t \geq 0}$ to Flory's equation and $g_t(x)$, $x \in [0, 1]$, generating function of $m \mu_t(dm)$. Then g_t solves the PDE

$$\frac{\partial g_t}{\partial t} = x(g_t - M_0) \frac{\partial g_t}{\partial x}, \quad \forall t > 0, x \in [0, 1], \quad (3.29)$$

the same as the one obtained for Smoluchowski's equation before gelation. It may be solved using the method of characteristics. Indeed, the mapping

$$\phi_t(x) = x e^{t(M_0 - g_0(x))} = x + \int_0^t (M_0 - g_0(x)) \phi_s(x) ds, \quad (3.30)$$

has the following properties.

- (d1) $\phi_t(0) = 0$, $\phi_t(1) = 1$.
- (d2) For all $t \geq 0$, $\phi_t'(x) = e^{t(M_0 - g_0(x))} (1 - t x g_0'(x))$.
- (d3) For $t \leq T_{\text{gel}}$, $\phi_t(\cdot)$ is increasing; therefore, $\phi_t(x) \in [0, 1]$ for all $x \in [0, 1]$ and $\phi_t(x) = 1$ if and only if $x = 1$.
- (d4) For $t > T_{\text{gel}}$, $\phi_t(\cdot)$ is increasing on $[0, m_t]$ and decreasing on $[m_t, 1]$, where m_t is the unique $x \in (0, 1)$ such that $\phi_t'(x) = 0$, i.e. such that $t x g_0'(x) = 1$.
- (d5) For $t > T_{\text{gel}}$, $\phi_t(m_t) > 1$, since $\phi_t(1) = 1$ and $\phi_t'(1) < 0$. Therefore there is a unique $l_t \in (0, m_t)$ such that $\phi_t(l_t) = 1$.
- (d6) For $t > T_{\text{gel}}$, $\phi_t'(l_t) \neq 0$, since $l_t < m_t$.

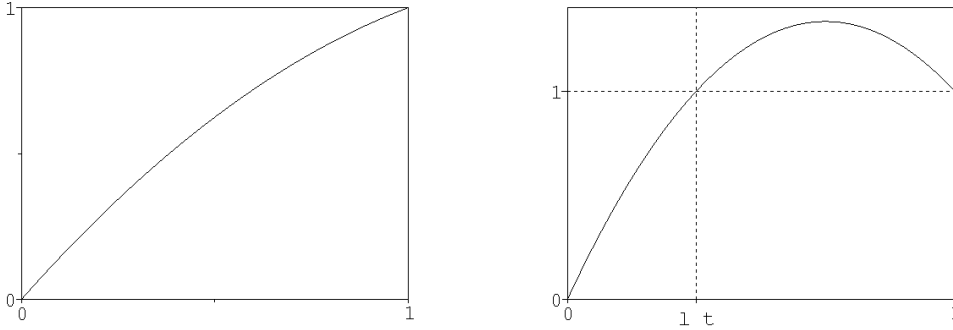


Figure 3.2: ϕ_t before and after gelation.

Setting $l_t := 1$ for $t \leq T_{\text{gel}}$, ϕ_t is thus a continuous bijection from $[0, l_t]$ to $[0, 1]$, with continuous inverse function $h_t : [0, 1] \mapsto [0, l_t]$. By using (3.29) and (3.30) and arguing as in part (i) and (ii) of the proof of Lemma 2.11, we can see that the function $u_t(x) := g_t(\phi_t(x)) - g_0(x)$ satisfies $u_t(x) = u_0(x) = 0$ for all $t \geq 0$ and $x \in [0, l_t]$. Therefore the only solution of the PDE (3.29) is given by

$$g_t(x) = g_0(h_t(x)), \quad t \geq 0, x \in [0, 1]. \quad (3.31)$$

Flory's equation has thus a unique solution on \mathbb{R}^+ , and its generating function is g_t .

3. We have seen in (d5) above that, for $t > T_{\text{gel}}$, there is a unique $l_t \in [0, 1]$ such that $\phi_t(l_t) = 1$. The relation $\phi_t(l_t) = 1$ with $l_t \in [0, 1]$ is equivalent to $l_t = e^{-t(M_0 - g_0(l_t))}$

with $l_t \in [0, 1)$. This relation implies that $t \mapsto l_t$ is analytic for $t > T_{\text{gel}}$. A differentiation shows that

$$\frac{dl_t}{dt} = -\frac{(M_0 - g_0(l_t))l_t}{1 - tg'_0(l_t)l_t} < 0, \quad t > T_{\text{gel}},$$

since $g'_0(l_t)l_t < g'_0(m_t)m_t = 1/t$ and $g_0(l_t) < g_0(1) = M_0$. Let ℓ be the limit of l_t as $t \downarrow T_{\text{gel}}$: then we obtain $\ell = e^{-T_{\text{gel}}(M_0 - g_0(\ell))}$, i.e. $\phi_{T_{\text{gel}}}(\ell) = 1$. By (d3) above, this is equivalent to $\ell = 1$.

4. Since $M_t = g_t(1) = g_0(h_t(1)) = g_0(l_t)$, the properties of $t \mapsto M_t = g_0(l_t)$ follow from those of $t \mapsto l_t$. Recall now that $\phi_t(l_t) = 1$, that is

$$\log(l_t) = t(g_0(l_t) - 1). \quad (3.32)$$

If the limit l of l_t as $t \rightarrow +\infty$ were nonzero, then passing to the limit in this equality would give $\log(l) = -\infty$. So $l = 0$ and

$$\log l_t \sim -t. \quad (3.33)$$

- Assume $m > 0$. Now, obviously $g_0(x) \leq x^m$, so

$$\log(tg_0(l_t)) = \log l_t + \log g_0(l_t) \leq \log t + m \log l_t \rightarrow -\infty.$$

Hence $tg_0(l_t) \rightarrow 0$ and (3.32) yields $\log l_t + t \rightarrow 0$. Hence $l_t^m \sim e^{-mt}$. Finally

$$\lim_{t \rightarrow +\infty} M_t e^{mt} = \lim_{t \rightarrow +\infty} \frac{g_0(l_t)}{l_t^m} = m\mu_0(\{m\})$$

since by dominated convergence, $g_0(x)x^{-m} \rightarrow m\mu_0(\{m\})$ when $x \rightarrow 0$. Now, by monotone convergence, if $m' > m$, then $g_0(x)x^{-m'} \rightarrow +\infty$ when x tends to 0, whence

$$\lim_{t \rightarrow +\infty} M_t e^{m't} = \lim_{t \rightarrow +\infty} \frac{g_0(l_t)}{l_t^{m'}} = +\infty.$$

- Assume now $m = 0$ and let $\varepsilon > 0$. By monotone convergence $g_0(x)x^{-\varepsilon} \rightarrow +\infty$ as $x \downarrow 0$, so using (3.33) we see that $g(l_t)e^{-\varepsilon t} \rightarrow +\infty$ as $t \uparrow +\infty$, which is the desired result.

5. Finally, (3.31) gives for $x < 1$ and $t > T_{\text{gel}}$

$$g'_t(x) = g'_0(h_t(x)) h'_t(x) = \frac{g'_0(h_t(x))}{\phi'_t(h_t(x))}.$$

When $x \uparrow 1$, $h_t(x) \uparrow l_t < 1$, and $\phi'_t(h_t(x)) \rightarrow \phi'_t(l_t) \neq 0$ by (d6) above. So $\langle \mu_t, m^2 \rangle = g'_t(1) < +\infty$.

6. Existence of a solution of (3.27) follows arguing as in Section 2.6.

3.3 Proof of Corollary 3.4

For all $t \leq T_{\text{gel}}$, $\langle \mu_t^F, m \rangle = \langle \mu_0^F, m \rangle$ and therefore μ_t^F solves (3.3), so that by uniqueness of Smoluchowski's equation we have that $\mu_t^S = \mu_t^F$. For $t > T_{\text{gel}}$ we have that $\langle \mu_t^F, m \rangle = g_0(l_t)$ while $\langle \mu_t^S, m \rangle = g_0(\ell_t)$, where l_t and ℓ_t are defined respectively by

$$l_t = e^{-t(M_0 - g_0(l_t))}, \quad l_t \in [0, 1)$$

and

$$\ell_t g_0'(\ell_t) = \frac{1}{t}.$$

In points (d4) and (d5) of the proof of Theorem 3.2, we have shown that $l_t < m_t$ where $t m_t g_0'(m_t) = 1$, so that $m_t = \ell_t < l_t$. Hence $\langle \mu_t^F, m \rangle = g_0(\ell_t) < g_0(l_t) = \langle \mu_t^S, m \rangle$.

4 The model with limited aggregations

We now turn to our main interest, namely Equation (3.2). We apply the same techniques as above in a slightly more complicated setting. After giving all details in Smoluchowski's case, we will give a shorter proof and focus on the differences with the proof of Theorem 2.3. As above, we can transform the system (3.2) into a non-local PDE problem, which we are able to solve, thus obtaining existence and uniqueness to (3.2). More precisely, we consider the following system.

Definition 4.1. Let $c_0(a, m) \geq 0$, $a \in \mathbb{N}$, $m \in \mathbb{N}^*$. We say that a family $(c_t(a, m))$, $t \geq 0$, $a \in \mathbb{N}$, $m \in \mathbb{N}^*$, is a solution of Smoluchowski's equation (3.34) if

- for every $t > 0$, $\int_0^t \langle c_s, a \rangle^2 ds < +\infty$,
- for all $a \in \mathbb{N}$, $m \in \mathbb{N}^*$ and $t > 0$,

$$\begin{aligned} c_t(a, m) = & c_0(a, m) + \\ & + \int_0^t \frac{1}{2} \sum_{a'=1}^{a+1} \sum_{m'=1}^{m-1} a'(a+2-a') c_s(a', m') c_s(a+2-a', m-m') ds \\ & - \int_0^t \sum_{a' \geq 1} \sum_{m' \geq 1} a a' c_s(a, m) c_s(a', m') ds. \end{aligned} \quad (3.34)$$

Because of the interpretation of a as a variable counting the number of arms a cluster possesses, it is more natural to state (3.34) in the discrete setting, as in [3]. In particular, since at each coagulation two arms are removed from the system, a non-integer initial number of arms would lead to an ill-defined dynamics. One could however with no difficulty consider an initial distribution of masses on $(0, +\infty)$.

As usual, we define for $c, f : \mathbb{N} \times \mathbb{N}^* \rightarrow \mathbb{R}^+$,

$$\langle c, f \rangle = \sum_{(a, m) \in \mathbb{N} \times \mathbb{N}^*} c(a, m) f(a, m)$$

writing a for the function $(a, m) \mapsto a$, m for $(a, m) \mapsto m$ and so on.

It is easy to see that (c_t) is a solution to this equation if and only if the function

$$k_t(x, y) := \langle c_t, a x^{a-1} y^m \rangle = \sum_{a=1}^{+\infty} \sum_{m=1}^{+\infty} a c_t(a, m) x^{a-1} y^m, \quad (3.35)$$

defined for $t \geq 0$, $y \in [0, 1]$ and $x \in [0, 1)$, satisfies

$$\begin{cases} k_t(x, y) = k_0(x, y) + \int_0^t \left[(k_s(x, y) - xA_s) \frac{\partial k_s}{\partial x}(x, y) - A_s k_s(x, y) \right] ds, \\ A_t := k_t(1, 1) = \langle c_t, a \rangle. \end{cases} \quad (3.36)$$

We may solve this PDE with the same techniques as above and obtain the following result. We consider below initial concentrations $c_0(a, m) \geq 0$, $a \in \mathbb{N}$, $m \in \mathbb{N}^*$ such that $\langle c_0, 1 \rangle < +\infty$, and we may define a similar but slightly more general gelation time as in [3].

Definition 4.2. Let $A_0 := \langle c_0, a \rangle \in (0, +\infty]$ and $K := \langle c_0, a(a-1) \rangle \in [0, +\infty]$. Then $K = +\infty$ whenever $A_0 = +\infty$, and we may let

$$T_{\text{gel}} = \begin{cases} \frac{1}{K-A_0} & \text{if } A_0 < K < +\infty \\ 0 & \text{if } K = +\infty \\ +\infty & \text{if } K \leq A_0 < +\infty. \end{cases} \quad (3.37)$$

We can then state our main result.

Theorem 4.3. Equation (3.34) has a unique solution (c_t) defined on \mathbb{R}^+ . When $T_{\text{gel}} < +\infty$, this solution enjoys the following properties.

1. The number of arms $A_t := \langle c_t, a \rangle$ is continuous, strictly decreasing, and, for all $t > 0$,

$$A_t \leq \frac{A_0}{1 + tA_0} \quad \text{if } A_0 < +\infty, \quad A_t \leq \frac{1}{t} \quad \text{if } A_0 = +\infty. \quad (3.38)$$

If we set

$$\alpha_t = \exp\left(\int_0^t A_s ds\right),$$

then α_t is given by

$$\alpha_t = 1 + A_0 t \quad \text{for } t < T_{\text{gel}}$$

and, for $t \geq T_{\text{gel}}$,

$$\alpha_t = \begin{cases} \Gamma^{-1}(1 + A_0 T_{\text{gel}} + t - T_{\text{gel}}) & \text{if } A_0 < +\infty \\ \Gamma^{-1}(1 + t) & \text{if } A_0 = +\infty \end{cases} \quad (3.39)$$

where

$$\Gamma(x) = 1 + A_0 T_{\text{gel}} + \int_{1+A_0 T_{\text{gel}}}^x \frac{dr}{k_0(H(1/r), 1)}, \quad x \geq 1 + A_0 T_{\text{gel}},$$

and $H : [G(0), G(1)) \rightarrow [0, 1)$ is the right-inverse of the increasing function

$$G(x) := x - \frac{k_0(x, 1)}{k'_0(x, 1)}, \quad x \in [0, 1). \quad (3.40)$$

2. Let

$$\beta_t = \int_0^t \frac{1}{\alpha_s^2} ds$$

and consider

$$\phi_t(x, y) := \alpha_t(x - \beta_t k_0(x, y)), \quad t \geq 0, x, y \in [0, 1].$$

Then

- $\phi_t(\cdot, 1)$ attains its maximum at a point ℓ_t such that $\phi_t(\ell_t, 1) = 1$. For $t \leq T_{\text{gel}}$, $\ell_t = 1$, and for $t > T_{\text{gel}}$, $0 < \ell_t < 1$ and ℓ_t is given by

$$\ell_t = H\left(\frac{1}{\alpha_t}\right). \quad (3.41)$$

- For every $y \in [0, 1]$, $\phi_t(\cdot, y)$ has a right inverse $h_t(\cdot, y) : [0, 1] \mapsto [0, 1]$.

3. The generating function $k_t(x, y) = \langle c_0, ax^{a-1}y^m \rangle$ is given by

$$k_t(x, y) = \frac{1}{\alpha_t} k_0(h_t(x, y), y) \quad (3.42)$$

for $y \in [0, 1]$, $x \in [0, 1]$.

4. In particular, for $t > 0$

$$A_t = \frac{k_0(\ell_t, 1)}{1 + \int_0^t k_0(\ell_s, 1) ds}, \quad \alpha_t = 1 + \int_0^t k_0(\ell_s, 1) ds. \quad (3.43)$$

5. The second moment $\langle c_t, a^2 \rangle$ is finite on $[0, T_{\text{gel}})$, infinite on $[T_{\text{gel}}, +\infty)$.
6. The mass $\langle c_t, m \rangle$ is continuous. It is constant on $[0, T_{\text{gel}})$ and strictly decreasing on $[T_{\text{gel}}, +\infty)$.

4.1 Proof

The only major difference with respect to the proof of Theorem 2.3 is the additional variable y in the generating function $k_t(x, y)$. However, the variable y plays the role of a parameter in the PDE (3.36), and this allows to adapt all above techniques.

Proof of Theorem 4.3. The case $K \leq A_0 < +\infty$, for which $T_{\text{gel}} = +\infty$ has already been treated in [3, Thm. 2], so that we can restrict here to the cases where $T_{\text{gel}} < +\infty$. When $T_{\text{gel}} > 0$, Thm. 2 in [3] also shows that $\alpha_t = 1 + A_0 t$ on $[0, T_{\text{gel}})$ (this however also requires that $\langle c_t, a^2 \rangle$ be bounded in a neighborhood of 0: see point 3 of the proof of Lemma 2.11).

1. First, by setting $u_t(x, y) := \alpha_t k_t(\phi_t(x, y), y) - k_0(x, y)$, we can see, arguing as in points (i)-(ii) of the proof of Lemma 2.11, that for all $y \in (0, 1]$ and $t > 0$ there exists $\ell_t^0(y) < \ell_t(y) \in (0, 1]$ such that

$$\alpha_t k_t(\phi_t(x, y), y) = k_0(x, y), \quad \forall t \geq 0, y \in (0, 1], x \in [\ell_t^0(y), \ell_t(y)] \quad (3.44)$$

and $\phi_t(\cdot, y) : [\ell_t^0(y), \ell_t(y)] \mapsto [0, 1]$ is a continuous bijection and has a continuous right-inverse $h_t(\cdot, y) : [0, 1] \mapsto [\ell_t^0(y), \ell_t(y)]$.

2. We denote for simplicity

$$k_t(x) := k_t(x, 1), \quad \phi_t(x) := \phi_t(x, 1), \quad t \geq 0, x \in [0, 1].$$

For $y = 1$, we set $\ell_t(1) = \ell_t$, i.e.

$$1 = \phi_t(\ell_t) = \alpha_t(\ell_t - \beta_t k_0(\ell_t)), \quad t \geq 0.$$

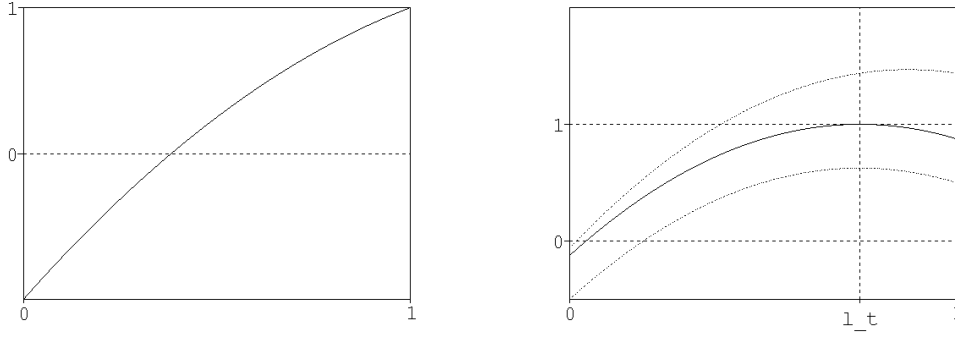


Figure 3.3: $\phi_t(\cdot, 1)$ before and after gelation. The dotted lines represent what ϕ_t may look like. The solid one is the actual ϕ_t .

Arguing as in points (iv)-(v) of the proof of Lemma 2.11, we can see that $\ell_t = 1$ for all $t \leq T_{\text{gel}}$ and $\ell_t < 1$ for all $t > T_{\text{gel}}$. Moreover, $t \mapsto \ell_t$ is continuous and monotone non-increasing. Since ϕ_t is increasing on $[0, \ell_t]$, $\phi_t'(\ell_t) \geq 0$, i.e.

$$\beta_t \leq \frac{1}{k_0'(\ell_t)},$$

so that

$$1 = \alpha_t(\ell_t - \beta_t k_0(\ell_t)) \geq \alpha_t G(\ell_t), \quad (3.45)$$

where we set $G(x) := x - \frac{k_0(x)}{k_0'(x)}$, $x \in [0, 1]$. Notice that

$$G'(x) = 1 - \frac{(k_0'(x))^2 - k_0(x)k_0''(x)}{(k_0'(x))^2} = \frac{k_0(x)k_0''(x)}{(k_0'(x))^2} > 0,$$

since k_0 is strictly convex (there is no gelation whenever $k_0'' \equiv 0$). Moreover $G(0) \leq 0$ and

$$G(1) = 1 - \frac{A_0}{K} \quad \text{if } A_0 < +\infty, \quad G(1) = 1 \quad \text{if } A_0 = +\infty.$$

Indeed, $k_0'(1) = K = \langle c_0, a(a-1) \rangle$ and, if $k_0(1) = A_0 = +\infty$, then

$$\lim_{x \uparrow 1} \frac{k_0(x)}{k_0'(x)} = 0$$

since, if $\liminf_{x \uparrow 1} \frac{k_0(x)}{k_0'(x)} > \varepsilon > 0$, then $k_0(1) \leq k_0(1 - \delta)e^{\delta/\varepsilon} < +\infty$, for some $\delta > 0$, contradicting $k_0(1) = +\infty$. In any case, G has an inverse H , and $H(1/x)$ is defined for $x \in [1 + A_0 T_{\text{gel}}, +\infty)$.

3. Computing (3.44) at $(x, y) = (\ell_t, 1)$ we obtain

$$k_0(\ell_t) = \alpha_t k_t(1) = \alpha_t A_t = \frac{d^+ \alpha_t}{dt}. \quad (3.46)$$

Let us notice that

$$\phi_t(x) = x + \int_0^t \left(A_s \phi_s(x) - \frac{k_0(x)}{\alpha_s} \right) ds.$$

Then by (3.46), analogously to (3.21) above,

$$0 = d\phi_t(\ell_t) = \left(A_t \phi_t(\ell_t) - \frac{k_0(\ell_t)}{\alpha_t} \right) dt + \phi_t'(\ell_t) d\ell_t = \phi_t'(\ell_t) d\ell_t.$$

In particular, for $d\ell_t$ -a.e. t , $\phi_t'(\ell_t) = 0$, i.e. $\beta_t = 1/k_0'(\ell_t)$, and therefore

$$1 = \alpha_t(\ell_t - \beta_t k_0(\ell_t)) = \alpha_t G(\ell_t), \quad d\ell_t - \text{a.e. } t.$$

Then, by (3.45), we can write (note that H is well-defined on the considered interval)

$$\ell_t \leq H\left(\frac{1}{\alpha_t}\right), \quad \forall t > T_{\text{gel}}, \quad \ell_t = H\left(\frac{1}{\alpha_t}\right), \quad d\ell_t - \text{a.e. } t.$$

Now, by (3.46), setting $\Lambda :]1 + A_0 T_{\text{gel}}, +\infty[\rightarrow]0, 1[$, $\Lambda(z) := k_0\left(H\left(\frac{1}{z}\right)\right)$,

$$\frac{d^+ \alpha_t}{dt} \leq \Lambda(\alpha_t), \quad \forall t > T_{\text{gel}}, \quad \frac{d^+ \alpha_t}{dt} = \Lambda(\alpha_t), \quad d\ell_t - \text{a.e. } t.$$

Since $\alpha_t > 1 + A_0 T_{\text{gel}}$ for any $t > T_{\text{gel}}$, we obtain that $k_0(\ell_t) \leq \Lambda(\alpha_t) < 1$ for all $t > T_{\text{gel}}$. In particular, $d\ell_t$ is not identically equal to 0. Suppose that for some $t > T_{\text{gel}}$ we have $\phi_t'(\ell_t) > 0$. We set

$$s := \sup\{r < t : \phi_r'(\ell_r) = 0\} = \max\{r < t : \phi_r'(\ell_r) = 0\}.$$

Then for all $r \in]s, t[$ we must have $\phi_r'(\ell_r) > 0$. Then for all $r \in]s, t[$ we have $\ell_r = \ell_s$. But, by definition of β ,

$$\beta_r > \beta_s = \frac{1}{k_0'(\ell_s)} = \frac{1}{k_0'(\ell_r)}$$

and this is a contradiction. Then

$$\frac{\partial \phi_t}{\partial x}(\ell_t, 1) = 0 \tag{3.47}$$

and for all $t > T_{\text{gel}}$, we have $\dot{\alpha}_t = \Lambda(\alpha_t)$ for all $t > T_{\text{gel}}$ and the only solution of this equation with $\alpha_{T_{\text{gel}}} = 1 + A_0 T_{\text{gel}}$ is given by (3.39).

4. In order to prove (3.43), let us note that by the preceding results

$$\frac{d\alpha_t}{dt} = \alpha_t A_t = \alpha_t k_t(1, 1) = k_0(\ell_t, 1),$$

and

$$A_t = \frac{d}{dt} \log \alpha_t = \frac{d}{dt} \log \left(1 + \int_0^t k_0(\ell_s, 1) ds \right) = \frac{k_0(\ell_t, 1)}{1 + \int_0^t k_0(\ell_s, 1) ds}.$$

The rest of the proof follows the same line as that of Theorem 2.3. \square

4.2 Convergence of the concentrations

We can now see what happens for the concentrations when time tends to infinity. Let us start by defining, for all $t \geq 0$, $x, y \in [0, 1]$, the generating function of c_t ,

$$g_t(x, y) = \sum_{a=0}^{+\infty} \sum_{m=1}^{+\infty} c_t(a, m) x^a y^m, \quad g_t(y) := g_t(0, y) = \sum_{m=1}^{+\infty} c_t(0, m) y^m. \tag{3.48}$$

In particular, $k_t = \frac{\partial g_t}{\partial x}$, with \bar{k}_t defined in (3.35). Note also that k_t does not involve $c_t(0, m)$, while g_t does.

Proposition 4.4. *When $t \rightarrow +\infty$, there exists limiting concentrations $c_\infty(m)$ such that*

$$c_t(a, m) \rightarrow c_\infty(m) \mathbb{1}_{\{a=0\}}$$

in $\ell^1(\mathbb{N} \times \mathbb{N}^*)$. Moreover, the generating function $g_\infty(y)$ of $(c_\infty(m))$ is given, for $y \in [0, 1]$, by

$$g_\infty(y) = \lim_{t \rightarrow +\infty} g_t(y) = \int_0^y \frac{\partial g_0}{\partial y}(h_\infty(0, y'), y') \, dy' \quad (3.49)$$

where

$$k_0(h_\infty(y), y) = \frac{1}{\beta_\infty} h_\infty(y), \quad \beta_\infty = \frac{1}{k'_0(H(0))} \in (0, +\infty],$$

and H is defined in Theorem 4.3.

Proof. Obviously, by (3.38), $A_t \rightarrow 0$ as $t \rightarrow +\infty$, so that the first claim follows. For the same reason, the formula

$$c_t(0, m) = c_0(0, m) + \frac{1}{2} \int_0^t \sum_{m'=1}^{m-1} c_s(1, m') c_s(1, m - m') \, ds$$

readily shows that $c_t(0, m)$ converges in $\ell^1(\mathbb{N}^*)$ to some $c_\infty(0, m)$. In particular

$$g_t(y) = \sum_{m'=1}^{+\infty} c_t(0, m) y^m \rightarrow \sum_{m'=1}^{+\infty} c_\infty(0, m) y^m := g_\infty(y) \quad (3.50)$$

for $y \in [0, 1]$. By differentiating $g_t(\phi_t(x, y))$ with respect to x , and using (3.42), we now see that

$$g_t(y) = \int_0^y \frac{\partial g_0}{\partial y}(h_t(0, y'), y') \, dy'. \quad (3.51)$$

Since $h_t(\cdot, y)$ is the right-inverse of $\phi_t(\cdot, y)$, then $h_t(y) := h_t(0, y)$ solves

$$k_0(h_t(y), y) = \frac{1}{\beta_t} h_t(y). \quad (3.52)$$

Note that by definition, $h_t(y) \leq \ell_t(y)$, and since ϕ_t is a decreasing function of y , then $\ell_t(y) \leq \ell_t$, so

$$h_t(y) \leq \ell_t. \quad (3.53)$$

Let us show that β_t has a limit when $t \rightarrow +\infty$. First (3.39) shows that $\alpha_t \rightarrow +\infty$, hence, by (3.41), $\ell_t \rightarrow \ell_\infty = H(0)$. Now, (3.47) gives $\beta_t = 1/k'_0(\ell_t)$, so β_t tends to

$$\beta_\infty = \frac{1}{k'_0(H(0))} = \frac{H(0)}{k_0(H(0))}. \quad (3.54)$$

Fix $y \in [0, 1]$, and let us find the limit of $h_t(y)$. Equations (3.52) and (3.53) show that every limit point $h_\infty(y)$ of $h_t(y)$ solves the equation

$$k_0(h_\infty(y), y) = \frac{1}{\beta_\infty} h_\infty(y) \quad (3.55)$$

with $h_\infty(y) \leq \ell_\infty$. But since $\ell_\infty = H(0) = k_0(\ell_\infty)/k'_0(\ell_\infty)$, then

$$k_0(\ell_\infty, y) < k_0(\ell_\infty) = \ell_\infty k'_0(\ell_\infty) = \frac{1}{\beta_\infty} \ell_\infty.$$

Since $k_0(\cdot, y)$ is convex, then this readily shows that Equation (3.55) has at most one solution in $[0, \ell_\infty)$. Hence $h_t(y)$ is bounded and has a unique limit point, so it converges to the unique $h_\infty(y)$ such that (3.55) holds. \square

5 The modified version

Let us finally consider Flory's version of the limited aggregations model. As in the case of Flory's equation (3.27), we can consider only initial concentrations c_0 such that $A_0 = \langle c_0, a \rangle < +\infty$. Then, the equation we are interested in is

$$\begin{aligned} \frac{d}{dt}c_t(a, m) &= \frac{1}{2} \sum_{a'=1}^{a+1} \sum_{m'=1}^{m-1} a'(a+2-a')c_t(a', m')c_t(a+2-a', m-m') \\ &\quad - \sum_{a' \geq 1} \sum_{m' \geq 1} aa'c_t(a, m)c_t(a', m') \\ &\quad - \left(\frac{A_0}{1+tA_0} - \sum_{a', m' \geq 1} a'c_t(a', m') \right) ac_t(a, m). \end{aligned} \quad (3.56)$$

This is Flory's version of Bertoin's equation with limited aggregations. Indeed, as for the standard Flory equation (3.27), an extra term is added to (3.34) accounting for interactions between the gel and the clusters in solution. Note indeed that

$$\frac{A_0}{1+tA_0} - \sum_{a', m' \geq 1} a'c_t(a', m')$$

is the "missing" number of arms in solution, in that if no clusters fall into the gel, then $A_t = A_0/(1+tA_0)$.

We consider here initial concentrations $c_0(a, m) \geq 0$, $a \in \mathbb{N}$, $m \in \mathbb{N}^*$ such that $\langle c_0, a \rangle \in (0, +\infty)$, and define the gelation time T_{gel} as in Definition 3.37. With the same techniques as above, we can prove the following result.

Theorem 5.1. *Equation (3.56) has a unique solution (c_t) defined on \mathbb{R}^+ . When $T_{\text{gel}} < +\infty$, this solution enjoys the following properties.*

1. The number of arms $A_t = \langle c_t, a \rangle$ is given for $t \geq 0$ by

$$A_t = \frac{1}{1+tA_0}k_0(l_t) \quad (3.57)$$

where $l_t = 1$ for $t \leq T_{\text{gel}}$ and, for $t > T_{\text{gel}}$, l_t is uniquely defined by

$$l_t = \frac{t}{1+tA_0}k_0(l_t), \quad l_t \in [0, 1].$$

Therefore $t \mapsto A_t$ is continuous and strictly decreasing on $[0, +\infty)$ and analytic on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$.

2. The function $\phi_t(x, y) = (1+tA_0)x - tk_0(x, y)$ has, for every $y \in [0, 1]$, a right-inverse $h_t(\cdot, y) : [0, 1] \rightarrow [0, l_t]$. The generating function $k_t(x, y) = \langle c_t, ax^{a-1}y^m \rangle$ defined in (3.35) is given for $t \geq 0$ by

$$k_t(x, y) = \frac{1}{1+tA_0}k_0(h_t(x, y), y). \quad (3.58)$$

3. The second moment $\langle c_t, a^2 \rangle$ is finite on $\mathbb{R}^+ \setminus \{T_{\text{gel}}\}$ and infinite at T_{gel} .
4. The mass $\langle c_t, m \rangle$ is continuous. It is constant on $[0, T_{\text{gel}}]$ and strictly decreasing on $[T_{\text{gel}}, +\infty)$.

Proof. The proof follows the same line of reasoning as the one of Theorem 3.2. First, for every $y \in [0, 1]$, $\phi_t(\cdot, y)$, as defined in the statement, has the following properties:

- (i) $\phi_t(0, y) \leq 0$, $\phi_t(1, y) \geq \phi_t(1, 1) = 1$.
- (ii) For $t \leq T_{\text{gel}}$, $\phi_t(\cdot, y)$ is increasing, and in particular, there are unique $0 \leq l_t^0(y) < l_t(y) \leq 1$ such that $\phi_t(l_t^0(y), y) = 0$ and $\phi_t(l_t(y), y) = 1$.
- (iii) For $t > T_{\text{gel}}$, $\phi_t(\cdot, y)$ is increasing then decreasing, and in particular, there are unique $0 \leq l_t^0(y) < l_t(y) < 1$ such that $\phi_t(l_t^0(y), y) = 0$ and $\phi_t(l_t(y), y) = 1$.

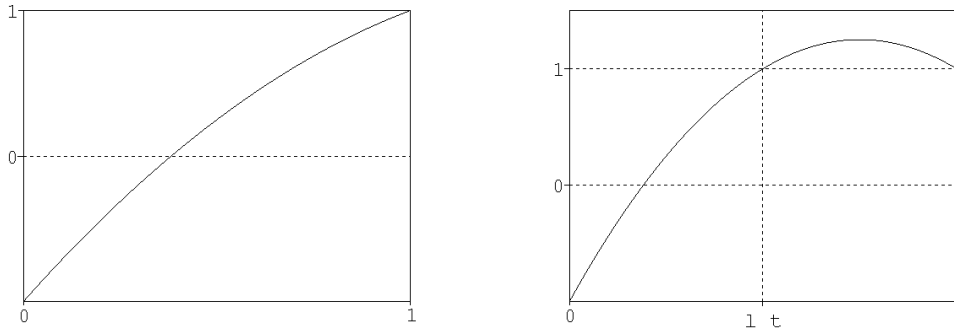


Figure 3.4: $\phi_t(\cdot, 1)$ before and after gelation.

In any case, it is easy to check that for $x \in [l_t^0(y), l_t(y)]$,

$$\exp\left(\int_0^t A_s ds\right) k_t(\phi_t(x, y), y) = k_0(x, y)$$

where A_t is defined by (3.57). Then, the properties above show that $\phi_t(\cdot, y)$ has a right inverse h_t defined on $[0, 1]$, and compounding by h_t in the previous equation shows that (3.58) holds. The other properties then follow easily. \square

6 Limiting concentrations

We compute here some explicit formulas for the concentrations and their limit for the two models above. In the standard Smoluchowski and Flory cases, clusters keep coagulating, and they all eventually disappear into the gel: $c_t(m) \rightarrow 0$ for every $m \geq 1$. When the aggregations are limited, there may remain some clusters in the system, since whenever a cluster with no arms is created, it becomes inert, and so it will remain in the medium forever. In the following, we consider monodisperse initial conditions, i.e. $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$ for a measure μ on \mathbb{N} . We also denote

$$\nu(m) = (m+1)\mu(m+1).$$

In [3], it is assumed that ν is a probability measure, what we do not require. The results of [3] can hence be recovered by taking $A_0 = 1$ below. Now, note the two following facts.

- Equations (3.38) and (3.57) readily show that

$$c_\infty(a, m) := \lim_{t \rightarrow +\infty} c_t(a, m) = 0, \quad a \geq 1, \quad (3.59)$$

that is, only clusters with no arms remain in the medium (else, a coagulation “should” occur).

- There is an arbitrary concentration of particles with no arms at time 0, and they are the only clusters with no arms and mass 1 which will still be in the medium in the final state. Hence, the limit concentrations $c_\infty(0, 1) = c_0(0, 1)$ have no physical meaning. We will thus only consider $c_\infty(0, m)$ for $m \geq 2$.

Note now that if at time 0, each particle has zero or more than two arms, then obviously, this property still holds for any positive time. Rigorously, this is easy to check with the representation formula (3.42) or (3.58). Then, because of (3.59),

$$c_\infty(m) = 0$$

for each $m \geq 2$. We thus rule out this trivial case by assuming that

$$\mu(1) > 0. \quad (3.60)$$

This is actually a technical assumption which is needed to apply Lagrange's inversion formula in the proof of the following corollaries.

6.1 Modified model

Corollary 6.1. *Let $c_t(a, m)$ be the solution to Flory's equation with limited aggregations (3.56) and with initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$ with $\mu(1) > 0$.*

- For all $t \geq 0$, $m \geq 2$, $a \geq 0$,

$$c_t(a, m) = \frac{(a + m - 2)!}{a!m!} \frac{t^{m-1}}{(1 + tA_0)^{a+m-1}} \nu^{*m}(a + m - 2).$$

- In particular, there are limiting concentrations $c_\infty(a, m) = c_\infty(m)\mathbb{1}_{\{a=0\}}$ with

$$c_\infty(m) = \frac{1}{m(m-1)} \nu^{*m}(m-2). \quad (3.61)$$

Proof. With the notation of Theorem 5.1, we have

$$(1 + tA_0)h_t(x, y) - tyk_0(h_t(x, y)) = x, \quad k_t(x, y) = \frac{1}{1 + tA_0} yk_0(h_t(x, y)).$$

Up to some obvious changes (just replace $1+t$ by $1+tA_0$), these are precisely the equations solved in Section 3.2 of [3] under the assumption that $\mu(1) > 0$. Theorem 2 and Corollary 2 therein hence give the desired result (with only $1+t$ replaced by $1+tA_0$). \square

If $A_0 = 1$, what we may always assume up to a time-change, then

$$c_\infty(0, m) = \frac{1}{m} \langle mu, 1 \rangle \mathbb{P}(T(\mu_0, \nu) = m),$$

where $\mu_0 = \mu/\langle \mu, 1 \rangle$, and $T(\mu_0, \nu)$ is the size of a Galton-Watson $\mathbb{GW}_{\mu_0, \nu}$ starting from an ancestor with reproduction law μ_0 , and such that every other individual has reproduction law ν . This Galton-Watson process is (sub)critical when $K \leq 1$ (i.e. there is no gelation), and supercritical when $K > 1$. Similarly to the explication given in Section 3.3.2, this may be interpreted by saying that a typical *finite* cluster in solution is a Galton-Watson tree $\mathbb{GW}_{\mu_0, \nu}$.

Denote now by p_ν its extinction probability, i.e. the smallest root of $k_0(x) = x$, so $p_\nu = 1$ when $K \leq 1$ and $p_\nu < 1$ when $K > 1$. Let us compute the mass at infinity, as in [3], by writing

$$\begin{aligned} M_\infty &:= \sum_{m \geq 1} m c_\infty(m) = c_\infty(1) + \sum_{m \geq 2} \frac{1}{m-1} \nu^{*m} (m-2) \\ &= c_\infty(1) + \sum_{a \geq 0} \nu(a) \sum_{m \geq a+2} \frac{1}{m-1} \nu^{*m-1} (m-2-a) \\ &= c_\infty(1) + \sum_{a \geq 0} \nu(a) \sum_{n \geq a+1} \frac{1}{n} \nu^{*n} (n-1-a). \end{aligned}$$

Now, the Lagrange inversion formula [58] shows that

$$\frac{a+1}{n} \nu^{*n} (n-1-a)$$

is precisely the coefficient of x^n in the analytic expansion of $\phi(x)$ around 0, where ϕ is the unique solution to $\phi(x) = xk(\phi(x))$. Hence

$$\sum_{n \geq a+1} \frac{1}{n} \nu^{*n} (n-1-a) = p_\nu,$$

where p_ν is defined above. Note also that $c_\infty(1) = \mu(0)$, so finally

$$M_\infty = c_\infty(1) + \sum_{a \geq 0} \nu(a) \frac{1}{a+1} p_\nu^{a+1} = \sum_{a \geq 0} \mu(a) p_\nu^a. \quad (3.62)$$

The mass at time 0 is $M_0 = \sum \mu(a)$, so when there is no gelation, $p_\nu = 1$ and no mass is lost in the gel. When there is gelation, $p_\nu < 1$ and the mass $M_0 - M_\infty > 0$ is lost in the gel.

6.2 Non-modified model

Corollary 6.2. *Let $c_t(a, m)$ be the solution to Smoluchowski's equation with arms (3.34) and with initial conditions $c_0(a, m) = \mu(a) \mathbb{1}_{\{m=1\}}$ with $\mu(1) > 0$.*

- For all $t \geq 0$, $m \geq 2$, $a \geq 0$,

$$c_t(a, m) = \frac{(a+m-2)! \beta_t^{m-1}}{a! m! \alpha_t^a} \nu^{*m} (a+m-2)$$

where α_t and β_t are defined in Theorem 4.3.

- In particular, there are limiting concentrations $c_\infty(a, m) = c_\infty(m) \mathbb{1}_{\{a=0\}}$ with

$$c_\infty(m) = \frac{1}{m(m-1)} \beta_\infty^{m-1} \nu^{*m} (m-2) \quad (3.63)$$

where β_∞ is defined by

$$\beta_\infty = \frac{1}{k'_0(\eta)} = \frac{\eta}{k_0(\eta)}$$

and η is the unique solution to

$$k'_0(\eta) = k_0(\eta)/\eta.$$

Moreover, $\beta_\infty = 1$ when there is no gelation, and $\beta_\infty > 1$ otherwise.

Proof. As for Corollary 6.1, the proof of the formula for $c_t(a, m)$ is the same as in [3, Section 3.2], just replacing $1 + tA_0$ by α_t and t by $\alpha_t\beta_t$. So we just have to find the limit of β_t . First (3.39) shows that $\alpha_t \rightarrow +\infty$, hence, by (3.41), $\ell_t \rightarrow \ell_\infty = H(0)$. Now, (3.47) gives $\beta_t = 1/k'_0(\ell_t)$, so β_t tends to

$$\beta_\infty = \frac{1}{k'_0(H(0))}$$

where by definition $\eta := H(0)$ is the unique solution to $k'_0(\eta) = k_0(\eta)/\eta$. Finally, when there is gelation, $\alpha_t < 1 + t$ after gelation because of (3.39), so by definition of β_t , $\beta_\infty > 1$. \square

By a similar computation as above, we may also compute the mass at infinity in this case and get

$$M_\infty = \sum_{a \geq 0} \mu(a) \eta^a$$

where η is defined in the corollary. Note that η is the slope of the straight line passing by 0 and tangent to the graph of k , so $\eta > p_\nu$. In particular, less mass is lost than in Flory's case.

A final remark is that despite the striking resemblance between Formulas (3.63) and (3.61), the meaning of the factor β_∞ is unclear. A probabilistic interpretation explaining its appearance is given in the next chapter.

Chapter 4

Self-organized criticality in a microscopic model for Smoluchowski's equation

The material of this chapter is a joint work [41] with Mathieu Merle to be submitted soon.

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1 Introduction

1.1 Smoluchowski's equation

The goal of this chapter is to study a discrete model for Smoluchowski's equation with limited aggregations (1.3). Let us, to begin with, recall some facts from the introduction of this thesis and from the previous chapter.

The standard Smoluchowski equation was introduced in 1916 in [57], to model pairwise coalescing particles. As time passes, clusters of particles are created. We assume that each particle has unit mass, so a cluster has mass m , the number of particles it contains. The coagulation phenomenon is characterized by a symmetric kernel $\kappa(m, m')$, modeling the "rate" at which two clusters of mass m and m' coalesce. When they do, a cluster of mass

$m + m'$ is formed. For the multiplicative kernel, the evolution of the concentration $c_t(m)$ of clusters of mass m at time t is given by Smoluchowski's equation

$$\frac{d}{dt}c_t(m) = \frac{1}{2} \sum_{m'=1}^{m-1} mm' c_t(m') c_t(m - m') - \sum_{m' \geq 1} mm' c_t(m) c_t(m'), \quad (4.1)$$

for $m \in \mathbb{N}^*$.

For that the multiplicative kernel, two facts have been known for a long time [39], when we start from monodisperse initial conditions $c_t(m) = \mathbb{1}_{\{m=1\}}$. First, there is a unique solution on $[0, 1]$, and it is mass-conservative, in that $\sum m c_t(m)$ remains constant. Secondly, if there is actually a solution on a larger interval, then the mass has to decrease after time 1. This phenomenon is called *gelation*. Physically, it is interpreted as the appearance of a giant cluster, containing a positive fraction of the particles. Actually, there may just as well be several giant clusters, though we talk about “the” giant cluster by abuse of language. In general, this giant particle is called the *gel*, and the particles not in the gel are *in solution*.

A proof of existence and uniqueness to Smoluchowski's equation with a multiplicative kernel and general initial conditions is given in the previous chapter. We will provide more details below for the model with limited aggregations we are interested in.

1.2 The model with limited aggregations

To avoid that a particle bonds to infinitely many other particles, one may initially give each particle a certain number of arms. These arms are used to create the bonds, in that two arms are used to create a link and cannot be reused again. Then, a cluster is characterized by its mass m and its number of available arms a , i.e. the total number of non-used arms of the particles it is formed of. This model has first been considered by Bertoin in [3], and the evolution of concentration $c_t(a, m)$ of (a, m) -clusters, i.e. clusters with a (available) arms and mass m is given by

$$\begin{aligned} \frac{d}{dt}c_t(a, m) &= \frac{1}{2} \sum_{a'=1}^{a+1} \sum_{m'=1}^{m-1} a'(a+2-a')c_t(a', m')c_t(a+2-a', m-m') \\ &\quad - \sum_{a' \geq 1} \sum_{m' \geq 1} aa'c_t(a, m)c_t(a', m'). \end{aligned} \quad (4.2)$$

The first term on the RHS accounts for the appearance of (a, m) -clusters, by coagulation of (a', m') -clusters with $(a+2-a', m-m')$ -clusters. The second term on the RHS accounts for the disappearance of such clusters, by coagulation with any other cluster.

In [3], Bertoin considers initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$ for a probability μ with a second moment (beware of a slight change of notations with [3] and the first chapters, more adapted to our case). We define ν the *size-biased shifted-by-one*, probability deduced from μ , or just size-bias of μ , given for $k \in \mathbb{N}$ by

$$\nu(k) = \frac{(k+1)\mu(k+1)}{\sum_{i \geq 1} i\mu(i)}.$$

Let also $A = \sum_{k \geq 1} k\mu(k)$, $K = \sum_{k \geq 1} k(k-1)\mu(k)$ and define

$$T_{\text{gel}}^0 = \begin{cases} +\infty & \text{if } K \leq A \\ \frac{1}{K-A} & \text{if } K > A. \end{cases} \quad (4.3)$$

Bertoin then shows that (4.2) has a unique solution up to time T_{gel}^0 , interpreted as the *gelation time*. Moreover, when $T_{\text{gel}}^0 = +\infty$, or equivalently when ν has mean at most 1, there are limiting concentrations $c_\infty(a, m) = \mathbb{1}_{\{a=0\}} c_\infty(m)$, where

$$c_\infty(m) = \frac{1}{m(m-1)} \nu^{*m}(m-2)$$

and which can be interpreted in terms of the total progeny of some Galton-Watson process. A rigorous explanation of this relation is given in the subsequent paper [4]. We refer to Section 3.3.2 of the introduction for an intuitive presentation of the result.

In the previous chapter, we showed that actually, for general initial conditions, Equation (4.2) has a unique solution on the whole of \mathbb{R}^+ . For monodisperse initial conditions $c_0(a, m) = \mu(a) \mathbb{1}_{\{m=1\}}$, the limiting concentrations can be computed. Assume to this end that ν has mean greater than 1, i.e. that $T_{\text{gel}}^0 < +\infty$. Then, the equation

$$\eta G'_\nu(\eta) = G_\nu(\eta), \quad (4.4)$$

where G_ν is the generating function of ν , has a unique solution.

Moreover, when $t \rightarrow +\infty$,

$$c_t(a, m) \rightarrow c_\infty(m) \mathbb{1}_{\{a=0\}}$$

in $\ell^1(\mathbb{N} \times \mathbb{N}^*)$, where

$$c_\infty(m) = \frac{1}{m(m-1)} \beta^{m-1} \nu^{*m}(m-2), \quad (4.5)$$

β is given by

$$\beta = \frac{\eta}{G_\nu(\eta)} = \frac{1}{G'_\nu(\eta)}. \quad (4.6)$$

The mass at infinity can also be computed and is provided by the formula

$$M_\infty = G_\mu(\eta). \quad (4.7)$$

1.3 Microscopic model

This last result is actually the initial motivation for the present work. As in [4], we wish to give a probabilistic interpretation for these concentrations. However, preliminary precautions are to be taken. Let us indeed recall the issues raised when one studies microscopic models for Smoluchowski's equation (4.1) with the multiplicative kernel.

Informally, consider N particles, and let two clusters of mass m and m' coagulate at rate mm' . Then, it is well-known [27, 45] that, as $N \rightarrow +\infty$, there is a limiting process which is deterministic and solves Smoluchowski's equation, but only before the gelation time. On \mathbb{R}^+ , the Equation which is solved is actually Flory's equation (see [17]), which is quite natural since, in the discrete model, any cluster can interact with any other, whatever their sizes are.

On the other hand, Smoluchowski's equation only takes into account coagulation of particles of finite size (informally, only finite values of m appear in the equation), or, in other words, there is no interaction with the gel. An appropriate microscopic model would then prevent "big" particles from interacting with the others. This idea has been formalized by Fournier and Laurençot in [19]. In this paper, they allow only the particles of size less than $\alpha(N)$ to coagulate, where $1 \ll \alpha(N) \ll N$, and show that the process

converges (up to a subsequence) to a solution of Smoluchowski's equation. That is the idea we take over to build our microscopic model, see Section 2.1.

When particles have arms, we can as well consider microscopic evolutions with or without interactions with the gel. The second will be called Smoluchowski's evolution, and that is the one we are interested in; the first will be called Flory's evolution, which is easier to study. It will be sometimes useful to compare the two models.

1.4 Brief description of results

In the present work, we aim at recovering Bertoin's equation of coagulation with limited aggregations in the supercritical regime, when particles in solution do not interact with those in the gel, while keeping as much as possible track of the random graph structure. This allows us to express limiting equations for the evolution of concentrations of particle with a given number of already linked arms, to solve these equations and recover the formula (4.5) for limiting distributions of [44]. We are also able to express the total concentration of particles in solution at any time after gelation and in particular recover (4.7).

The fact that we are able to track the evolution of the concentrations is due to a remarkable (though not very surprising) feature of the asymptotics of our discrete model: it exhibits *self-organized criticality* (SOC) after gelation. More precisely, the limiting distribution of already linked arms at any time t past gelation has a size-bias law with mean 1.

It is worth noticing that the SOC here is really a microscopic phenomenon, in that it concerns the law of the number of arms of the particles. In particular, it could not have been deduced from the result of [44], since the distribution of already linked arms cannot be recovered from the variables $c_t(a, m)$ of (4.2).

The phenomenon of SOC is well-known in the Physics literature, but is seldom proved mathematically. It has however been observed in the beautiful paper [47] by Tóth and Ráth. In this article, they study a model of fire on trees, where two phenomena compete (growth of trees vs. fires) to reach criticality. The criticality is lost when one of the two phenomena has a far bigger influence than the other: informally, either all trees burn, or none. In our case, we also observe that criticality is attained because the connections of arms compete with the falls into the gel.

1.5 Plan of the paper

In this paper, we shall first provide a rigorous presentation of the model we consider, as well as the statement of the results. We also discuss them, explain the phenomenon of post-gelation self-organized criticality (SOC), and show how our results relate to those obtained in [44]. In particular, we provide an explanation of the limiting concentrations in terms of Galton-Watson processes. The remaining sections are devoted to the proof of these results. We first provide some preliminary lemmas and present useful techniques we will use throughout the text, before turning to the core results of the paper: first, the subcriticality, which enables us to show the tightness of various relevant quantities. It is then an easy matter to show the actual SOC. The last difficult proposition we prove is a result of asymptotic independence, stating that two subclusters connected to a particle have essentially independent behaviors. All this allows us to write an equation for the evolution of some relevant quantities, which, in the limit, turns out to be an easy PDE. Solving it allows us to deduce an expression for various interesting quantities, and in

particular, we obtain the uniqueness of the limit points, what ends our proof. Finally, so as not to weigh down the proofs, several technical issues are deferred to the appendices.

2 Presentation of the model and of the results

2.1 Construction of the model

2.1.1 Assumptions

Let us, to begin with, introduce the assumptions we shall make. We start with N particles, denoted $[N] = \{1, \dots, N\}$, respectively having $a_1 = a'_{\sigma(1)}, \dots, a_n = a'_{\sigma(N)}$ arms, where a'_1, \dots, a'_N are fixed (and depend on N) and σ is a uniformly chosen permutation of $\{1, \dots, N\}$, independent from the dynamic. We introduce σ only to make the particles exchangeable; doing otherwise would make some results quite cumbersome to state¹. We write

$$\mu^{(N)} := \frac{1}{N} \sum_{i=1}^N \delta_{a'_i}$$

the empirical measure of the number of arms. The a_i arms of the i -th particle are denoted $(i, 1), \dots, (i, a_i)$.

We also fix a probability measure $\mu \neq \delta_0$ on \mathbb{N} with a finite second moment. Here are our two main assumptions on $\mu^{(N)}$ and μ .

Assumption 2.1. *The sequence $(\mu^{(N)})_{N \geq 1}$ converges sharply to μ , i.e. $(\mu^{(N)})$ converges weakly to μ and the second moment of $\mu^{(N)}$ converges to the second moment of μ .*

Assumption 2.2. *The measure μ has three moments, i.e.*

$$\langle \mu, k^3 \rangle < \infty.$$

We also fix a sequence $\alpha(N) \geq 1$, and our last assumption, is the following.

Assumption 2.3. *The sequence $\alpha(N)$ has the property that*

$$\frac{\alpha(N)}{N} \xrightarrow{N \rightarrow \infty} 0 \quad \text{and} \quad \frac{\alpha(N)}{N^{1/3}} \xrightarrow{N \rightarrow \infty} +\infty.$$

It is a stronger assumption than that of Fournier and Laurençot [19], viz. $1 \ll \alpha(N) \ll N$, and the reason for that choice will appear clearly when introducing Janson's result, Theorem 2.6.

2.1.2 Model

We may now introduce our model. It is defined by only two quantities, namely a family $\{e_{(i,j)}, i, j \in \mathbb{N}\}$ of i.i.d. exponential random variables with parameter 1, and a sequence $(\alpha(N))_{N \geq 1}$. The clock $e_{(i,j)}$ should be interpreted as the time when the j -th arm of the i -th particle (if it exists) is activated, and $\alpha(N)$ as a threshold: clusters of size greater than $\alpha(N)$ fall into the gel. Let us note that the exponential variables may or may not depend on N , but this does not really matter since we are only interested in convergence in law. Moreover, a coupling ensuring a stronger convergence seems quite hard to formulate.

¹However, we will not reuse σ in the text. One should just remember that the particles are exchangeable at time 0.

At each time $t \geq 0$, our model provides a configuration (i.e. a multigraph) \mathcal{C}_t . We say that a particle is *in solution* if its connected component has size less (or equal) than $\alpha(N)$, else we say that it is *in the gel*. An arm is in solution or in the gel if the particle to which it is attached is in solution or in the gel.

Now, when it exists, the j th arm of the i th particle is activated at time $e_{(i,j)}$. Hence, at any time t , an arm will be said to be *active* if the corresponding clock has rung, and *available* if it has not. Finally, an arm is said to be *free* if it is not bound.

We join pair of free arms of particles whose respective clocks ring consecutively *when they are both in solution*. To be more precise, when it exists, the j -th arm of the i -th particle is activated at time $e_{(i,j)}$.

- If i is in the gel at that time, or if i is still in solution *and* there is no other free active arm in solution, nothing happens.
- If on the other hand, i is in solution *and* there is another free active in solution, say (k, l) , then the arms (i, j) and (k, l) get bound together. In the latter case, if the newly formed cluster contains more than $\alpha(N)$ particles, it falls instantaneously into the gel and will remain there forever. Otherwise it stays in solution.

Note also that we proceed in such a way that the process is càdlàg.

The graph structure is obvious: particles stand for vertices, and bounds between particles for edges. Hence, at a given time $t > 0$, two particles i, j are *neighbors* at time t if at least one arm of i was paired with an arm of j before time t .

The major difficulty in dealing with this model is gelation: any cluster reaching size greater than $\alpha(N)$ instantaneously falls into the gel² and stops interacting with the other clusters.

Remark 2.4. It is important to note that this model is only given by exponential clocks, and that the state at a time t depends only on

- the activated arms;
- the order of these activations.

Moreover, conditionally on the activated arms, this order is uniform.

Thanks to this remark, we may present equivalent constructions of the model at a fixed time t . First, activate each arm independently with probability $1 - e^{-t}$. We then need to bind these arms, and to this end, we may

- either choose independently a uniform ordering of these arms, and bind them as in the algorithm described above (the first and second in solution, the third and fourth in solution, and so on);
- or choose a couple of free active arms in solution uniformly among all such couples and bind them, then choose another couple of free active arms in solution uniformly, and so on;
- or choose a free active arm uniformly in solution, another free active arm uniformly in solution, and bind them; then choose a free active arm in solution and another free active arm in solution, and so on.

In any case, we stop when all the active arms, except possibly the last one, are paired.

²This is just a physical metaphor: nothing actually happens to these clusters!

2.2 Notation

2.2.1 General notations

The following notations and definitions will be used a lot throughout the text.

- The probabilities considered are always probabilities on $\mathbb{N} = \{0, 1, 2, \dots\}$.
- For a probability π with positive and finite mean, its size-biased shifted-by-one counterpart (or simply “its size-bias”) is denoted $\hat{\pi}$, i.e., for every $k \geq 0$,

$$\hat{\pi}(k) = \frac{(k+1)\pi(k+1)}{\sum_{i \geq 1} i\pi(i)},$$

which is also a probability measure. If $\pi = \delta_0$, we let $\hat{\pi} = \delta_0$.

- For a nonnegative function f on \mathbb{N} , we write

$$\langle \pi, f \rangle := \sum_{k \geq 0} \pi(k)f(k).$$

We will write k for the function $k \mapsto k$, k^2 for $k \mapsto k^2$, and so on; for instance $\langle \pi, k \rangle$ is the mean of π .

- We say that π is subcritical, critical or supercritical when $\langle \hat{\pi}, k \rangle < 1$, $\langle \hat{\pi}, k \rangle = 1$ or $\langle \hat{\pi}, k \rangle > 1$.
- A sequence of events (E_N) is said to hold *with high probability*, written w.h.p., if

$$\lim_{N \rightarrow +\infty} \mathbb{P}(E_N) = 1.$$

- A *configuration* G is a multigraph, and for a vertex i , $G(i)$ is its connected component, or cluster, and $|G(i)|$ its cardinality, or size.
- Recall also that we fixed a probability measure $\mu \neq \delta_0$ on \mathbb{N} with a finite second moment, and let $\nu = \hat{\mu}$. The mean of ν is denoted $\zeta := \langle \nu, k \rangle$ and we define the gelation time as

$$T_{\text{gel}} = -\log \left(1 - \frac{1}{\zeta} \right)$$

if $\zeta > 1$, and $+\infty$ else.

The following notations are less important and will be recalled when necessary. We give them here anyhow so the reader can refer to this section if necessary.

- For ξ a probability on $\mathbb{N} \times \mathbb{N}$, we let

$$\hat{\xi}(k, l) = \frac{(k+1)\xi(k+1, l)}{\sum_{i, j \geq 1} i\xi(i, j)},$$

and $\xi = \delta_{(0,0)}$ if $\xi(i, j) = 0$ for $i \geq 1$.

- For a nonnegative function f on \mathbb{N}^2 , we write

$$\langle \xi, f \rangle := \sum_{i, j \geq 0} \xi(i, j)f(i, j).$$

We will write i for the function $(i, j) \mapsto i$, j for $(i, j) \mapsto j$ and so on; for instance $\langle \xi, i \rangle$ is the mean of the first marginal of ξ .

- The generating function of a probability π is denoted $G_\pi : x \mapsto \langle \pi, x^k \rangle$.
- Finally, for $p \in [0, 1]$, $\text{Bin}(\pi, p)$ is the mixing of a binomial law and of π , in that

$$\text{Bin}(\pi, p)(k) = \sum_{i \geq k} \binom{i}{k} \pi(i) p^k (1-p)^{i-k}, \quad k \in \mathbb{N}.$$

This law is obtained by taking a variable X with law π , and then by picking, conditionally on X , a variable with law $\text{Bin}(X, p)$. In our context, this can be interpreted as follows: a particle has a number of arms given (asymptotically) by a law μ . At a time t , each is activated independently with probability $1 - e^{-t}$. Then the number of activated arms of this particle has law $\text{Bin}(\mu, 1 - e^{-t})$.

2.2.2 Notions of convergence

In the whole text, convergences are understood as convergences in law.

- We denote \mathcal{M}_1 the space of probability measures on \mathbb{N} , and it is endowed with the weak topology³.
- We say that a sequence $(\pi^{(N)})_{N \geq 1}$ of probabilities on \mathbb{N} converges *sharply* to a probability π if $\pi^{(N)}$ converges to π weakly, and $\langle \pi^{(N)}, k^2 \rangle \rightarrow \langle \pi, k^2 \rangle < +\infty$.
- Similarly, a sequence $(\xi^{(N)})$ of probabilities on \mathbb{N}^2 is said to converge *sharply* to a probability ξ if $\xi^{(N)}$ converges to ξ weakly and $\langle \pi^{(N)}, f \rangle \rightarrow \langle \pi, f \rangle < +\infty$ for $f(i, j) = i^2$ and j^2 .

We will in the sequel consider random probability measures π , so for instance $\langle \pi, k \rangle$ is a real random variable. In that case, we have similar definitions.

- We say that a sequence $(\pi^{(N)})$ of *random* probabilities on \mathbb{N} converges *sharply* to a (random) probability π if $\pi^{(N)}$ converges to π in law in \mathcal{M}_1 , and $\langle \pi^{(N)}, k^2 \rangle \rightarrow \langle \pi, k^2 \rangle < +\infty$ in law in \mathbb{R} .
- The space of càdlàg mappings from $\mathbb{R}^+ = [0, +\infty)$ to a Polish space E is denoted $\mathbb{D}(\mathbb{R}^+, E)$, and it is endowed with the Skorokhod topology.
- We say that a càdlàg \mathcal{M}_1 -valued process $(\pi_t^{(N)}, t \geq 0)_{N \in \mathbb{N}}$ converges sharply to $(\pi_t, t \geq 0)$ if $(\pi^{(N)})$ converges in law to π in $\mathbb{D}(\mathbb{R}^+, \mathcal{M}_1)$ and $\langle \pi^{(N)}, k^2 \rangle \rightarrow \langle \pi, k^2 \rangle < +\infty$ in law in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$.

Finally, we will say that a sequence of probabilities, random probabilities or probability-valued processes is *sharply tight* if it is tight and that the convergence of a subsequence implies its sharp convergence.

³which is the topology of convergence in law. However, we shall speak of convergence in law only when we refer to convergence of random elements.

2.2.3 Characteristics of the model

Let us now introduce some quantities related to the model. Obviously, all quantities depend on N (which we write as an exponent (N) or an index N), but we will omit it in some notations.

As for the already defined notations, some will be used a lot in the text and we give them here, others are more seldom used and will be recalled, though we give them here as a memorandum.

- Recall that \mathcal{C}_t is the configuration at time t , $\mathcal{C}_t(i)$ is the cluster of i and $|\mathcal{C}_t(i)|$ its cardinality, or size.
- We write $i \overset{t}{\leftrightarrow} j$ if i and j are linked at time t .
- We say that a cluster is *giant* if it is in the gel, i.e. if it has size greater than $\alpha(N)$.
- We let $I_S(t)$ denote the set of particles in solution at time t , $I_G(t) = [N] \setminus I_S(t)$ the set of particles in the gel at time t .

The most important quantities are $n_t^{(N)}$, $\pi_t^{(N)}$ and $\rho_t^{(N)}$ defined as follows.

- We denote N_t the cardinal of $I_S(t)$, that is the number of particles in solution, and

$$n_t^{(N)} = N_t/N$$

the concentration in solution.

- For $t \geq 0$, $i \in [N]$, $A_t(i)$ is the number of active arms of i and for $k \in \mathbb{N}$, we let $\mathbf{N}_t(k) = \#\{i \in I_S(t), A_t(i) = k\}$ denote the number of particles in solution which have k activated arms at that time. Then the probability $\pi_t^{(N)}$ is the empirical distribution of already connected arms of particles in solution

$$\pi_t^{(N)}(k) := \frac{\mathbf{N}_t(k)}{N_t}.$$

- Its size-bias is denoted

$$\rho_t^{(N)} := \hat{\pi}_t^{(N)}.$$

Let us now introduce less important quantities, though technically crucial.

- The N -th gelation time $T_{\text{gel}}^{(N)}$ is the first time at which a cluster falls into the gel:

$$T_{\text{gel}}^{(N)} = \inf\{t \geq 0, \exists i \in [N] |\mathcal{C}_t(i)| > \alpha(N)\}.$$

- The number of available arms of i is denoted $B_t(i) = a_i - A_t(i)$.
- The empirical distribution of the activated and available arms of the particles at time t is denoted $\xi_t^{(N)}$, i.e.

$$\xi_t^{(N)}(k, l) = \frac{1}{N_t} \#\{i \in I_S(t), A_t(i) = k, B_t(i) = l\}.$$

- For $k \in \mathbb{N}$, the concentration of particles with k activated arms and in solution is denoted

$$\mathbf{n}_t^{(N)}(k) = \mathbf{N}_t(k)/N.$$

- In the same vein, we also introduce the concentration of particles with k active arms, n in total, and in solution at time t :

$$\mathbf{p}_t^{(N)}(n, k) = \frac{1}{N} \#\{i \in I_S(t), a_i = n, A_t(i) = k\}.$$

Note that the Greek lowercase letters denote probability measures, uppercase Latin letters are integers, and bold letters are vectors (indexed by \mathbb{N} or $\mathbb{N} \times \mathbb{N}$), notations that we try to maintain throughout the text.

The configuration of connected components of particles in solution at time t is closely linked with configuration models having a total of vertices close to N_t and a distribution of degrees close to $\pi_t^{(N)}$. Before stating our results, we recall a few facts about the configuration model.

2.3 Configuration model with fixed sequence of degrees

We shall now recall some facts about the configuration model, which we already discussed in the introduction of this thesis.

2.3.1 Definition

Let $N \in \mathbb{N}$ and $D_N := (d_1, \dots, d_N) \in \mathbb{N}^N$ be fixed. We also define

$$\mathbb{D}_N = \frac{1}{N} \sum_{i=1}^N \delta_{d_i}$$

as the empirical measure of the degrees. To each vertex i we associate d_i half-edges (arms) which we denote by (i, j) , $1 \leq j \leq d_i$. We define the configuration model with N vertices and the fixed degrees D_N as a uniformly chosen pairing amongst all possible pairings of the $\{(i, j), 1 \leq i \leq N, 1 \leq j \leq d_i\}$, leaving possibly one edge unpaired when $\sum_{i=1}^N d_i$ is odd⁴.

For each half edge (i, j) paired with an half edge (k, l) we draw one edge between vertex i and vertex k . The configuration which is obtained is a multigraph which we denote $CM(N, D_N)$ or also $CM(N, \mathbb{D}_N)$. Note that there may exist loops and multiple-edges, and there is possibly one unpaired half-edge attached to one of the vertices, which does not play any role in the cluster configuration. A good reference about random graphs and the configuration model is van der Hofstad's course [54].

2.3.2 Construction of the configuration model

There are multiple equivalent ways to construct a configuration model $CM(N, D_N)$, and it will be important in the sequel to understand them. Denote K_N the total number of arms.

⁴In the classical definitions of the configuration model, the choice is to either condition on $\sum d_i$ being even, or to add an extra edge for the last vertex. Although these two alternate definitions may be nicer when proving results on graphs, they are not well-suited for our model.

- A first way is to choose a uniform random ordering of the arms, and to bind together the $(2i-1)$ -th and $2i$ -th arms, for all $1 \leq i \leq \lfloor K_N/2 \rfloor$. To do so, one may for instance set i.i.d. exponential clocks on the arms, what gives a natural ordering of them.
- Another way is to pick a vertex arbitrarily⁵. Then choose one of its arms, and bind it with another one uniformly chosen among all available arms. Then, take any arm of this newly-formed cluster, and bind it to another arm uniformly chosen among all the available ones. Go on until no more arms are available in the cluster, then start over with any other vertex. It is important to note that, at each step, the choice of the first vertex or arm really is arbitrary, in that we may choose them according to any random or deterministic rule.

2.3.3 Giant component

In this paragraph we suppose that the following (obviously very similar to Assumption 2.1) is in force.

Assumption 2.5. \mathbb{D}_N converges to μ sharply.

Under this assumption, the existence of a giant component (that is, a connected component C_N of size of order N , i.e. $|C_N| = \Theta(N)$) is, asymptotically as $N \rightarrow \infty$, equivalent to $\zeta' = \langle \hat{\mu}, k \rangle > 1$ (see [42] or [26]). On the other hand, when $\zeta' < 1$, Janson [25] established a bound on the size of the largest connected component.

Theorem 2.6 (Janson [25]). *Consider the random graph $CM(N, D_N)$ with largest component denoted by \mathcal{C}_{\max} . Suppose that \mathbb{D}_N satisfies Assumption 2.5 and that*

$$\mathbb{D}_N([k, \infty)) = O(k^{-\gamma}),$$

uniformly in N and $k \geq 1$, for some $\gamma > 2$. Then there exists a constant A such that, w.h.p.,

$$|\mathcal{C}_{\max}| \leq AN^{1/\gamma}.$$

As we will see further (Section 5.1), our assumption that μ possesses 3 moments ensures that a limiting point π_t of $\pi_t^{(N)}$ possesses as well 3 moments. The above result will thus be very useful to ensure that while π_t is subcritical, then asymptotically almost surely no cluster falls into the gel, since we take $\alpha(N) \gg N^{1/3}$.

2.4 A modified Smoluchowski's equation

We define here the modified Smoluchowski equation which will turn out to be the limit of our model. Let us first introduce some notations. Define $S = \mathbb{N} \times \mathbb{N}^*$. For $p = (a, m), p' = (a', m') \in S$, let $p \cdot p' = aa'$ and $p \circ p' = (a + a' - 2, m + m')$. We write $p' \preceq p$ if $a' \leq a + 1$ and $m' \leq m - 1$. If $p \preceq p'$, we define $p' \setminus p = (a + 2 - a', m - m') \in S$. We also define the modified Smoluchowski equation,

$$c_t(p) = c_0(p) + \int_0^t \frac{1}{\langle c_s, a \rangle} \left(\frac{1}{2} \sum_{p' \preceq p} p' \cdot (p \setminus p') c_s(p') c_s(p \setminus p') - \sum_{p' \in S} p \cdot p' c_s(p) c_s(p') \right) ds, \quad (4.8)$$

⁵Actually, a slight issue arises when K_n is odd. For instance, say we always start with the particle 1. Then all of its arm will eventually be bound. However, a uniform pairing of the arms has a positive probability to leave an arm of 1 unpaired. A way to solve this is to first remove an arm chosen uniformly at random – which is the arm which would remain unpaired in a uniform pairing, and then start the process. Obviously, when K_n is large, this issue becomes irrelevant, and we will ignore it most of the time.

for $p \in S$. It obviously resembles Equation (4.2), and we will explicit the relation between the two in Section 2.5.1. Let us now define what we call a solution to this equation.

Definition 2.7. We say that a family of nonnegative continuous functions $(c_t(p), p \in S)_{t \geq 0}$ is a solution to (4.8) if there is a time $T \in [0, +\infty]$ such that

- $\inf_{s \in [0, t]} \langle c_s, a \rangle > 0$ for all $t < T$ and $\langle c_t, a \rangle = 0$ for $t \geq T$,
- $\sup_{s \in [0, t]} \langle c_s, a \rangle < +\infty$ for all $t > 0$,
- (c_t) verifies (4.8) on $[0, T)$.

The time T is just a technical device, and we will see below that either $T = 0$ and the solution is trivial, or $T = +\infty$.

The relation between these two equations is quite obvious when we think about microscopic models. The standard equation (4.2) is indeed obtained by setting exponential clocks on *pairs of arms*, and this one is obtained, when, as in our model, we set exponential clocks on *arms*. It should then be intuitively obvious that the solutions to these equations are related only by a time-change, as Theorem 2.8 below shows.

2.5 Statement of results

2.5.1 Well-posedness of the modified Smoluchowski equation

Due to the similarity between Equations (4.2) and (4.8), using the techniques developed in the previous chapter, there is no difficulty in checking that (4.8) has a unique solution which is just a time-change of the solution to (4.2). Hence, we shall not prove this result.

For initial conditions $(c_0(p), p \in S)$ with $\langle c_0, a \rangle > 0$, we let

$$\zeta' = \frac{\langle c_0, a(a-1) \rangle}{\langle c_0, a \rangle}$$

and

$$T'_{\text{gel}} = -\ln \left(1 - \frac{1}{\zeta'} \right)$$

if $\zeta' > 1$, and $+\infty$ else. Note that $T'_{\text{gel}} = T_{\text{gel}}$ for monodisperse initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$. Then, we have the following result.

Theorem 2.8. *For any initial conditions (c_0) with $\langle c_0, a^2 \rangle < +\infty$, the modified Smoluchowski equation (4.8) has a unique solution c on \mathbb{R}^+ . It enjoys the following properties.*

1. *The mass $\langle c_t, m \rangle$ is continuous on \mathbb{R}^+ , constant before time T'_{gel} , decreasing afterwards. In other words, there is gelation at time T'_{gel} .*
2. *The total number of arms $A_t = \langle c_t, a \rangle$ is continuous on \mathbb{R}^+ , analytic on $\mathbb{R}^+ \setminus \{T'_{\text{gel}}\}$, and remains positive for all $t \geq 0$ if $A_0 > 0$.*
3. *If $A_0 > 0$, the ODE $s'(t) = A_{s(t)}$, $s(0) = 0$, has a unique solution on \mathbb{R}^+ . This solution is a diffeomorphism of \mathbb{R}^+ and $c_{s(t)}$ is the solution to Smoluchowski's equation (4.2) with initial conditions (c_0) .*
4. *For b a solution to (4.2) and $B_t = \langle b_t, a \rangle$ with $B_0 > 0$, the ODE $s'(t) = 1/B_{s(t)}$, $s(0) = 0$, has a unique solution on \mathbb{R}^+ . This solution is a diffeomorphism of \mathbb{R}^+ and $b_{s(t)}$ is the solution to the modified Smoluchowski's equation (4.8) with initial conditions (c_0) .*

5. The limiting concentrations $c_\infty(p) := \lim_{t \rightarrow +\infty} c_t(p)$ exist and are the same as for Smoluchowski's equation (4.2).

This result means that a solution to (4.8) is obtained just through a change of time of a solution to (4.2). This change of time is a diffeomorphism of \mathbb{R}^+ , and in particular, the final states of both equations coincide. Hence, even though we were initially interested in giving a microscopic model for (4.2), we shall rather do so for (4.8).

Finally, we recall that a representation formula for the solutions to (4.2) is given in the previous chapter (see Theorem 4.3), which allow to compute various quantities concerning (4.8). However, using the formulas of [44] and applying the time-change described above leads to painful calculations. To obtain formulas for (4.8), it would actually easier to mimic the proof of Theorem 4.3 and compute them directly.

2.5.2 Convergence to Smoluchowski's equation

Consider the concentration $c_t^{(N)}(a, m)$ of clusters with a available arms and mass m at time t , in symbols

$$c_t^{(N)}(a, m) = \frac{1}{mN} \#\{i \in [N], |\mathcal{C}_t(i)| = m, \sum_{j \in \mathcal{C}_t(i)} B_t(j) = a\}.$$

Obviously, Assumption 2.1 ensures that, for large enough M , all the $c_t^{(N)}$ belong to

$$E = \{\eta \in [0, 1]^S, \sum_{(a, m) \in S} (a + m)\eta(a, m) \leq M\}.$$

This space is endowed with the ℓ^1 norm.

For the sake of consistency, the first result to check is that these concentrations actually solve, in the limit, Smoluchowski's equation. This is essentially achieved by using classical martingale techniques, as in [27, 45, 17], the delicate issue being the cut-off at threshold $\alpha(N)$, which can be dealt with as in [19]. For that reason, in Section 3, we will only compute the jump rates of $c^{(N)}$ and point out the differences with the case of [19], leaving the very slight changes to the reader.

Theorem 2.9. *The sequence $(c^{(N)}(p), p \in S)_{N \geq 1}$ converges in $\mathbb{D}(\mathbb{R}^+, E)$ to the unique solution of the modified Smoluchowski equation (4.8), starting from monodisperse initial conditions $c_0(a, m) = \mu(a)\mathbb{1}_{\{m=1\}}$.*

2.5.3 Pre-gelation results

The following pre-gelation results are easy to prove, since before gelation, our model essentially behaves like the well-known configuration model. We shall prove this result in Section 4. Recall that, for a law λ and $p \in [0, 1]$, the law $\text{Bin}(\lambda, p)$ is obtained by picking a variable X with law λ , and, conditionally on X , a binomial variable with parameters X and p , and that $T_{\text{gel}}^{(N)}$ is the first time when a giant cluster appears in the system, viz.

$$T_{\text{gel}}^{(N)} = \inf\{t \geq 0, \exists i \in [N] |\mathcal{C}_t(i)| > \alpha(N)\}.$$

Proposition 2.10. *1. For any $t < T_{\text{gel}}^{(N)}$, the configuration of particles has the law of a configuration model with parameters N and $\text{Bin}(\mu^{(N)}, 1 - e^{-t})$, conditioned on having no giant cluster. Moreover,*

$$\text{Bin}(\mu^{(N)}, 1 - e^{-t}) \xrightarrow[N \rightarrow \infty]{} \text{Bin}(\mu, 1 - e^{-t})$$

sharply. Finally, as $N \rightarrow +\infty$, the conditioning becomes trivial, in that the probability that a giant cluster exists tends to 0.

2. When $\zeta \leq 1$, there is no event of gelation w.h.p. More precisely,

$$\mathbb{P}(T_{\text{gel}}^{(N)} = \infty) \xrightarrow{N \rightarrow \infty} 1.$$

3. When $\zeta > 1$,

$$T_{\text{gel}}^{(N)} \xrightarrow{N \rightarrow \infty} T_{\text{gel}}$$

in probability.

In particular, this proves that T_{gel} really is the first time that a cluster falls into the gel. One may however argue that a better definition for T_{gel} would be the first time that a positive fraction of the particles falls into the gel, but, as Theorem 2.11 shows, these two times actually coincide.

2.5.4 Post-gelation results

In the interesting case when $\zeta > 1$, the analysis becomes much harder after the gelation time. Our main result is the convergence of $(n^{(N)})_{N \geq 1}$ and $(\pi^{(N)})_{N \geq 1}$ to a deterministic limit, for which we give an explicit expression. Recall that for a probability λ , G_λ denotes its generating function, and $\nu = \hat{\mu}$ is the size-bias of μ .

Theorem 2.11. 1. The sequence $(n^{(N)})$ converges in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$ as $N \rightarrow +\infty$ to some continuous n , and the sequence $(\pi^{(N)})$ converges in $\mathbb{D}(\mathbb{R}^+, \mathcal{M}_1)$ as $N \rightarrow +\infty$ to some continuous π . Moreover, for $t < T_{\text{gel}}$,

$$\pi_t = \text{Bin}(\mu, 1 - e^{-t}),$$

and for $t \geq T_{\text{gel}}$, π_t is critical.

2. For $t > T_{\text{gel}}$, the equation

$$(\sigma(t) - e^{-t})G'_\nu(\sigma(t)) = G_\nu(\sigma(t)) \quad (4.9)$$

has a unique solution. If we set $\sigma(t) = 1$ for $t \leq T_{\text{gel}}$, then σ is continuous and $\sigma(t) < 1$ for $t > T_{\text{gel}}$.

3. For all $t \geq 0$, the mass in solution is given by

$$n_t = G_\mu(\sigma(t)) \quad (4.10)$$

and π_t has generating function

$$G_{\pi_t}(x) = \frac{1}{n_t} G_\mu(\sigma(t)x + e^{-t}(1-x)), \quad x \in [0, 1]. \quad (4.11)$$

This is our main result, and its proof will take up the main part of this chapter. It relies on several intermediary result, which are interesting on their own, and which we will describe in Section 2.7. The relation between them, as well as the guideline of the proofs, will be given in Section 2.8.

In particular, we may obtain the limiting quantities as $t \rightarrow +\infty$. As will be explained in Section 2.6.4, this provides a probabilistic interpretation for the expression of the limiting concentrations (4.5) obtained in [44], thus achieving our initial goal. Recall from (4.4) that η is the unique solution to

$$G_\nu(\eta) = \eta G'_\nu(\eta).$$

Corollary 2.12. *As $t \rightarrow +\infty$, $\sigma(t) \rightarrow \eta$. Hence, the mass converges to a limit*

$$n_\infty = G_\mu(\eta) \quad (4.12)$$

and the limiting distribution of arms in solution converges to a critical probability π_∞ with generating function

$$G_{\pi_\infty}(x) = \frac{G_\mu(\eta x)}{G_\mu(\eta)}, \quad x \in [0, 1]. \quad (4.13)$$

2.6 Discussion of the results

2.6.1 Before gelation

What happens at a time $t < T_{\text{gel}}^{(N)}$ is rather easy to understand: if the particle i started with a_i arms, its distribution of active arms at time t is $\text{Bin}(a_i, 1 - e^{-t})$. As $N \rightarrow \infty$, the distribution of the number of arms of the particles has a limit $\text{Bin}(\mu, 1 - e^{-t})$, whose size-bias has mean

$$\zeta_t = \zeta(1 - e^{-t}),$$

where we recall that $\zeta = \langle \nu, k \rangle$. Moreover, the cluster configuration is a uniform random pairing of these arms. By Janson's result, and since we took $\alpha(N) \gg N^{1/\gamma}$, it will be easy to see that, with probability going to one as $N \rightarrow \infty$, there exists no giant cluster as long as $\zeta_t < 1$, while we must already have seen a particle fall as soon as $\zeta_t > 1$. A consequence is that $T_{\text{gel}}^{(N)}$ has a deterministic limit as $N \rightarrow \infty$, which is precisely such that $\zeta_{T_{\text{gel}}} = 1$.

2.6.2 Relation to Smoluchowski's equation

Let us discuss the link between the asymptotics of our model and the continuous model given by Smoluchowski's equation (4.8). As we saw, we prove, on the one hand, that the concentrations $c_t^{(N)}(a, m)$ converge to a solution to Smoluchowski's equation (4.8). On the other hand, we obtain the convergence of some relevant quantities of our model, namely $n_t^{(N)}$ and $\pi_t^{(N)}$. However, it is not mathematically obvious that, for instance, the mass

$$M_t := \sum_{a \geq 0, m \geq 1} m c_t(a, m)$$

in Smoluchowski's equation should coincide with the asymptotic mass

$$n_t = \lim_{N \rightarrow +\infty} n_t^{(N)} = \lim_{N \rightarrow +\infty} \sum_{a \geq 0, m \geq 1} m c_t^{(N)}(a, m).$$

We give in Theorem 2.11 a formula for n_t , and, as noted after Theorem 2.8, a formula for M_t can be computed, and it turns out that the two formulas coincide. Hence, we have proved, as was obviously to be expected, that $n^{(N)} \rightarrow M$ in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$. However, as we shall see, getting there is quite a long road. Moreover, we will have to use subtle characteristics of the model (e.g. the graph structure), which are not encoded in the knowledge of the jump rates of $c_t^{(N)}(a, m)$. Proving directly that $n^{(N)} \rightarrow M$ (or even the tightness of $(n^{(N)})$) seems to be quite a challenging question.

Let us end with an intriguing remark on the formulas that can be obtained for (4.8). In fact, though there is only an easy time-change between (4.8) and (4.2), the formulas for the former are much nicer. For instance, we can obtain

$$A_t := \langle c_t, a \rangle = A_0(\sigma'(t) + e^{-t})G_\nu(\sigma(t)),$$

which is worth comparing to the dreadful expression given in Theorem 4.3 of Chapter 3 for (4.2). It is probably even more striking for the mass, but we let the courageous reader compute it with the formulas of Chapter 3 and compare it to (4.10).

2.6.3 State after gelation and self-organized criticality

Knowing the mass and the law of the number of activated arms of the particles in solution, we can draw a quite simple picture of the state in solution. As we will see below in Lemma 5.4, the configuration of particles in solution is a configuration model $CM(n_t^{(N)}N, \pi_t^{(N)})$ conditioned on having no giant cluster. Though this conditioning may not be trivial, we will see (cf. in particular Lemma 5.6) that essentially, a typical cluster in solution can be described as for the configuration model (see Section 3.3.2 in the introduction), that is, if one picks a particle in solution uniformly at random, then it is the root of a Galton-Watson $\mathbb{GW}_{\pi_t, \hat{\pi}_t}$ tree. After gelation, π_t is critical, which is really showing that our model exhibits a phenomenon of *post-gelation self-organized criticality*.

Let us add a few words on these trees in solution. If we allowed links between the particles in the gel and in solution, we would be dealing at time t with a configuration model with parameters N and $\mu_t^{(N)} := \text{Bin}(\mu^{(N)}, 1 - e^{-t})$, which is close, for large N , to $\mu_t := \text{Bin}(\mu, 1 - e^{-t})$. Bertoin and Sidoravicius [4] have shown in that case that a typical cluster in solution is a $\mathbb{GW}_{\mu_t, \hat{\mu}_t}$ -tree conditioned on being finite⁶. Hence, except at the gelation time when μ_t is critical, these *finite* trees are subcritical Galton-Watson trees – recall that a supercritical Galton-Watson tree conditioned on being finite is subcritical.

In the present case, we show that a typical cluster in solution is a $\mathbb{GW}_{\pi_t, \hat{\pi}_t}$ tree, which are thus critical Galton-Watson trees after gelation. In particular, after gelation, *the clusters in solution are much larger in Smoluchowski's model than in Flory's*.

2.6.4 The limiting concentrations

Let us now consider the final state of our system. First, note that Formulas (4.7) and (4.12) agree. Now, as already noted above, a typical particle in solution can be thought of as the root of a $\mathbb{GW}_{\pi_\infty, \hat{\pi}_\infty}$ -tree. So let us argue as in Section 3.3.2 of the introduction, by defining $p(m)$, the probability that a particle, picked uniformly at random, is in a cluster of size m in the final state. On the one hand, $p(m) = mc_\infty(m)$ by definition. On the other hand, this is also the probability that a Galton-Watson tree $\mathbb{GW}_{\pi_\infty, \hat{\pi}_\infty}$ has size m . Recalling from Corollary 2.12 that π_∞ has generating function $G_{\pi_\infty}(x) = G_\mu(\eta x)/G_\mu(\eta)$, it is easy to use Dwass' [9] formula to show that this probability is

$$\frac{1}{m-1} \beta^{m-1} \nu^{*m}(m-2).$$

We have therefore recovered (4.5), thus achieving our initial goal.

2.6.5 The Poisson case

To further interpret the results it is useful to consider the simple case when one starts with an initial distribution of arms that is Poisson. More precisely, let $\lambda > 1$ and

$$\mu(k) = e^{-\lambda} \lambda^k / k!.$$

⁶Actually, this is proved only when μ_t is (sub)critical, but it is implicit in their work that it still holds when μ_t is supercritical.

The Poisson case is closely related to Erdős-Rényi's random graph model, since Erdős-Rényi's model with connection probability λ/n is asymptotically the same as a configuration model with $\text{Poisson}(\lambda)$ distribution. In particular, assuming $\lambda > 1$ corresponds to assuming that gelation will occur, what can be seen also by noticing that $\hat{\mu} = \mu$.

When $\lambda > 1$, gelation occurs at time

$$T_{\text{gel}} = \log\left(\frac{\lambda}{\lambda - 1}\right).$$

Not surprisingly, this goes to ∞ as $\lambda \rightarrow 1$ (in that case the total number of arms is barely supercritical, therefore it takes almost all arms to be paired before gelation can occur). On the other hand it goes to 0 when $\lambda \rightarrow \infty$ (a contrario, the mean number of total arms for a particle is then very large, and it only takes a very small proportion of those arms to be paired to one another to allow formation of large clusters).

The concentration n_t of particles in solution is of course 1 before gelation occurs, while, for $t \geq T_{\text{gel}}$, it is

$$n_t = \sum_{n,k \geq 1} p_t(n, k) = \exp(\lambda(e^{-t} + \lambda^{-1} - 1)).$$

We can also compute the solution given in Theorem 2.11 and obtain for $t \geq T_{\text{gel}}$:

$$\sigma(t) = \lambda^{-1} + e^{-t},$$

We can then deduce, that for $t < T_{\text{gel}}$, π_t is $\text{Poisson}(\lambda(1 - e^{-t}))$, and for any $t \geq T_{\text{gel}}$ it remains $\text{Poisson}(1)$.

It is interesting to note that in that case, the final microscopic state is already reached at T_{gel} . Of course, particles keep falling into the gel for $t > T_{\text{gel}}$, so that on the time interval $]T_{\text{gel}}, +\infty[$, the proportion of particles in solution n_t decreases from 1 to $e^{1-\lambda}$. However, microscopic quantities have reached their final state: the number of already connected arms of a particle still in solution is $\text{Poisson}(1)$, and a typical cluster in solution is a Galton-Watson tree with offspring distribution $\text{Poisson}(1)$. The SOC is thus probably even more striking in this case.

2.7 Intermediate results

The proofs of the post-gelation results are quite lengthy, and take up the main part of this paper. To make the reading easier, let us highlight the main steps of the proofs.

2.7.1 Results at a fixed time

A first combinatorial result is the exact law of the configuration in solution at any time $t \geq 0$.

Lemma 2.13. *At any time $t \geq 0$, conditionally on N_t and $\pi_t^{(N)}$, the state in solution has the same distribution as a configuration model $CM(N_t, \pi_t^{(N)})$, conditioned on having no giant cluster.*

The conditioning in this result is not so easy to deal with. Actually, if $\pi_t^{(N)}$ is subcritical, the conditioning has, by Janson result, probability tending to one, and thus does not play any significant role. When $\pi_t^{(N)}$ is supercritical, its probability tends to 0, so it really matters. Finally, when $\pi_t^{(N)}$ is critical, it is not easy to tell, and it may depend a lot on $\alpha(N)$ (see in particular Th. 2.4. in [26]). As already stated, this last case is actually the one we are in.

At a fixed time, it is also easy to obtain the tightness of any relevant quantity.

Lemma 2.14. • For any $t \geq 0$, the sequences of variables $(n_t^{(N)})$, $(\pi_t^{(N)})$, $(\rho_t^{(N)})$ and $(\xi_t^{(N)})$ are tight.

- The convergence (along some subsequence) of any of the above quantities implies its sharp convergence.

2.7.2 Subcriticality

A first important result is the asymptotic subcriticality of the model, which is a first step towards the tightness.

Proposition 2.15. For any $t \geq 0$

$$\limsup_{N \rightarrow +\infty} \langle \rho_t^{(N)}, k \rangle \leq 1, \quad \text{a.s.}$$

This is essentially achieved by showing that the number of configurations which give rise to a supercritical distribution of active arms in solution is much less than the number of configurations which give rise to a subcritical distribution.

2.7.3 Tightness

The fact that at some time t the model is subcritical allows us to say, that for a small s , it can be at most slightly supercritical at time $t + s$. This ensures that only a proportion of order s of the mass can disappear into the gel on that time-interval. In formulas, we show the following.

Proposition 2.16. For every compact subset K of $(0, +\infty)$, there is a constant C , depending only on this compact and μ , such that, for all $t \in K$ and $s \leq 1$,

$$\limsup_{N \rightarrow +\infty} \mathbb{E} \left(\frac{n_t^{(N)} - n_{t+s}^{(N)}}{n_t^{(N)}} \right) \leq Cs.$$

A similar result concerns the variance of the increments.

Proposition 2.17. For any $t, s \geq 0$,

$$\limsup_{N \rightarrow +\infty} \mathbb{V} \left(n_t^{(N)} - n_{t+s}^{(N)} \right) = 0.$$

These results allow us to estimate a modulus of continuity for $(n^{(N)})$, thus implying its tightness. Proposition 2.17 also shows that any limit point is deterministic, and Proposition 2.16 ensures that it is also Lipschitz-continuous. The tightness of any other quantity is then easy to obtain.

Proposition 2.18. • The sequence $(n^{(N)})$ is tight in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$.

- The sequences $(\pi^{(N)})$ and $(\rho^{(N)})$ are tight in $\mathbb{D}(\mathbb{R}^+, \mathcal{M}_1)$. Moreover, the convergence along a subsequence of any of these two quantities implies its sharp convergence.
- The sequence $(\mathbf{n}^{(N)}(k), k \geq 0)$ is tight in $\mathbb{D}(\mathbb{R}^+, \ell^1(\mathbb{N}))$.
- The sequence $(\mathbf{p}^{(N)}(n, k), n, k \in \mathbb{N}^2)$ is tight in $\mathbb{D}(\mathbb{R}^+, \ell^1(\mathbb{N}^2))$.
- Any limit point of $(n^{(N)})$ or $(\mathbf{n}^{(N)}(k))$ is locally Lipschitz.
- Any limit point of $(\langle \rho^{(N)}, k \rangle)$ is continuous.

2.7.4 Self-organized criticality

Our next step is to show that the model cannot be strictly subcritical after gelation, thus showing the self-organized criticality. With the above results, the proof is quite simple. The idea goes as follows. Take ρ a limit point of $\rho^{(N)}$ and let $m(t) = \langle \rho_t, k \rangle$. Assume that for some $t > T_{\text{gel}}$, $m(t) < 1$. Then no more particles can fall into the gel on a small time-interval, so because of new activations of arms, m has to increase on that interval. Hence, whenever m is below 1, it increases. But $m(T_{\text{gel}}) = 1$, so, along with the continuity of m of the previous proposition, this readily implies that $m(t) = 1$ after gelation.

Proposition 2.19. *For $t \geq T_{\text{gel}}$,*

$$\lim_{N \rightarrow +\infty} \langle \rho_t^{(N)}, k \rangle = 1.$$

2.7.5 Asymptotic independence

Before stating the next result, let us try to draw the heuristic picture of what occurs between a given time t past gelation and a time $t + \varepsilon$. At t we are essentially dealing with a configuration model with a degree sequence that is close to critical. Typical clusters are thus of finite size. Therefore, for any ε , a typical cluster *which is going to be involved in a gelation event in the time window $[t, t + \varepsilon]$* is also of finite size⁷ at time t , in particular, it is typically a finite tree. This then allows us to show a form of asymptotic independence between the evolution of the size of the different subcomponents attached to a given cluster in solution at time t .

Denote now $\tilde{\mathbb{P}}_t^{(k)}$ the probability knowing that particle 1 is in solution at time t and has k activated arms. Obviously, 1 has no particular role, and could be replaced by any $i \in [N]$. Recall that $i \overset{t}{\leftrightarrow} j$ means that i and j are neighbors at time t , and let, for $i \overset{t}{\leftrightarrow} j$, $\mathcal{C}_t^{\setminus j}(i)$ be the cluster of i when deleting the link(s) between i and j . Finally, let $\beta(N)$ be a sequence such that $1 \ll \beta(N) \ll \alpha(N)$.

Proposition 2.20. *For any $t \geq T_{\text{gel}}$, $k \geq 1$,*

$$\lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)} \left(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N), \#\{i : i \overset{t}{\leftrightarrow} 1, |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(i)| < \alpha(N) - \beta(N)\} \neq 1 \right) = 0.$$

In words, this means that, if 1 falls into the gel on the time-interval $[t, t + \varepsilon]$, then that is because one and only one of the subclusters to which it is attached has reached a large size $\alpha(N) - \beta(N)$. Hence, we may write

$$\tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| \geq \alpha(N)) = \sum_{i \overset{t}{\leftrightarrow} 1} \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_t^{\setminus 1}(i)| \geq \alpha(N) - \beta(N)) + o(\varepsilon). \quad (4.14)$$

This result is particularly important when we consider the *instantaneous rate of gelation*. It is defined as the rate at which a particle with k activated arms falls into the gel in a small time-interval, in symbols:

$$f_k(t) := \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \lim_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| \geq \alpha(N)).$$

The limit in N has to be understood along a subsequence such that $(\mathbf{n}^{(N)})$ converges. This is then easy to see that this quantity is well-defined a.e. thanks to the Lipschitz continuity of a limit point $\mathbf{n}(k)$ of $(\mathbf{n}^{(N)}(k))$.

⁷In fact, when $\varepsilon \rightarrow 0$ it is typically of a size of order ε^{-2} .

Now, it is not hard to see that, when 1 is in solution at time t and $i \xrightarrow{t} 1$,

$$\lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \lim_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}^1(i)| \geq \alpha(N) - \beta(N)) = f_1(t),$$

still along a subsequence. Hence, Formula (4.37) provides the following result.

Corollary 2.21. *For all $t \geq 0$, $f_k(t) = k f_1(t)$.*

Informally, this result ensures that the k clusters attached to a particle with k activated arms behave independently, and thus such a particle has k times more chances to fall into the gel than a particle with only one activated arm.

2.7.6 Asymptotic evolution of the system

All these results allow us to precisely describe the evolution of the relevant quantities of the model. The first equation below is obtained as follows. We work up to convergent subsequences. Denote $f(t) := f_1(t)$, and note that f may a priori be random. Consider a particle in solution at time t , with k activated arms and n initially activable arms. On a time interval $[t, t + dt]$, it may:

- fall into the gel with probability $f_k(t) dt = k f(t) dt$;
- or activate one arm with probability $(n - k)(1 - e^{-dt}) = (n - k) dt$.

Such a particle may also appear if a particle with n initially activable arms and $k - 1$ available arms activates one arm. Any other event, such as two of the above occurring at the same time, or a particle activating more than one arm, is very unlikely (of order $(dt)^2$).

Proposition 2.22. *1. Any limit point $(\mathbf{p}_t(n, k), 0 \leq k \leq n)_{t \geq 0}$ of $(\mathbf{p}^{(N)})$ enjoys the relation*

$$\frac{d}{dt} \mathbf{p}_t(n, k) = (-k f(t) - (n - k)) \mathbf{p}_t(n, k) + (n - k + 1) \mathbf{p}_t(n, k - 1).$$

2. In particular, the generating function

$$h_t(x, y) = \sum_{k, n \geq 0} \mathbf{p}_t(n, k) x^k y^n, \quad x, y \in [0, 1),$$

solves the PDE

$$\frac{\partial h_t}{\partial t} = x(1 - f(t) - x) \frac{\partial h_t}{\partial x} + (x - 1)y \frac{\partial h_t}{\partial y}, \quad (4.15)$$

with initial condition $h_0(x, y) = G_\mu(y)$.

This PDE is easy to solve using the method of characteristics.

Proposition 2.23. *The PDE (4.15) has a unique solution given, for $x, y \in [0, 1)$, by*

$$h_t(x, y) = G_\mu \left(y e^{-t} (1 + x \theta(t) / \theta'(t)) \right), \quad (4.16)$$

where

$$\theta(t) = \int_0^t \exp \left(- \int_0^s (1 - f(r)) dr \right) ds.$$

This would end our proof, if we were to actually know $f(t)$. But a priori, it may even be random. However, an elegant way to recover it is to see that it is only defined by the criticality of our model. In other words, writing, for π a limit point of $(\pi^{(N)})$, $\langle \hat{\pi}, k \rangle = 1$ provides an equation for f which has a unique solution. Recall that σ is defined by Equation (4.9).

Corollary 2.24. *For all $t \geq 0$,*

$$f(t) = \frac{\sigma'(t)}{e^{-t} - \sigma(t)}. \quad (4.17)$$

This is finally our last step. Note indeed that this determines f uniquely, and thus shows that the PDE (4.15) has a unique (deterministic) solution. Since any relevant quantity, say n_t and π_t , can be derived upon knowing its solution, and hence $\mathbf{p}_t(n, k)$, we see that there is a unique limit point to any quantity of interest, and so our proof is over.

2.8 Organization of the proofs

Let us now explain how the rest of this chapter is organized. The main part of this paper is devoted to proving Theorem 2.11, and the following list can serve as a reference to find one's way in its proof.

- Let us recall that we shall not prove the well-posedness of the modified Smoluchowski equation (4.8) (Theorem 2.8), for it can be tackled as in the previous chapter.
- The two next sections deal with two easy results: Section 3 sketches of proof of the convergence to Smoluchowski's equation stated in Theorem 2.9, whereas in Section 4, we prove Proposition 2.10 concerning the pre-gelation behavior of our system, what is quite easy with Janson's result Theorem 2.6.
- We then plunge into the core of this paper, namely the proof of the post-gelation results, summarized in Theorem 2.11. We start in Section 5 by giving some important but easy lemmas that we will use a lot throughout the text.
- The first important result, namely the subcriticality of the model (Proposition 2.15) is proved in Section 6. This uses the construction of the exploration process of a configuration model, which is detailed in Appendix A.
- The longest proof concerns the tightness of several processes, as is stated in Proposition 2.18. It is dealt with in Section 7, though several technical issues are deferred to Appendices B, C and D.
- With the help of these results, showing the actual post-gelation SOC (Proposition 2.19) is an easy matter which is taken care of in Section 8.
- The last important result is the asymptotic independence, which mainly consists in proving Proposition 2.20, as is done in Section 9. To avoid more technical convolutions, some elements of the proof are just hinted at.
- In the final Section 10, we gather up all these results to study the asymptotic evolution in time of the system. It is given by a PDE which is easy to solve. We thus show that any limit point of the tight sequences $(n^{(N)})$ and $(\pi^{(N)})$ are uniquely determined, what ends the proof of Theorem 2.11. Proving Corollary 2.12 is then straightforward.

As will appear in the proofs, this order could hardly be changed. Indeed, subcriticality is necessary to prove tightness, which we need to prove the SOC and the asymptotic independence. The latter allows us to write down the PDE describing the evolution in time of the system, and the former implies that this PDE has a unique deterministic solution.

3 Convergence to Smoluchowski's equation

The goal of this section is to give a sketch of proof of Theorem 2.9, which we now recall.

Theorem 3.1. *The sequence $(c^{(N)}(p), p \in S)_{N \geq 1}$ converges in $\mathbb{D}(\mathbb{R}^+, E)$ to the unique solution of the modified Smoluchowski equation (4.8), starting from monodisperse initial conditions $c_0(a, m) = \mu(a) \mathbb{1}_{\{m=1\}}$.*

Its proof is analogous to the one given in [19], where the authors prove the result for the standard Smoluchowski's equation (4.1). Hence, we will only introduce the main ideas, referring to the aforementioned paper for some details.

Recall that $c_t^{(N)}(p)$ is the concentration of $p = (a, m)$ -clusters in solution, i.e. clusters with a (available) arms⁸ and mass m in solution. Let

$$NA_t^{(N)} = \sum_{m=1}^{\alpha(n) + \infty} \sum_{a=1} a N c_t^{(N)}(a, m)$$

be the total number of arms in solution. We always assume in the following computations that there are at least two arms in solution, else nothing happens. Starting from a time t , these concentrations evolve as follows. First, one has to wait an exponential time with parameter $A_t^{(N)}$ to see an arm in solution activated. Then, another arm in solution is activated after an independent exponential time with parameter $A_t^{(N)} - 1$, and a link is created. At this point, three cases are possible.

- A coagulation occurs between two different p - and p' -clusters, with probability

$$\frac{Np.c_t^{(N)}(p)}{NA_t^{(N)}} \times \frac{Np'.c_t^{(N)}(p')}{NA_t^{(N)} - 1} + \frac{Np'.\eta(p')}{NA_t^{(N)}} \times \frac{Np.c_t^{(N)}(p)}{NA_t^{(N)} - 1} = \frac{2p.p'.c_t^{(N)}(p)c_t^{(N)}(p')}{A_t^{(N)}(A_t^{(N)} - 1/N)}.$$

- A coagulation occurs between two distinct $p = (a, m)$ -clusters with probability

$$\frac{Np.c_t^{(N)}(p)}{NA_t^{(N)}} \times \frac{Np.c_t^{(N)}(p) - a}{NA_t^{(N)} - 1} = \frac{p.pc_t^{(N)}(p)^2}{A_t^{(N)}(A_t^{(N)} - 1/N)} - \frac{1}{N} \frac{p.pc_t^{(N)}(p)}{A_t^{(N)}(A_t^{(N)} - 1/N)},$$

- A link between two arms of the same $p = (a, m)$ -clusters is created with probability

$$\frac{aNc_t^{(N)}(p)}{NA_t^{(N)}} \times \frac{a - 1}{NA_t^{(N)} - 1} = \frac{a(a - 1)c_t^{(N)}(p)}{NA_t^{(N)}(A_t^{(N)} - 1)}.$$

⁸In this section only, "arm" means "activable arm".

In the first two cases, the system jumps to a new state $c_t^{(N)} + \frac{1}{N}\Delta_{p,p'}$, where

$$\begin{cases} \Delta_{p,p'}(p) = \Delta_{p,p'}(p') = -1 & \text{if } p \neq p' \\ \Delta_{p,p'}(p) = -2 & \text{if } p = p' \\ \Delta_{p,p'}(p \circ p') = +1. \end{cases}$$

In the third case, the system jumps to a new state $c_t^{(N)} + \frac{1}{N}\Delta'_p$, where, for $p = (a, m)$,

$$\begin{cases} \Delta'_p(p) = -1 \\ \Delta'_p((a-2, m)) = +1. \end{cases}$$

An important fact to note is that $c^{(N)}$ is not a Markov process since the time between two jumps is not exponential, but it is however a semi-Markov process. This is actually no big trouble, and we can proceed just as in the usual Markov case studied in [19]. First, the tightness (in $\mathbb{D}(\mathbb{R}^+, E)$) is easily obtained as in [19] or [43]. To see that a limit point has to solve Smoluchowski's equation (4.8), we can use the above remarks to write martingales (even though the process is only semi-Markov, see e.g. [30]), and proceed just as in [19], with some easy changes.

4 Pre-gelation results

In this section, we prove Proposition 2.10, which is quite an easy matter when using Theorem 2.6. Let us recall the result.

Proposition 4.1. *1. For any $t < T_{\text{gel}}^{(N)}$, the configuration of particles has the law of a configuration model with parameters N and $\text{Bin}(\mu^{(N)}, 1 - e^{-t})$, conditioned on having no giant cluster. Moreover,*

$$\text{Bin}(\mu^{(N)}, 1 - e^{-t}) \xrightarrow{N \rightarrow \infty} \text{Bin}(\mu, 1 - e^{-t})$$

sharply. Finally, as $N \rightarrow +\infty$, the conditioning becomes trivial, in that the probability that a giant cluster exists tends to 0.

2. When $\zeta \leq 1$, there is no event of gelation w.h.p. More precisely,

$$\mathbb{P}(T_{\text{gel}}^{(N)} = \infty) \xrightarrow{N \rightarrow \infty} 1.$$

3. When $\zeta > 1$,

$$T_{\text{gel}}^{(N)} \xrightarrow{N \rightarrow \infty} T_{\text{gel}}$$

in probability.

Proof. First, note that when $t < T_{\text{gel}}^{(N)}$ we have kept all N particles in solution until time t so the first part of the result follows from Lemma 5.4 below.

Let us now turn to the proof of the convergence of $T_{\text{gel}}^{(N)}$. Consider the following coupling.

- $C_1^{(N)}(t)$ corresponds to the configuration following the evolution of our discrete model until time t (where particles which fall into the gel do not interact anymore with particles which stay in solution).

- $C_2^{(N)}(t)$ is such that it has the same number of initially activable arms, the exponential clocks are exactly the same, but we now allow for pairings of an arm of a particle in solution with an arm of a particle in the gel. In other words, this is Flory's model.

By the construction of a configuration model of Section 2.3, the configuration of $C_2^{(N)}(t)$ is exactly that of a configuration model with parameters N and $\text{Bin}(\mu^{(N)}, 1 - e^{-t})$. Moreover, both configurations $C_1^{(N)}(t), C_2^{(N)}(t)$ exactly coincide at any time $t < T_{\text{gel}}^{(N)}$. Therefore,

$$\mathbb{P}(T_{\text{gel}}^{(N)} > t) = \mathbb{P}((C_2^{(N)}(t))_{\max} < \alpha(N)),$$

where $(C_2^{(N)}(t))_{\max}$ is the largest component of $C_2^{(N)}(t)$.

Let $\pi_t = \text{Bin}(\mu, 1 - e^{-t})$. Then $\pi_t^{(N)} \rightarrow \pi_t$ strongly because of Assumption 2.1. Now, note that $\langle \hat{\pi}_t, k \rangle = (1 - e^{-t})\zeta$, so $\langle \hat{\pi}_t, k \rangle \leq 1 \Leftrightarrow t \leq T_{\text{gel}}$. Hence, according to Janson's theorem 2.6, if $t < T_{\text{gel}}$, $C_2^{(N)}(t)$ has w.h.p. no cluster of size greater than $\alpha(N)$ (since $\alpha(N) \gg N^{1/3}$). On the other hand, if $t > T_{\text{gel}}$, $C_2^{(N)}(t)$ has w.h.p. a giant component, which is in particular of size greater than $\alpha(N)$ for N large enough, since $\alpha(N) \ll N$. This readily yields the expected result. \square

5 Preliminary results

This section is devoted to providing some easy results which we will use throughout the text. To avoid irrelevant distinctions, we will assume from now on that we are in the interesting case when gelation occurs, i.e. $\zeta = \langle \hat{\mu}, 1 \rangle > 1$, so in particular $\mu([3, +\infty)) > 0$.

5.1 Lower- and upper-bounds

The following results ensure that some quantities are bounded above or below, as long as this is the case at time 0. Although they are easy to prove, they are a main ingredient in many of our proofs. For instance, proving that, at any time, a positive fraction of the particles remains in solution ensures that $\pi_t^{(N)}$ has as many moments as μ . Recall also that $\xi_t^{(N)}$ is the empirical distribution of active and available arms of the particles in solution at time t .

Lemma 5.1. *For any $t > 0$ and any compact K of $(0, +\infty)$, there exist positive constants $c_0(t)$, $c_0(K)$, $M(t)$ and $M(K)$ such that w.h.p.*

$$\inf_{s \in [0, t]} n_s^{(N)} \geq c_0(t), \quad \inf_{s \in K} \langle \xi_s^{(N)}(i, j), j \rangle \geq c_0(t), \quad \inf_{s \in K} \langle \pi_s^{(N)}, k \rangle \geq c_0(K), \quad (4.18)$$

and

$$\sup_{s \in [0, t]} \langle \pi_s^{(N)}, k^3 \rangle \leq M(t), \quad \sup_{s \in K} \langle \rho_s^{(N)}, k^2 \rangle \leq M(K). \quad (4.19)$$

In words, there is w.h.p., at any time $t > 0$, a positive concentration in solution, a positive proportion of active arms and a positive proportion of available arms. Also, $\pi_t^{(N)}$ has as many moments as μ and consequently $\rho^{(N)}$ has 2 moments.

Proof. First, note that Assumption 2.1 ensures that there is w.h.p. a positive fraction of particles with, say, k arms for some $k \geq 0$. At any time t , each of these particles also has a positive probability to have activated no arm, and thus to remain in solution, whence the bound on $n^{(N)}$ follows. Similar reasonings give the other lower bounds. The bound on $\langle \pi_s^{(N)}, k^3 \rangle$ stems from the bound on $n^{(N)}$, and implies the bound on $\langle \rho_s^{(N)}, k^2 \rangle$. \square

Remark 5.2. An important corollary to this result is that, at any time t , the measure $\pi_t^{(N)}$ verifies the assumptions of Janson's Theorem 2.6, what will be really useful in some proofs.

5.2 Tightness at a fixed time

Lemma 5.3. • For any $t \geq 0$, the sequences of variables $(n_t^{(N)})$, $(\pi_t^{(N)})$, $(\rho_t^{(N)})$ and $(\xi_t^{(N)})$ are tight.

- Convergence (along some subsequence) of any of the above quantities implies its sharp convergence.

Proof. Obviously, $n_t^{(N)} \leq 1$ for all N , so the tightness of $(n_t^{(N)})$ is clear. To avoid convolutions, let us verify the tightness of $(\pi_t^{(N)})$ (tightness of other quantities can be obtained by a similar reasoning). This amounts to proving that for any $\varepsilon, \varepsilon' > 0$, there is a compact $K \subset \mathbb{R}$ such that, for large enough N ,

$$\mathbb{P}(\pi_t^{(N)}(K) > 1 - \varepsilon) > 1 - \varepsilon'.$$

Now, recall from (4.18) that, with probability greater than $1 - \varepsilon'$ say, $n_t^{(N)} > c_0(t) > 0$ for large enough N . Take $\alpha > 0$ such that $\alpha/\varepsilon < c_0(t)$ and a compact K such that $\mu^{(N)}(K) > 1 - \alpha$ for large enough N , what is possible by convergence of $\mu^{(N)}$ to μ . Now, one has, with probability greater than $1 - \varepsilon'$,

$$\pi_t^{(N)}(K^c) = \frac{1}{N_t} \sum_{k \in K^c} N_t(k) \leq \frac{1}{c_0(t)} \mu^{(N)}(K^c) \leq \frac{1}{c_0(t)} \alpha < \varepsilon$$

whence the result follows.

The second part of the statement readily follows from (4.19) and dominated convergence. \square

5.3 Configuration at a fixed time

We shall first point out some features of our model. Recall that all the randomness is given by a sequence of i.i.d. exponential random variables. At a time t , the state of the system depends only on:

- the arms which are activated,
- the order of these activations.

Note that the precise times of activation of the arms are independent from these two variables.

For a set B of arms, denote $\mathfrak{S}(B)$ the set of orderings of B . Define \mathcal{A}_t the set of activated arms at time t , and, conditionally on \mathcal{A}_t , $\sigma_t \in \mathfrak{S}(\mathcal{A}_t)$ is the ordering of these arms. We then just stated that conditionally on \mathcal{A}_t , σ_t is a uniform variable in $\mathfrak{S}(\mathcal{A}_t)$.

5.3.1 Configuration at the current time

Lemma 5.4. At any time $t \geq 0$, conditionally on N_t and $\pi_t^{(N)}$, the state in solution has the same distribution as a configuration model $CM(N_t, \pi_t^{(N)})$, conditioned on having no giant cluster.

Proof. Let us start by conditioning on the initial distribution of arms and on \mathcal{A}_t . Fix S a set of particles and a set B of activated arms of S . We write \mathfrak{S}_1 for the set of orderings of \mathcal{A}_t which leave exactly S in solution. Then,

$$I_S(t) = S \Leftrightarrow \sigma_t \in \mathfrak{S}_1.$$

Hence, conditioning on $I_S(t) = S$ is conditioning on $\sigma_t \in \mathfrak{S}_1$. But σ_t is uniform on $\mathfrak{S}(\mathcal{A}_t)$ so $\sigma_t | \sigma_t \in \mathfrak{S}_1$ is uniform on \mathfrak{S}_1 . It is obvious that a uniform ordering in \mathfrak{S}_1 induces a uniform pairing of the arms of B conditioned on having no giant cluster, so the result readily follows. \square

A slightly more precise result, but with the same proof, takes into account the activable arms. Recall that $\xi_t^{(N)}$ is the empirical measure of active and available arms at time t .

Lemma 5.5. *At any time $t \geq 0$, conditionally on N_t and $\xi_t^{(N)}$, the state in solution has the same distribution as a uniform pairing of the activated arms, conditioned on having no giant cluster.*

5.3.2 Configuration at an earlier time

As pointed out in the previous section, the conditioning of the previous results may have a probability tending to 0 as $N \rightarrow +\infty$, and hence turn out to be quite tedious to deal with. To circumvent this issue, we shall “go back in time”, by looking at the set $I_S(t)$ of particles which are still in solution at time t , at a time $s < t$. These particles have an empirical distribution of activated arms at time s denoted by

$$\pi_{t,s}^{(N)} = \frac{1}{N_t} \sum_{i \in I_S(t)} \delta_{A_s(i)}.$$

Lemma 5.6. *Conditionally on $I_S(t)$ and $\pi_{t,s}^{(N)}$, the configuration of the particles of $I_S(t)$ at time s has the same distribution as a configuration model $CM(N_t, \pi_{t,s}^{(N)})$ conditioned on having no giant cluster. Moreover, the sequence $(\pi_{t,s}^{(N)})$ is sharply tight and there is an $\varepsilon > 0$ such that, for any of its limit point $\pi_{t,s}$,*

$$\langle \hat{\pi}_{t,s}, k \rangle \leq (1 - \varepsilon) \limsup_{N \rightarrow +\infty} \langle \hat{\pi}_t^{(N)}, k \rangle.$$

Now, when we know that actually $\limsup \langle \hat{\pi}_t^{(N)}, k \rangle \leq 1$, this will show that at time s , the configuration of the particles of $I_S(t)$ is a *subcritical* configuration model conditioned on having no giant cluster. But the subcriticality implies that this conditioning has probability tending to 1, and hence there will be no harm in forgetting about it.

Proof. The fact that it is a conditioned configuration model is proved as in Lemma 5.4 above, and the tightness as in Lemma 5.3. Now, at time s , there is an asymptotically positive proportion (depending only on μ and s) of particles with no active arm and a positive number of available arms at time s . But there is also positive probability (depending only on μ , s and t) for each of them, to, on the time-interval $[s, t]$,

- activate exactly one arm,
- bind to exactly another particle.

These clusters of size 2 will thus be in solution at time t (if $\alpha(N) \geq 2$). In other words, there is a positive fraction (depending only on μ , s and t) of particles in solution at t , with one active arm at t , none at s . The result easily follows. \square

Without further effort, we may also take into account the number of available arms. So let

$$\xi_{t,s}^{(N)} = \frac{1}{N_t} \sum_{i \in I_S(t)} \delta_{(A_s(i), B_s(i))}$$

the empirical distribution of the number of active arms and available arms of the particles of $I_S(t)$ at time s .

Lemma 5.7. *Conditionally on $I_S(t)$ and $\xi_{t,s}^{(N)}$, the configuration of the particles of $I_S(t)$ at time s has the same distribution as a uniform pairing of the active arms conditioned on having no giant cluster. Moreover, the sequence $(\xi_{t,s}^{(N)})$ is sharply tight, and there is an $\varepsilon > 0$ such that, for any of its limit point $\xi_{t,s}$,*

$$\langle \hat{\xi}_{t,s}, i \rangle \leq (1 - \varepsilon) \limsup_{N \rightarrow +\infty} \langle \hat{\xi}_t^{(N)}, i \rangle.$$

6 Subcriticality

6.1 Introduction

Our goal in this paragraph is to establish that our model is subcritical, more precisely the following result.

Proposition 6.1. *For any $t \geq 0$*

$$\limsup_{N \rightarrow +\infty} \langle \rho_t^{(N)}, k \rangle \leq 1, \quad \text{a.s.} \quad (4.20)$$

Now, fix $t \geq 0$, and assume thanks to Lemma 5.3 that $\rho_t^{(N)} \rightarrow \rho_t$ sharply along some subsequence. We will also assume by tightness that $n_t^{(N)}$ converges to some n_t , and we recall from (4.18) that then, $n_t > c_0(t)$ for some $c_0(t) > 0$ depending only on μ and t .

6.2 A preliminary lemma

We shall prove that the following result, which is enough to show that (4.20) holds.

Lemma 6.2. *For any $\delta > 0$ and $u \in [c_0(t), 1]$, there exists $\varepsilon_u > 0$ such that*

$$\lim_{N \rightarrow +\infty} \mathbb{P} \left(\langle \rho_t^{(N)}, k \rangle \geq 1 + 2\delta, n_t^{(N)} \in (u - \varepsilon_u, u + \varepsilon_u) \right) = 0.$$

Before proving the lemma, let us see how it implies the subcriticality of ρ_t . We have

$$\begin{aligned} \mathbb{P} \left(\langle \rho_t^{(N)}, k \rangle \geq 1 + 2\delta \right) &\leq \mathbb{P} \left(n_t^{(N)} \leq c_0(t) \right) \\ &+ \mathbb{P} \left(\langle \rho_t^{(N)}, k \rangle \geq 1 + 2\delta, \bigcup_{u \in [c_0(t), 1]} \{n_t^{(N)} \in (u - \varepsilon_u, u + \varepsilon_u)\} \right). \end{aligned} \quad (4.21)$$

By Borel characterization of compact sets, there exists a finite n and some $u_1, \dots, u_n \in [c_0(t), 1]$ such that $\bigcup_{i=1}^n (u_i - \varepsilon_u, u_i + \varepsilon_u) \supset [c_0(t), 1]$. Thus

$$\begin{aligned} & \mathbb{P} \left(\langle \rho_t^{(N)}, k \rangle \geq 1 + 2\delta, \bigcup_{u \in [c_0(t), 1]} \{n_t^{(N)} \in (u - \varepsilon_u, u + \varepsilon_u)\} \right) \\ & \leq \sum_{i=1}^n \mathbb{P} \left(\langle \rho_t^{(N)}, k \rangle \geq 1 + 2\delta, n_t^{(N)} \in (u_i - \varepsilon_{u_i}, u_i + \varepsilon_{u_i}) \right). \end{aligned}$$

By Lemma 6.2, each term in the above (finite) sum goes to 0 as $N \rightarrow +\infty$, so the second term of the sum in the RHS of (4.21) goes to 0 as $N \rightarrow +\infty$. So does the first since $n_t^{(N)} \rightarrow n_t > c_0(t)$, and therefore,

$$\lim_{N \rightarrow +\infty} \mathbb{P} \left(\langle \rho_t^{(N)}, k \rangle \geq 1 + 2\delta \right) = 0.$$

Since $\langle \rho_t^{(N)}, k \rangle \rightarrow \langle \rho_t, k \rangle$ by sharp convergence, this obviously implies

$$\langle \rho_t, k \rangle \leq 1 + 2\delta, \quad \text{a.s.}$$

This holds for any $\delta > 0$, so the result follows. Hence, it remains to establish the lemma.

6.3 Proof of Lemma 6.2

Recall that $\mathcal{A}_t(i)$ is the set of all active arms of i at time t , and define $\mathcal{A}_t = \bigcup_{i \in [N]} \mathcal{A}_t(i)$ the set of activated arms at time t .

Let us from now on condition on \mathcal{A}_t . Recall from Section 2.1 that then, the configuration of the system at time t is only given by a uniform ordering of the arms of \mathcal{A}_t . We thus take Ω the set of all these orderings, which is endowed with the uniform probability \mathbb{P} .

To a set $S \subset [N]$ of particles, we associate

$$\pi(S) = \frac{1}{\#S} \sum_{i \in S} \delta_{\mathcal{A}_t(i)}$$

the empirical measure of the degree of active arms of S , and $\rho(S) = \hat{\pi}(S)$.

Now, an $\omega \in \Omega$ induces a configuration of the particles, so we can define $I_G(\omega)$ the set of all particles falling into the gel in the configuration induced by ω , $I_S(\omega) = [N] \setminus I_G(\omega)$ the set of all particles remaining in solution, $\pi(\omega) = \pi(I_S(\omega))$ the empirical measure of the degree of active arms of the particles in solution, $\rho(\omega) = \rho(I_S(\omega))$ its size-bias, and $n(\omega) = \#I_S(\omega)/N$ the concentration in solution.

Consider now a set $\mathcal{C}_1, \dots, \mathcal{C}_p$ of disjoint giant clusters. Define

$$\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p} = \left\{ \omega \in \Omega, I_G(\omega) \supset \bigcup_{i=1}^p \mathcal{C}_i \right\},$$

the set of all ω which make $\mathcal{C}_1, \dots, \mathcal{C}_p$ fall into the gel. It can be split into those events for which only $\mathcal{C}_1, \dots, \mathcal{C}_p$ fall into the gel

$$\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^0 := \left\{ \omega \in \Omega, I_G(\omega) = \bigcup_{i=1}^p \mathcal{C}_i \right\},$$

and those events for which there at least another cluster which falls into the gel:

$$\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^{\geq 1} := \left\{ \omega \in \Omega, I_G(\omega) \supseteq \bigcup_{i=1}^p \mathcal{C}_i \right\}.$$

Now, fix $\delta > 0$ and $u \in [c_0(t), 1]$. Since the sequence $(\langle \mu^{(N)}, k^2 \rangle)_{N \geq 1}$ is bounded, we may choose $\varepsilon > 0$, depending only on μ and δ , such that, for any set S of particles with $\#S \geq (u - \varepsilon)N$, for any set $S' \subset S$ with $\#S' \leq 2\varepsilon N$, we have

$$|\langle \rho(S \setminus S'), k \rangle - \langle \rho(S), k \rangle| \leq \delta.$$

Finally, define

$$\mathcal{J} = [N] \setminus \bigcup_{i=1}^p \mathcal{C}_i.$$

We shall first establish the following.

Lemma 6.3. *Assume that*

$$n(\mathcal{J}) \in (u - \varepsilon, u + \varepsilon), \quad \langle \rho(\mathcal{J}), k \rangle \geq 1 + \delta.$$

Then, there exists $\kappa > 0$, which depends only on u , ε and δ , and such that, for large enough N ,

$$\mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^0) \leq \exp(-\kappa N) \mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^{\geq 1}).$$

Proof. This result can be restated by saying that

$$\mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^{\geq 1} | \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}) \geq \exp(\kappa N) \mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^0 | \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}).$$

But

$$\mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^0 | \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p})$$

is the probability that there is no giant cluster among the particles of \mathcal{J} , and we assumed that

$$\rho(\mathcal{J}) \geq 1 + \delta.$$

When $\mathcal{C}_1, \dots, \mathcal{C}_p$ fall into the gel, the ordering of the arms of the particles of \mathcal{J} is still uniform, and thus the configuration of the particles of \mathcal{J} is the same as taking a uniform ordering of these arms, and then applying our algorithm (i.e. bind together the first and second arm in solution, the third and fourth in solution, and so on).

In particular, the probability that a cluster falls into the gel is the same as the probability for the configuration model to have a cluster of size greater than $\alpha(N)$. Since we assumed $\langle \rho(\mathcal{J}), k \rangle \geq 1 + \delta$, it is well-known (see Section 2.3) that \mathcal{J} has a component of size of order N w.h.p. However, we need a little more here to get the factor $\exp(-\kappa N)$.

It is actually implicit in the literature (see e.g. the proofs in [26]) that such a model has probability decreasing exponentially fast to have no cluster of order N , and that the parameter of the exponential depends only on $\langle \rho(\mathcal{J}), k \rangle$.

To be more precise, we refer to Appendix A for the definition and properties of the exploration process of a typical component of a configuration model. Proposition A.3 then ensures that the size of the connected component are bounded by below by the lengths of the excursions between infima of a random walk \underline{V} , as long as, in the notations of Appendix A, less than $f(N)$ steps of the exploration process have been taken.

From the construction of \underline{V} , it is clear that for N large and $f(N) \ll N$, the steps of \underline{V} can be chosen in such a way that their mean is greater than $1 + \delta/2$. Now, assume also that

$f(N) \gg \alpha(N)$. It is then classical from the random walk theory (see e.g. [52]) that there is a constant κ depending only on δ such that an excursion of size $\alpha(N)$ appears among the $f(N)$ first steps of the random walk, with probability bounded below by $1 - e^{-\kappa N}$. In other words

$$\mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^{\geq 1} | \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}) \geq 1 - e^{-\kappa n}$$

whence the result follows easily after noticing that

$$\mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^0 | \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}) + \mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^{\geq 1} | \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}) = 1.$$

□

When the distribution in solution is supercritical and $\mathcal{C}_1, \dots, \mathcal{C}_p$ fall into the gel, Lemma 6.3 ensures that there is a high probability that other clusters than $\mathcal{C}_1, \dots, \mathcal{C}_p$ also fall into the gel. But when $2\varepsilon N/\alpha(N)$ additional clusters have fallen into the gel, then the concentration has dropped by at least 2ε (since any cluster which falls into the gel has size greater than $\alpha(N)$). As a result, the concentration is in fact very likely to have already dropped below $u - \varepsilon$. This is the content of the following lemma.

Lemma 6.4. *Assume that*

$$n(\mathcal{J}) \in (u - \varepsilon, u + \varepsilon), \quad \langle \rho(\mathcal{J}), k \rangle \geq 1 + 2\delta.$$

Then, for large enough N ,

$$\mathbb{P}(\omega \in \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}, n(\omega) \in (u - \varepsilon, u + \varepsilon)) \leq \frac{N}{\alpha(N)} \exp(-\kappa N) \mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}).$$

Proof. Define, for $i \geq 1$,

$$\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^i$$

be the set of ω such that $I_G(\omega)$ contains $\mathcal{C}_1, \dots, \mathcal{C}_p$, as well as precisely i other clusters.

For $\omega \in \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^i$ with $i \leq \varepsilon N/\alpha(N)$, one has $0 \leq c(\mathcal{J}) - c(\omega) \leq 2\varepsilon$ (since the clusters which fall into the gel have mass at most $2\alpha(N)$), so,

$$\left\{ \omega \in \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}, c(\omega) \in (u - \varepsilon, u + \varepsilon) \right\} \subset \bigcup_{i=1}^{\varepsilon N/\alpha(N)} \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^i, \quad (4.22)$$

and, by the very choice of ε , $\rho(\omega) \geq 1 + \delta$. By Lemma 6.3, we thus have

$$\mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^i) \leq e^{-\kappa N} \mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^{\geq i+1}). \quad (4.23)$$

If we denote

$$p_i = \mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}^{\geq i})$$

then (4.23) reads

$$p_i - p_{i+1} \leq e^{-\kappa N} p_{i+1}, \quad i \leq \varepsilon N/\alpha(N).$$

From this, it is easy to deduce that

$$p_{\varepsilon N/\alpha(N)} \leq \frac{N}{\alpha(N)} e^{-\kappa N} p_0.$$

Finally, (4.22) and the above result yield

$$\begin{aligned} \mathbb{P}(\omega \in \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}, n(\omega) \in (u - \varepsilon, u + \varepsilon)) &\leq p_{\varepsilon N/\alpha(N)} \\ &\leq \frac{N}{\alpha(N)} e^{-\kappa N} p_0 = \frac{N}{\alpha(N)} e^{-\kappa N} \mathbb{P}(\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}) \end{aligned}$$

and the proof is over. □

We can now finish the proof of Lemma 6.2. Note that the event

$$\{\langle \rho(\omega), k \rangle \geq 1 + 2\delta, n(\omega) \in (u - \varepsilon, u + \varepsilon)\}$$

is included in the union of the events

$$\left\{ \omega \in \Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}, n(\omega) \in (u - \varepsilon, u + \varepsilon) \right\}$$

for all p and giant $\mathcal{C}_1, \dots, \mathcal{C}_p$ such that, for $\mathcal{J} = [N] \setminus \bigcup_{i=1}^p \mathcal{C}_i$, $n(\mathcal{J}) \in (u - \varepsilon, u + \varepsilon)$ and $\rho(\mathcal{J}) \geq 1 + 2\delta$.

By summing over all possibilities such that the $\Omega_{\mathcal{C}_1, \dots, \mathcal{C}_p}$ are disjoint, we thus get from Lemma 6.4 that

$$\mathbb{P}(\langle \rho(\omega), k \rangle \geq 1 + 2\delta, n(\omega) \in (u - \varepsilon, u + \varepsilon)) \leq \frac{N}{\alpha(N)} e^{-\kappa N}$$

and Lemma 6.2 follows, after integrating over \mathcal{A}_t and using dominated convergence.

7 Tightness

7.1 Introduction

The goal of this section is to prove Proposition 2.18, which we recall now.

Proposition 7.1. • *The sequence $(n^{(N)})$ is tight in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$.*

- *The sequences $(\pi^{(N)})$ and $(\rho^{(N)})$ are tight in $\mathbb{D}(\mathbb{R}^+, \mathcal{M}_1)$. Moreover, the convergence along a subsequence of any of these two quantities implies its sharp convergence.*
- *The sequence $(\mathbf{n}^{(N)}(k), k \geq 0)$ is tight in $\mathbb{D}(\mathbb{R}^+, \ell^1(\mathbb{N}))$.*
- *The sequence $(\mathbf{p}^{(N)}(n, k), n, k \in \mathbb{N}^2)$ is tight in $\mathbb{D}(\mathbb{R}^+, \ell^1(\mathbb{N}^2))$.*
- *Any limit point of $(n^{(N)})$ or $(\mathbf{n}^{(N)}(k))$ is locally Lipschitz.*
- *Any limit point of $(\langle \rho^{(N)}, k \rangle)$ is continuous.*

We shall show in particular the tightness of the sequence $(n^{(N)})$. The tightness of the other sequences is then an easy matter, and the rest of the proposition is deduced from 4.19.

The proof relies mainly on the two following results, concerning the expectation and variance of the increments of $(n^{(N)})$, which allow to obtain a modulus of continuity for $(n^{(N)})$ and hence deduce its tightness.

Proposition 7.2. *For every compact subset K of $(0, +\infty)$, there is a constant C , depending only on this compact and μ , such that, for all $t \in K$ and $s \leq 1$,*

$$\limsup_{N \rightarrow +\infty} \mathbb{E} \left(\frac{n_t^{(N)} - n_{t+s}^{(N)}}{n_t^{(N)}} \right) \leq Cs.$$

Proposition 7.3. *For any $t, s \geq 0$,*

$$\limsup_{N \rightarrow +\infty} \mathbb{V} \left(n_t^{(N)} - n_{t+s}^{(N)} \right) = 0.$$

With the help of these results, the tightness is an easy but technical matter, which we defer to Appendix B.

7.2 Main steps of the proof of Proposition 7.2

The proof of Proposition 7.2 is long, and we will break it up in pieces so as not to exhaust the reader. This first section provides the main ingredients and ideas of the proof. Section 7.3 is devoted to giving a bound on the quantity of gel in a two-type uniform pairing. In some loose sense, this quantity bounds the quantity of gel that appears in our model between time t and $t + s$. Finally, most technical matters are deferred to Appendices C and D.

7.2.1 Idea of the proof

An issue that we already raised concerning the state at some time t is the conditioning in Lemma 5.4, and to deal with it, we will “go back in time” as in Lemma 5.7. So fix $t > 0$, $1 > s > 0$ and take (a small) $h \in (0, t)$. We look at the particles of $I_S(t)$ at time $t - h$. At that time, they have active arms, which we call *old* (O), and available arms, which we call *young* (Y), and in the notations of Lemma 5.7 they have an empirical distribution of O and Y arms $\xi_{t,t-h}^{(N)}$. Moreover, we denote $\mathcal{C}_{t,t-h}^{(N)}$ their configuration at time $t - h$, and $\mathcal{C}_{t,t-h+s}^{(N)}$ their configuration at time $t - h + s$.

Now, delete all the links, so we are in the presence of N_t particles with empirical distribution of O and Y arms $\xi_{t,t-h}^{(N)}$. To obtain the configuration of these particles at time $t - h + s$, we can proceed as follows.

1. Condition on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$.
2. Pick a uniform pairing of the O arms, conditioned on having no giant cluster. By Lemma 5.7, this gives precisely the configuration $\mathcal{C}_{t,t-h}^{(N)}$.
3. Activate each Y arm independently after an exponential time with parameter 1. Bind the first two activated arms *in solution*, then the two next, and so on, without forgetting that if a cluster of size greater than $\alpha(N)$ is created, then it falls into the gel. Doing this is just looking at the process that we study. After a time $s + h$, we have thus obtained a configuration which has the same law as $\mathcal{C}^{(N)}$.

Obviously, this model is quite hard to study as such. To make it easier, let us build a graph $\tilde{G}^{(N)}$ as follows.

1. Condition on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$.
2. Pick a uniform pairing of the O arms, conditioned on having no giant cluster.
3. Activate each Y arm independently after an exponential time with parameter 1. Bind the first two activated arms *in solution or not*, then the two next, and so on, until time $s + h$.

Let us finally build a graph $G^{(N)}$, which will be our main interest in the sequel.

1. Condition on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$.
2. Pick a uniform pairing of the O arms.
3. Activate each Y arm independently after an exponential time with parameter 1. Bind the first two activated arms *in solution or not*, then the two next, and so on, until time $s + h$.

This graph is much easier to study, because it is a two-type uniform pairing, in that it can be constructed in the following way. First, condition on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$. Then activate each Y arm independently with probability $1 - e^{-(s+h)}$ and finally choose a uniform pairing of the O arms and independently a uniform pairing of the activated Y arms.

Remark 7.4. It is important to note that, in our constructions, there are two types of randomness. The first is the choice of the number of particles and their number of arms, given by $I_S(t)$ and $\xi_{t,t-h}^{(N)}$. The second is the choice of a pairing of these arms.

7.2.2 Relation between the different models

We have thus defined three different graphs. From Lemma 5.7 and Proposition 6.1, the law of the O arms in $\xi_{t,t-h}^{(N)}$ is subcritical, and Janson's Theorem 2.6 ensures that it is very unlikely to have a large cluster. In particular, the conditioning in the construction of $\tilde{G}^{(N)}$ is asymptotically trivial, so $\tilde{G}^{(N)}$ and $G^{(N)}$ should be really close.

Let us now compare $\tilde{G}^{(N)}$ and $\mathcal{C}^{(N)}$. Loosely speaking, the first corresponds to Flory's evolution, the second to Smoluchowski's. In fact, more mass is lost in Flory's evolution than in Smoluchowski's, as is stated below. For the continuous equations, this is in fact well-known (see in particular [44, Cor. 3.4]). Denote below $\tilde{G}_{\text{gel}}^{(N)}$ and $\mathcal{C}_{\text{gel}}^{(N)}$ the quantity of gel (i.e. the number of particles in giant clusters) in $\tilde{G}^{(N)}$ and $\mathcal{C}^{(N)}$.

Lemma 7.5. *For every $i \in I_S(t)$, one has*

$$\mathbb{P}\left(|\mathcal{C}^{(N)}(i)| > \alpha(N) \mid I_S(t), \xi_{t,t-h}^{(N)}\right) \leq \mathbb{P}\left(|\tilde{G}^{(N)}(i)| > \alpha(N) \mid I_S(t), \xi_{t,t-h}^{(N)}\right).$$

In particular

$$\mathbb{E}\left(\mathcal{C}_{\text{gel}}^{(N)} \mid I_S(t), \xi_{t,t-h}^{(N)}\right) \leq \mathbb{E}\left(\tilde{G}_{\text{gel}}^{(N)} \mid I_S(t), \xi_{t,t-h}^{(N)}\right)$$

and

$$\mathbb{E}\left(\frac{1}{N_t} \mathcal{C}_{\text{gel}}^{(N)}\right) \leq \mathbb{E}\left(\frac{1}{N_t} \tilde{G}_{\text{gel}}^{(N)}\right).$$

The proof of this result is quite long and uses particular techniques, so we shall rather deal with it in Appendix C.

7.2.3 End of the proof

Thanks to these two remarks, we may only study an upper-bound for the quantity of gel in $G^{(N)}$. For a (one-type) configuration model, this can be done by bounding the exploration process of a typical component by a random walk — or equivalently, by the exploration process of a Galton-Watson tree. Since $G^{(N)}$ is a two-type configuration model, it is then natural to try to bound this quantity by the exploration process of a two-type Galton-Watson tree. We will prove in Section 7.3 the following result.

Lemma 7.6. *There is a constant C , uniform for $t - h$ and t in the compacts of $(0, +\infty)$ and depending only on μ , such that*

$$\mathbb{E}\left(\frac{1}{N_t} G_{\text{gel}}^{(N)}\right) \leq C(s + h).$$

Taking this for granted, we can gather up all the results and end the proof. We shall first construct the graphs $\mathcal{C}^{(N)}$, $\tilde{G}^{(N)}$ and $G^{(N)}$ defined at the beginning of this proof. To this end, note that we may define, on a large enough probability space, for each $N \geq 1$,

- the random variables $I_S(t)$ and $\xi_{t,t-h}^{(N)}$,
- conditionally on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$, a set of $N_t = \#I_S(t)$ particles with O and Y arms, whose empirical distribution of O and Y arms is $\xi_{t,t-h}^{(N)}$.

This being given, we can build a uniform pairing $\mathcal{P}^{(N)}$ of the O arms, and conditionally on $\mathcal{P}^{(N)}$, a graph constructed by activating independently each Y arms with probability $1 - e^{-(s+h)}$, and then choosing a uniform pairing of the activated Y arms.

Note that this last graph is precisely (in law) the graph $G^{(N)}$ of Section 7.2.1. If we define E_N the event that $\mathcal{P}^{(N)}$ has no giant cluster, then by construction,

$$\tilde{G}^{(N)} \stackrel{(d)}{=} G^{(N)} | E_N. \quad (4.24)$$

Now, notice that

$$\frac{n_t^{(N)} - n_{t+s}^{(N)}}{n_t^{(N)}} = \frac{1}{N_t} \mathbf{c}_{\text{gel}}^{(N)} \quad (4.25)$$

According to Lemma 7.5,

$$\mathbb{E} \left(\frac{1}{N_t} \mathbf{c}_{\text{gel}}^{(N)} \right) \leq \mathbb{E} \left(\frac{1}{N_t} \tilde{G}_{\text{gel}}^{(N)} \right).$$

and so, by (4.24) and (4.25),

$$\begin{aligned} \mathbb{E} \left(\frac{n_t^{(N)} - n_{t+s}^{(N)}}{n_t^{(N)}} \right) &\leq \mathbb{E} \left(\frac{\tilde{G}_{\text{gel}}^{(N)}}{N_t} \right) \\ &= \mathbb{E} \left(\frac{G_{\text{gel}}^{(N)}}{N_t} \middle| E_N \right) \\ &\leq \frac{1}{\mathbb{P}(E_N)} \mathbb{E} \left(\frac{G_{\text{gel}}^{(N)}}{N_t} \right). \end{aligned}$$

Now, recall that $\mathcal{P}^{(N)}$ is a uniform pairing of the O arms, and that the distribution of the O arms is subcritical. Indeed, by Lemma 5.7 and Proposition 6.1, there is a $\varepsilon > 0$ such that

$$\begin{aligned} \limsup_{N \rightarrow +\infty} \langle \hat{\xi}_{t,t-h}^{(N)}, k \rangle &\leq (1 - \varepsilon) \limsup_{N \rightarrow +\infty} \langle \hat{\xi}_t^{(N)}, k \rangle \\ &= (1 - \varepsilon) \limsup_{N \rightarrow +\infty} \langle \hat{\pi}_t^{(N)}, k \rangle \\ &\leq 1 - \varepsilon, \quad \text{a.s.} \end{aligned}$$

Note that $\langle \hat{\xi}_{t,t-h}^{(N)}, k \rangle$ is precisely the mean of the size-bias of the distribution of O arms. Since $N_t \rightarrow +\infty$, Janson's Theorem 2.6 and Remark 5.2 ensures that w.h.p., $\mathcal{P}^{(N)}$ has no giant cluster, more precisely

$$\mathbb{P}(E_N | I_S(t), \xi_{t,t-h}^{(N)}) \rightarrow 1 \quad \text{a.s.}$$

so by dominated convergence

$$\mathbb{P}(E_N) \rightarrow 1. \quad (4.26)$$

Finally,

$$\mathbb{E} \left(\frac{n_t^{(N)} - n_{t+s}^{(N)}}{n_t^{(N)}} \right) \leq \frac{1}{\mathbb{P}(E_N)} \mathbb{E} \left(\frac{G_{\text{gel}}^{(N)}}{N_t} \right)$$

and so, with (4.26) and Lemma 7.6,

$$\limsup_{N \rightarrow +\infty} \mathbb{E} \left(\frac{n_t^{(N)} - n_{t+s}^{(N)}}{n_t^{(N)}} \right) \leq C(s+h)$$

and having $h \rightarrow 0$ shows the result, after recalling that C only depends on μ and is uniform as long as $t-h$ remains in a compact of $(0, +\infty)$. Finally, all it remains to prove is Lemma 7.6.

7.3 Quantity of gel in a two-type pairing

The goal of this section is to prove Lemma 7.6, i.e. to bound the quantity of gel in $G^{(N)}$, where $G^{(N)}$ is constructed by, taking, conditionally on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$, a uniform pairing of the O arms, and a uniform pairing of the Y arms. This is essentially achieved by bounding above the exploration process of a component of the graph $G^{(N)}$ by the exploration process of a (slightly supercritical) two-type Galton-Watson process, for which it is easy to check that it has little probability of growing too big.

7.3.1 Upper-bound for the exploration process

The proof is achieved by constructing at the same time a typical component of $G^{(N)}$ and its exploration process. The construction is actually quite long and technical, and we shall defer it to Appendix D.

Let us define, for random laws γ, γ^O and γ^Y on $\mathbb{N} \times \mathbb{N}$, the two-type Galton-Watson tree $\mathbb{GW}_{\gamma, \gamma^O, \gamma^Y}$, obtained by first picking the reproduction laws, and then constructing the Galton-Watson tree with reproduction law γ for the ancestor, γ^O and γ^Y for the O and Y individuals. We also define $T(\gamma, \gamma^O, \gamma^Y)$ to be its size.

In Appendix D we will obtain, for each N , random laws γ_N, γ_N^O and γ_N^Y . They enjoy the following properties, where we denote, for a law λ on $\mathbb{N} \times \mathbb{N}$, λ^1 and λ^2 its first and second marginal.

- The laws γ_N, γ_N^O and γ_N^Y are deterministic functions of $I_S(t)$ and $\xi_{t,t-h}^{(N)}$.
- The sequences $(\gamma_N), (\gamma_N^O)$ and (γ_N^Y) are sharply tight.
- If (X, Y) has law γ^Y , then Y has, conditionally on X a binomial law with parameters X and $1 - e^{-(s+h)}$.
- There are constants $a, M > 0$, depending only on μ and uniform for $t-h$ and t in the compacts of $(0, +\infty)$, such that, for any limit points γ, γ^O and γ^Y of these sequences:

$$\langle \gamma^1, k \rangle = \langle \gamma^2, k \rangle \leq M, \quad \langle \gamma_O^1, k \rangle \leq 1, \quad \langle \gamma_O^2, k \rangle \leq M(s+h), \quad (4.27)$$

and

$$\langle \gamma_Y^1, k \rangle \leq M, \quad \langle \gamma_Y^2, k \rangle \leq M(s+h). \quad (4.28)$$

Moreover

$$2\gamma_O(2, 0) \geq a. \quad (4.29)$$

Remark 7.7. These bounds are actually quite natural. The bounds in M essentially mean that there is a positive fraction of O or Y arms on the particles of $I_S(t)$ at time $t-h$, whereas the bounds in $M(s+h)$ stem from the fact that each Y arm is activated independently on the time-interval $[t-h, t+s]$ with probability $1 - e^{-(s+h)} \leq s+h$. In fact, we only use one important result, namely the subcriticality (Proposition 6.1), which ensures that $\langle \gamma_O^1, k \rangle \leq 1$. Hence, the Galton-Watson process with these reproduction laws is slightly supercritical (loosely, the mean number of children of an individual is less than $1 + M(s+h)$), so we will be able to bound its extinction probability quite easily.

Finally, denote $V^{(N)}$ the exploration process of $G^{(N)}$, $T^{(N)}$ the hitting time of -1 by $V^{(N)}$, $\bar{V}^{(N)}$ the exploration process of a $\mathbb{GW}_{\gamma_N, \gamma_N^O, \gamma_N^Y}$ -tree and $T(\gamma_N, \gamma_N^O, \gamma_N^Y)$ its size, i.e. the hitting time of -1 by $\bar{V}^{(N)}$. The result is the following.

Lemma 7.8. *Conditionally on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$, there is a coupling between $V^{(N)}$ and $\bar{V}^{(N)}$ such that, for all $k \leq T^{(N)} \wedge \alpha(N)$,*

$$V_k^{(N)} \leq \bar{V}_k^{(N)}.$$

In particular, $T^{(N)} \wedge \alpha(N) \leq T(\gamma_N, \gamma_N^O, \gamma_N^Y) \wedge \alpha(N)$, and

$$\frac{1}{N_t} \mathbb{E}(G_{\text{gel}}^{(N)} | I_S(t), \xi_{t,t-h}^{(N)}) \leq \mathbb{P}(T(\gamma_N, \gamma_N^O, \gamma_N^Y) > \alpha(N) | \gamma_N, \gamma_N^O, \gamma_N^Y).$$

Hence

$$\mathbb{E} \left(\frac{1}{N_t} G_{\text{gel}}^{(N)} \right) \leq \mathbb{E} \left(\mathbb{P}(T(\gamma_N, \gamma_N^O, \gamma_N^Y) > \alpha(N) | \gamma_N, \gamma_N^O, \gamma_N^Y) \right).$$

7.3.2 Summary

Our problem finally boils down to computing the probability for that Galton-Watson tree to grow big. That being said, now is probably the right time to look back at our results.

1. We have constructed a random graph $G^{(N)}$ which contains, in mean and w.h.p., more gel than our initial model.
2. Conditionally on N_t and $\xi_{t,t-h}^{(N)}$, this graph is a uniform pairing of the O arms and of the Y arms.
3. The quantity of gel in $G^{(N)}$ can be bounded by the probability that a two-type Galton-Watson process grows big.
4. The reproduction laws of this Galton-Watson process are deterministic functions of N_t and $\xi_{t,t-h}^{(N)}$.

Because of Lemma 7.8, our main interest is now to study the limit superior of

$$\mathbb{P}(T(\gamma_N, \gamma_N^O, \gamma_N^Y) > \alpha(N) | \gamma_N, \gamma_N^O, \gamma_N^Y).$$

Be careful to the definition of this object. We *first* pick laws γ_N, γ_N^O and γ_N^Y , and *then* we construct a two-type Galton-Watson tree with these reproductions laws, and we compute (conditionally on the reproduction laws) the probability that $T(\gamma_N, \gamma_N^O, \gamma_N^Y) > \alpha(N)$. To study this limit, we will first prove an easy lemma on Galton-Watson processes, then study the limit of these measures, and finally compute the extinction probability of the Galton-Watson process with these limit reproduction laws.

7.3.3 A lemma about extinction probabilities

This section is devoted to proving a lemma about extinction probabilities of some two-type Galton-Watson processes. We consider here non-random laws. So take laws λ , λ^O and λ^Y on $\mathbb{N} \times \mathbb{N}$. A couple of laws (λ^O, λ^Y) such that $\lambda^O(\mathbb{N} \times \{0\}) < 1$, $\lambda^O(\{0\} \times \mathbb{N}) < 1$, and $(\lambda^O, \lambda^Y) \neq (\delta_{(0,1)}, \delta_{(1,0)})$ will be called non-degenerate⁹.

Lemma 7.9. *Consider λ_N , λ_N^O , λ_N^Y laws on $\mathbb{N} \times \mathbb{N}$ and assume that they converge weakly to λ , λ^O and λ^Y for a non-degenerate (λ^O, λ^Y) . Then*

$$\mathbb{P}(T(\lambda_N, \lambda_N^O, \lambda_N^Y) > \alpha(N)) \rightarrow \mathbb{P}(T(\lambda, \lambda^O, \lambda^Y) = +\infty).$$

Proof. We will use the following easy result. For a r.v. X on $\mathbb{N} \cup \{+\infty\}$, denote $\phi_X(x) = \mathbb{E}(r^X \mathbb{1}_{\{X < +\infty\}})$ its generating function. Consider (X_N) and X r.v. on $\mathbb{N} \cup \{+\infty\}$ and denote $\phi_n = \phi_{X_N}$ and $\phi = \phi_X$. Then the following equivalences hold.

X_N converges to X in total variation

$$\Leftrightarrow \mathbb{P}(X_n = k) \rightarrow \mathbb{P}(X = k) \text{ for all } k \in \mathbb{N} \cup \{+\infty\}.$$

$$\Leftrightarrow \sum_{k \in \mathbb{N}} |\mathbb{P}(X_n = k) - \mathbb{P}(X = k)| \rightarrow 0$$

$$\Leftrightarrow \sum_{k \in \mathbb{N} \cup \{+\infty\}} |\mathbb{P}(X_n = k) - \mathbb{P}(X = k)| \rightarrow 0$$

$$\Leftrightarrow \phi_n \text{ converges to } \phi \text{ uniformly on } [0, 1]$$

$$\Leftrightarrow \phi_n \text{ converges pointwise to } \phi \text{ on } [0, 1].$$

Denote respectively g_N , g_N^O , g_N^Y , the generating functions of the sizes of the Galton-Watson trees $T(\lambda_N, \lambda^O, \lambda^Y)$, $T(\lambda_N^O, \lambda^O, \lambda^Y)$ and $T(\lambda_N^Y, \lambda^O, \lambda^Y)$, and ditto for g , g^O , g^Y . Then, it is well-known that for $r \in [0, 1]$

$$\begin{cases} g_N^O(r) &= r\phi_N^O(g_N^O(r), g_N^Y(r)) \\ g_N^Y(r) &= r\phi_N^Y(g_N^O(r), g_N^Y(r)) \end{cases}, \quad g_N(r) = \phi_N(g_N^O(r), g_N^Y(r)).$$

For $r \in [0, 1)$, the system has a unique solution. For $r = 1$ and in the non-degenerate cases, it has the only solution $(1, 1)$ in the (sub)critical case, and two solutions, $(1, 1)$ and another “smaller” one, in the supercritical case (see [24]).

Now, by uniform convergence of ϕ_N^O and ϕ_N^Y , for every $r \in [0, 1)$, every limit point of $(g_N^O(r), g_N^Y(r))$ (which is a bounded sequence) verifies

$$\begin{cases} g^O(r) &= r\phi^O(g^O(r), g^Y(r)) \\ g^Y(r) &= r\phi^Y(g^O(r), g^Y(r)). \end{cases}$$

This system has a unique solution, so $(g_N^O(r), g_N^Y(r))$ converges to that unique solution.

For $r = 1$, $(g_N^O(1), g_N^Y(1))$ is the smallest solution to

$$\begin{cases} g_N^O(1) &= \phi_N^O(g^O(1), g^Y(1)) \\ g_N^Y(1) &= \phi_N^Y(g^O(1), g^Y(1)). \end{cases} \quad (4.30)$$

Hence, every limit point of $(g_N^O(1), g_N^Y(1))$ solves

$$\begin{cases} g^O(1) &= \phi^O(g^O(1), g^Y(1)) \\ g^Y(1) &= \phi^Y(g^O(1), g^Y(1)). \end{cases}$$

⁹This is just the two-dimensional equivalent to the condition that the reproduction law is not δ_1 for a one-type Galton-Watson process, see [24].

In the (sub)critical case, this system has a unique solution $(1, 1)$, so $(g_N^O(1), g_N^Y(1)) \rightarrow (1, 1)$. In the supercritical case, it has two solutions (see [24]), one of them being $(1, 1)$. However, it is easy to see, by uniform convergence of (ϕ_N^O, ϕ_N^Y) to (ϕ^O, ϕ^Y) , that for large enough N , the smallest solution of (4.30) remains bounded away from $(1, 1)$, so $(g_N^O(1), g_N^Y(1))$ actually converges to the solution of the system above which is not $(1, 1)$.

Using the first result recalled, we have thus proven that (g_N^O, g_N^Y) to (g^O, g^Y) uniformly on $[0, 1]$. So $g_N(r) = \phi_N(g_N^O(r), g_N^Y(r))$ converges to $g(r) = \phi(g^O(r), g^Y(r))$ uniformly on $[0, 1]$. Hence, $T(\lambda_N, \lambda_N^O, \lambda_N^Y)$ converges to $T(\lambda, \lambda^O, \lambda^Y)$ in total variation, whence the result follows. \square

7.3.4 Extinction probability

The goal of this section is to compute a lower-bound for $\mathbb{P}(T(\gamma, \gamma^O, \gamma^Y) < +\infty)$, where γ , γ^O and γ^Y are defined in Appendix D. Having said that, we will only use the properties of these measures given in Section 7.3.1. Proposition 7.2 will be then easily proved after gathering the pieces.

Lemma 7.10. *There is a constant C depending only on μ and uniform for $t - h$ in the compacts of $(0, +\infty)$ such that*

$$\mathbb{P}(T(\gamma, \gamma^O, \gamma^Y) = +\infty | \gamma, \gamma^O, \gamma^Y) \leq C(s + h), \quad \text{a.s.}$$

Proof. Let us first proceed with some remarks on the reproduction laws. We denote ϕ , ϕ_O and ϕ_Y the generating functions of γ , γ_O and γ_Y . We may then reformulate (4.27), (4.28) and (4.29) by saying that there are constants $a, M > 0$, depending only on μ and uniform for $t - h$ and t in the compacts of $(0, +\infty)$, such that a.s.:

$$\frac{\partial \phi}{\partial x}(1, 1) = \frac{\partial \phi}{\partial y}(1, 1) \leq M, \quad \frac{\partial \phi_O}{\partial x}(1, 1) \leq 1, \quad \frac{\partial \phi_O}{\partial y}(1, 1) \leq M(s + h),$$

and

$$\frac{\partial \phi_Y}{\partial x}(1, 1) \leq M, \quad \frac{\partial \phi_Y}{\partial y}(1, 1) \leq M(s + h)$$

and finally

$$\frac{\partial^2 \phi_O}{\partial x^2}(0, 0) \geq a.$$

Now, let us condition on γ , γ^O and γ^Y . Let p_O (resp. p_Y) be the extinction probability of a Galton-Watson process $\mathbb{GW}_{\gamma^O, \gamma^O, \gamma^Y}$ started from a O (resp. Y) ancestor, and let $q = \mathbb{P}(T(\gamma, \gamma^O, \gamma^Y) = +\infty)$. Then, it is well-known (see e.g. [24]) that

$$\begin{cases} p_O &= \phi_O(p_O, p_Y) \\ p_Y &= \phi_Y(p_O, p_Y) \\ q &= \phi(p_O, p_Y). \end{cases}$$

Let us first compute an easy a priori bound on p_Y . Recall from Section 7.3.1 that, if (X, Y) has law γ_Y , then Y has, conditionally on X , a binomial law with parameters X

and $1 - e^{-(s+h)}$. Hence, we have

$$\begin{aligned}
 p_Y &= \phi_Y(p_O, p_Y) \\
 &\geq \sum_{j \geq 0} \gamma_Y(j, 0) p_O^j \\
 &= \sum_{j \geq 0} \gamma_Y^1(j) (e^{-(s+h)})^j p_O^j \\
 &\geq 1 + \sum_{j \geq 0} j \gamma_Y^1(j) (e^{-(s+h)})^j p_O - 1 = 1 + \langle \gamma_Y^1, k \rangle (e^{-(s+h)}) p_O - 1,
 \end{aligned}$$

the last inequality coming from the convexity of $x \mapsto \sum \gamma_Y^1(j) x^j$. Hence,

$$1 - p_Y \leq \langle \gamma_Y^1, k \rangle (s + h) + \langle \gamma_Y^1, k \rangle (1 - p_O) \leq M(s + h) + M(1 - p_O).$$

Now, Taylor's formula gives, for $x, y \in [0, 1]$,

$$\begin{aligned}
 \phi_O(x, y) &= \phi_O(1, 1) + (x - 1) \frac{\partial \phi_O}{\partial x}(1, 1) + (y - 1) \frac{\partial \phi_O}{\partial y}(1, 1) \\
 &\quad + \int_0^1 (1 - t) \left(\frac{\partial^2 \phi_O}{\partial x^2}(1 - t + tx, 1 - t + ty) (x - 1)^2 \right. \\
 &\quad + \frac{\partial^2 \phi_O}{\partial y^2}(1 - t + tx, 1 - t + ty) (y - 1)^2 \\
 &\quad \left. + \frac{\partial^2 \phi_O}{\partial x \partial y}(1 - t + tx, 1 - t + ty) (x - 1)(y - 1) \right) dt \\
 &\geq 1 - (1 - x) - (1 - y)M(s + h) + \frac{a}{2}(x - 1)^2
 \end{aligned}$$

and hence

$$p_O = \phi_O(p_O, p_Y) \geq p_O - (1 - p_Y)M(s + h) + \frac{a}{2}(p_O - 1)^2 \geq p_O - M(s + h)^2 - M(1 - p_O)(s + h) + \frac{a}{2}(p_O - 1)^2$$

so

$$-M^2(s + h)^2 - M(s + h)(1 - p_O) + \frac{a}{2}(p_O - 1)^2 \leq 0,$$

whence it is easy to deduce that

$$1 - p_O \leq \frac{M}{a} \left(1 + \sqrt{1 + 2a} \right) (s + h).$$

Finally, by convexity, we may compute the following bound, for some constants C' , C'' , C depending only on M and a , and thus depending only on μ and uniform when $t - h$ and t remain in the compacts of $(0, +\infty)$:

$$\begin{aligned}
 q &= \phi(p^O, p^Y) \\
 &\geq \phi(1, 1) - \frac{\partial \phi}{\partial x}(1, 1)(1 - p_O) - \frac{\partial \phi}{\partial y}(1, 1)(1 - p_Y) \\
 &\geq 1 - C's - C''s \\
 &\geq 1 - Cs.
 \end{aligned}$$

□

7.3.5 End of the proof

Finally, it just remains to prove Lemma 7.6, which is now straightforward. Indeed, Lemma 7.8 gives

$$\mathbb{E} \left(\frac{1}{N_t} G_{\text{gel}}^{(N)} \right) \leq \mathbb{E} \left(\mathbb{P}(T(\gamma_N, \gamma_N^O, \gamma_N^Y) > \alpha(N) | \gamma_N, \gamma_N^O, \gamma_N^Y) \right).$$

Then, Lemma 7.9 and dominated convergence yield

$$\limsup_{N \rightarrow +\infty} \mathbb{E} \left(\frac{1}{N_t} G_{\text{gel}}^{(N)} \right) \leq \mathbb{E} \left(\mathbb{P}(T(\gamma, \gamma^O, \gamma^Y) = +\infty | \gamma, \gamma^O, \gamma^Y) \right)$$

and the proof is completed by an appeal to Lemma 7.10.

7.4 Proof of Proposition 7.3

Let us now recall Proposition 7.3. We shall only sketch the proof, for the details are actually quite similar to the proof of Proposition 7.2 we just made, and to the proof of the asymptotic independence, Proposition 9.1, below.

Proposition 7.11. *For any $t, s \geq 0$,*

$$\limsup_{N \rightarrow +\infty} \mathbb{V} \left(n_t^{(N)} - n_{t+s}^{(N)} \right) = 0.$$

Proof. Fix $t, s \geq 0$ and let us condition $I_S(t)$. First note that

$$n_t^{(N)} - n_{t+s}^{(N)} = \frac{1}{N} \sum_{i \in I_S(t)} \mathbf{1}_{\{i \in I_G(t+s)\}}$$

so that $\mathbb{V} \left(n_t^{(N)} - n_{t+s}^{(N)} | I_S(t) \right)$ equals

$$\begin{aligned} & \frac{1}{N^2} \sum_{i \in I_S(t)} \left(\mathbb{P}(i \in I_G(t+s)) - \mathbb{P}(i \in I_G(t+s))^2 \right) \\ & + \frac{1}{N^2} \sum_{i, j \in I_S(t), i \neq j} \left(\mathbb{P}(i \in I_G(t+s) | j \in I_G(t+s)) - \mathbb{P}(i \in I_G(t+s)) \right) \times \mathbb{P}(j \in I_G(t+s)) \end{aligned}$$

The first term is lesser than $1/N$. Let us deal with the other. For $i \neq j \in I_S(t)$, we need to compare the probability that i falls into the gel on the time interval $[t, t+s]$ to the probability that this happens knowing that j falls into the gel on that same time-interval. But, on the one hand, there is probability tending to 0 that i and j find themselves in the same cluster (this can be seen as for the proof of Proposition 9.1 below).

When this is not the case, and that j falls into the gel, i binds to arms outside the cluster of j , and thus the event that it falls into the gel has the same probability as the event that it falls into the gel in a model where the cluster of j has been removed. This cluster has size $\alpha(N) \ll N$, so removing it obviously barely changes the empirical measure of the number of arms, and so the probability that i falls into the gel. Hence

$$\mathbb{P}(i \in I_G(t+s) | j \in I_G(t+s)) - \mathbb{P}(i \in I_G(t+s)) \rightarrow 0$$

whence the result follows easily. □

8 Self-organized criticality

We now turn to the proof of the actual criticality of ρ_t after gelation, i.e. of the following result.

Proposition 8.1. *For $t \geq T_{\text{gel}}$,*

$$\lim_{N \rightarrow +\infty} \langle \rho_t^{(N)}, k \rangle = 1.$$

Proof. Let us assume that, up to some subsequence $n^{(N)} \rightarrow n$ and $\rho^{(N)} \rightarrow \rho$ sharply in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$ and $\mathbb{D}(\mathbb{R}^+, \mathcal{M}_1)$. Define $m(t) = \langle \rho_t, k \rangle$, and recall from Proposition 7.1 that it is continuous. Assume now by contradiction that for some $t_0 > T_{\text{gel}}$, on an event of positive probability,

$$m(t_0) < 1 - 2\varepsilon.$$

We now work conditionally on that event.

Now obviously,

$$m(T_{\text{gel}}) = 1$$

so by continuity of m , there is a $T_{\text{gel}} < t < t_0$ such that

$$1 - 2\varepsilon > m(t) > m(t_0).$$

Hence,

$$\langle \rho_t^{(N)}, k \rangle < 1 - \varepsilon$$

w.h.p.

This implies that the concentration remains constant on a small time-interval after t . Note indeed that by Lemma 5.4, the state in solution is a configuration model conditioned on having no giant cluster. Hence, we may carry out the exact same proof as for Proposition 7.2, the only difference being that one has to replace

$$\langle \gamma_O^1, k \rangle \leq 1$$

by

$$\langle \gamma_O^1, k \rangle \leq 1 - \varepsilon.$$

The subsequent calculations then easily show that, for small enough s , the extinction probability of the (now strictly subcritical) Galton-Watson process therein is 0, and thus

$$\limsup \mathbb{E} \left(\frac{n_{t+s}^{(N)} - n_t^{(N)}}{n_t^{(N)}} \right) = 0$$

for small enough s . Hence, only a negligible proportion of the particles fall into the gel in the time interval $[t, t + s]$. This readily implies that m cannot decrease on $[t, t + s]$. Repeating the argument, we have thus shown that when m is strictly below 1, then it has to grow, thus yielding a contradiction. \square

9 Asymptotic independence

9.1 Introduction and assumptions

The aim of this section is to prove Proposition 2.20 and Corollary 2.21.

Recall from Lemma 5.4 that at time t , the configuration of the particles in solution has, conditionally on N_t and $\pi_t^{(N)}$, the same distribution as a configuration model $CM(N_t, \pi_t^{(N)})$ conditioned on having no giant cluster. That being said, we will assume in all the proofs that the state at time t is really a configuration model $CM(N_t, \pi_t^{(N)})$ *without conditioning*. This could be dealt with by “going back in time”, as is explained in Lemma 5.7: if we look at the particles of $I_S(t)$ at a time $t-h < t$ close to t , then their configuration is a subcritical configuration model conditioned on having no giant cluster, and the subcriticality implies that this conditioning is asymptotically trivial.

We already used in details that technique in the proof of Proposition 7.1. In the present case¹⁰, this would only lead to technical contortions and cumbersome notation. The interested reader could anyhow deal with that issue with ease, using in particular the continuity properties of Proposition 7.1, as well as (4.19).

Besides, in all the proofs, we assume the convergence of all the tight quantities of Proposition 7.1.

Finally, the constants in the proofs will only depend on μ and t . We will thus only write a constant C which may change from line to line, but only depends on μ and t .

9.2 Proof of Proposition 2.20

9.2.1 Goal

Let us first recall Proposition 2.20. Denote $\tilde{\mathbb{P}}_t^{(k)}$ the probability knowing that particle 1 is in solution at time t and has k activated arms. Obviously, 1 has no particular rôle, and could be replaced by any $i \in [N]$. Recall that $i \overset{t}{\leftrightarrow} j$ means that i and j are neighbors at time t , and let, for $i \overset{t}{\leftrightarrow} j$, $\mathcal{C}_t^{\setminus j}(i)$ be the cluster of i when deleting the link(s) between i and j . Finally, let $\beta(N)$ be a sequence such that $1 \ll \beta(N) \ll \alpha(N)$.

Proposition 9.1. *For any $t \geq T_{\text{gel}}$,*

$$\lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)} \left(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N), \#\{i : i \overset{t}{\leftrightarrow} 1, |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(i)| < \alpha(N) - \beta(N)\} \neq 1 \right) = 0.$$

In words, this means that, if 1 falls into the gel on the time-interval $[t, t + \varepsilon]$, then that is because one and only one of the subclusters to which it is attached has reached a large size $\alpha(N) - \beta(N)$.

9.2.2 Preliminary lemmas

The first part of the proof consists of several lemmas which are quite easy to prove upon knowing that we are dealing at time t with a critical configuration model. We denote here $\tilde{\mathbb{P}}_t$ the probability knowing that 1 is in solution at time t (but once again, this could just be any other particle).

Lemma 9.2. *For any $t \geq 0$, the following holds.*

¹⁰And, if one looks closely, as was already the case in the proof of Proposition 7.1.

1. One has

$$\lim_{N \rightarrow \infty} \tilde{\mathbb{P}}_t \left(\exists i \neq j : i \overset{t}{\leftrightarrow} 1, j \overset{t}{\leftrightarrow} 1, j \in \mathcal{C}_t^{\setminus 1}(i) \right) = 0. \quad (4.31)$$

This limit also holds for any $k \geq 0$ when replacing $\tilde{\mathbb{P}}_t$ with $\tilde{\mathbb{P}}_t^{(k)}$.

2. For any $\delta > 0$ there exists A large enough and depending only on δ, μ, t, k such that

$$\limsup_{N \rightarrow \infty} \tilde{\mathbb{P}}_t^{(k)} (|\mathcal{C}_t(1)| \geq A\varepsilon^{-2}) \leq \delta\varepsilon. \quad (4.32)$$

3. For any $k \geq 2$, there exists a constant C depending only on δ, μ, t, k such that for any ℓ_i, ℓ_j ,

$$\limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)} (|\mathcal{C}_t^{\setminus 1}(i)| = \ell_i, C_t^{\setminus 1}(j) = \ell_j | i \overset{t}{\leftrightarrow} 1, j \overset{t}{\leftrightarrow} 1) \leq C\ell_i^{-3/2}\ell_j^{-3/2}. \quad (4.33)$$

Proof. Recall that we may assume, up to minor modifications, that the state in solution at time t is, conditionally on N_t and $\pi_t^{(N)}$, a configuration model $CM(N_t, \pi_t^{(N)})$. The lemma is thus only a result on the configuration model. Note however that the parameters of the configuration model are random.

1. For the first result, note that, conditionally on $N_t, \pi_t^{(N)}$ and $\mathcal{C}_t(1) \leq \alpha(N)$ the event

$$\{\exists i \neq j : i \overset{t}{\leftrightarrow} 1, j \overset{t}{\leftrightarrow} 1, j \in \mathcal{C}_t^{\setminus 1}(i)\}$$

is included in the event that $\mathcal{C}_t(1)$ has at least one cycle. But \mathcal{C}_t is a critical configuration model, and the law of the number of arms is not degenerate, i.e. equal to $\delta_2/2$, because of (4.18). Hence, Bertoin and Sidoravicius' result [4] ensures that in that case a typical cluster is a tree, and thus has no cycle. In particular (and since $N_t \rightarrow +\infty$), $\mathcal{C}_t(1)$ has no cycle w.h.p., i.e.

$$\lim_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t \left(\exists i \neq j : i \overset{t}{\leftrightarrow} 1, j \overset{t}{\leftrightarrow} 1, j \in \mathcal{C}_t^{\setminus 1}(i) | N_t, \pi_t^{(N)} \right) = 0 \quad \text{a.s.}$$

The result follows after taking expectations and using dominated convergence.

2. The second part of the lemma would be well-known if the parameters of the configuration model were deterministic: this is essentially stating that the law of the size of a critical Galton-Watson has a tail which decreases in $t^{-1/2}$.

We need to be a little more careful here. We first need to define precisely the exploration process of a configuration model. This is explained in Appendix A., and we shall employ the same notation.

Condition first on N_t and $\pi_t^{(N)}$, and take $n = N_t, 1 \ll f(n) \ll N$. When working under $\tilde{\mathbb{P}}_t^{(k)}$, we may slightly modify our coupling by further imposing $V(1) = \bar{V}(1) = \underline{V}(1) = k$.

In the construction of the exploration process, we construct a family \bar{H} such that

$$\mathcal{H} \prec \bar{H}.$$

To this list correspond a measure $\bar{\rho}$, i.e., for U a uniform r.v. on $[0, 1]$, $\bar{\rho}$ is the law of $\phi_{\bar{H}}(U)$.

Recall that \bar{H} is constructed by removing a small¹¹ number $f(N_t)$ to the size-biased list of the number of arms of the N_t initial particles. Hence $\bar{\rho}$ is very close to $\rho_t^{(N)}$. It is not hard, though a bit long, to make this result more precise. The details are already given in the case of the two-type uniform pairing of Appendix D, so we shall not provide them here. We can actually show, thanks to the bounds of Lemma 5.1, that there are constants c and M such that, for any $\eta > 0$ and w.h.p.

$$\langle \bar{\rho}, k \rangle \leq 1 + \eta, \quad c \leq \langle \bar{\rho}, k^2 \rangle \leq M, \quad \langle \bar{\rho}, k^3 \rangle \geq c.$$

Now, under $\tilde{\mathbb{P}}^{(k)}$, 1 has k neighbors. Then, recall from Proposition A.3 that the size of its connected component is bounded by the hitting time T of -1 by a random walk \bar{V} started from $k - 1$, whose law of the steps is $\bar{\rho}$.

On the one hand, the probability that \bar{V} never reaches -1 can be bounded by a constant depending only on η and c . Since η can be chosen as small as we wish independently of c , we can take it such that

$$\mathbb{P}(T = +\infty) \leq \varepsilon.$$

Then, Berry-Esseen's theorem ensures that we can find C , which depends only on M and c , such that

$$\mathbb{P}(T \geq k, T < +\infty) \leq Ck^{-1/2}.$$

Finally, we obtain

$$\begin{aligned} \tilde{\mathbb{P}}^{(k)}(|\mathcal{C}_t(1)| \geq A\varepsilon^{-2}) &\leq \mathbb{P}(T \geq A\varepsilon^{-2}) \\ &\leq \varepsilon + CA^{-1/2}\varepsilon, \end{aligned}$$

so (4.32) is verified for large enough A .

3. The last part of the lemma is a similar reasoning. All one has to do is to modify slightly the coupling of Appendix A by imposing that the exploration process visits 1, then the component of i , then the component of j , provided these two last components are disjoint, which happens w.h.p. by the first part of the lemma.

□

9.2.3 An intermediary result

We will now prove the following result, which readily implies Proposition 9.1.

Lemma 9.3. *For any $t \geq T_{\text{gel}}$,*

$$\lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t \left(\exists i \neq j; i \overset{t}{\leftrightarrow} 1, j \overset{t}{\leftrightarrow} 1, |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(i)| \wedge |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(j)| \geq \sqrt{\beta(N)} \right) = 0.$$

Moreover, the same limit holds when replacing $\tilde{\mathbb{P}}_t$ by $\tilde{\mathbb{P}}_t^{(k)}$.

¹¹Because $n = N_t$ is of order N and $f(n) \ll N$

Proof. Take $i \overset{t}{\leftrightarrow} 1$ (if there is any), and consider its cluster disconnected from 1, $\mathcal{C}_t^{\setminus 1}(i)$. Say it has size k . It is then easy to see (for instance as a variation of the proof of Lemma 7.6) that there is a constant C such that any of the k particles of $\mathcal{C}_t^{\setminus 1}(i)$ has, on the time interval $[t, t + \varepsilon]$, probability less than $C\varepsilon^2$ to activate at least one arm *and* that the cluster to which it connects falls into the gel (informally, these two events are independent and each has probability of order ε to happen).

Then, using (4.33), we get

$$\limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t \left(|\mathcal{C}_t^{\setminus 1}(i)| = k, |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(i)| \geq \sqrt{\beta(N)} \mid i \overset{t}{\leftrightarrow} 1 \right) \leq Ck^{-1/2}\varepsilon^2. \quad (4.34)$$

On the other hand, suppose that 1 is in solution at t and both $i \neq j$ are its direct neighbors. Assume, what is true w.h.p. thanks to (4.31), that $\mathcal{C}_t^{\setminus 1}(i)$ and $\mathcal{C}_t^{\setminus 1}(j)$ are disjoint. Then, knowing that $\mathcal{C}_t^{\setminus 1}(j)$ reaches size $\sqrt{\beta(N)}$ on $[t, t + \varepsilon]$ is independent from the event that $\mathcal{C}_t^{\setminus 1}(i)$ activates one arm on $[t, t + \varepsilon]$. The latter happens with probability bounded by

$$C |\mathcal{C}_t^{\setminus 1}(i)|\varepsilon \wedge 1 \leq C(|\mathcal{C}_t^{\setminus 1}(i)|\varepsilon \wedge 1)$$

taking $C > 1$ if necessary. This, with Equation 4.34 provides

$$\begin{aligned} & \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t \left(|\mathcal{C}_t^{\setminus 1}(i)| = k, |\mathcal{C}_t^{\setminus 1}(j)| = k', |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(i)| \geq \sqrt{\beta(N)}, |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(j)| \geq \sqrt{\beta(N)} \right) \\ & \leq Ck^{-1/2}\varepsilon^2(k')^{-3/2}(k'\varepsilon \wedge 1). \end{aligned} \quad (4.35)$$

Note that we broke the symmetry in this equation.

It remains to sum the different contributions. We can decompose the event

$$\mathcal{E}_t := \left\{ |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(i)| \wedge |\mathcal{C}_{t+\varepsilon}^{\setminus 1}(j)| \geq \sqrt{\beta(N)} \right\}$$

according to the size of these two components at time t . First, according to (4.35)

$$\begin{aligned} & \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t \left(\mathcal{E}_t, |\mathcal{C}_t^{\setminus 1}(i)| \leq \delta\varepsilon^{-1}, |\mathcal{C}_t^{\setminus 1}(j)| \leq \delta\varepsilon^{-1} \right) \\ & \leq C\varepsilon^3 \sum_{k=1}^{\delta\varepsilon^{-1}} \sum_{k'=1}^{\delta\varepsilon^{-1}} k^{-1/2}(k')^{-1/2} \leq C\sqrt{\delta\varepsilon^{-1}}\sqrt{\delta\varepsilon^{-1}}\varepsilon^3 \leq C\delta\varepsilon. \end{aligned}$$

Still with (4.35),

$$\begin{aligned} & \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t \left(\mathcal{E}_t, 1 \leq |\mathcal{C}_t^{\setminus 1}(i)| \leq \delta\varepsilon^{-1}, \delta\varepsilon^{-1} < |\mathcal{C}_t^{\setminus 1}(j)| \leq A\varepsilon^{-2} \right) \\ & \leq C\varepsilon^2 \sum_{k=1}^{\delta\varepsilon^{-1}} k^{-1/2} \sum_{k'=\delta\varepsilon^{-1}+1}^{A\varepsilon^{-2}} (k')^{-3/2} \leq C\varepsilon^2\sqrt{\delta\varepsilon^{-1}} \sum_{k' \geq 1} (k')^{-3/2} \leq C\delta^{1/2}\varepsilon^{3/2}. \end{aligned}$$

The same inequality holds for the symmetric event, using the symmetric version of (4.35).

Finally,

$$\begin{aligned} & \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t \left(\mathcal{E}_t, \delta\varepsilon^{-1} < |\mathcal{C}_t^{\setminus 1}(i)| \leq A\varepsilon^{-2}, \delta\varepsilon^{-1} < |\mathcal{C}_t^{\setminus 1}(j)| \leq A\varepsilon^{-2} \right) \\ & \leq C\varepsilon^3 \sum_{k=\delta\varepsilon^{-1}+1}^{A\varepsilon^{-2}} k^{-1/2} \sum_{k'=\delta\varepsilon^{-1}+1}^{A\varepsilon^{-2}} (k')^{-1/2} \leq C\delta\varepsilon^2. \end{aligned}$$

We may then conclude with (4.33) that

$$\begin{aligned} \limsup_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t(\mathcal{E}_t) &\leq \tilde{\mathbb{P}}_t\left(|\mathcal{C}_t^{\setminus 1}(i)| \geq A\varepsilon^{-2}, |\mathcal{C}_t^{\setminus 1}(j)| \geq A\varepsilon^{-2}\right) + C(\delta\varepsilon + \delta^{1/2}\varepsilon^{3/2} + \delta\varepsilon) \\ &\leq \delta\varepsilon + C(\delta\varepsilon + \delta^{1/2}\varepsilon^{3/2} + \delta\varepsilon), \end{aligned}$$

so, since δ is arbitrary, the result follows.

The corresponding result for $\tilde{\mathbb{P}}_t^{(k)}$ is readily obtained by using the fact that the law of the number of arms of a particle in solution at t has finite variance, see (4.19). \square

9.2.4 Conclusion

From Lemma 9.3, Proposition 9.1 follows easily. Indeed, say that 1 has k neighbors at time t . Then, Lemma 9.3 states that, if $\mathcal{C}_t(1)$ reaches size $\alpha(N)$ on the time-interval $[t, t + \varepsilon]$, then at most one of the subclusters of 1 has reached size $\sqrt{\beta(N)}$. But if none does, then $\mathcal{C}_t(1)$ has size less than $k\sqrt{\beta(N)} + 1$, thus less than $\alpha(N)$ for large N .

Consequently, if 1 falls into the gel on $[t, t + \varepsilon]$, then exactly one of its subclusters reaches size greater than $\sqrt{\beta(N)}$. A fortiori, this cluster also reaches size $\alpha(N) - k\sqrt{\beta(N)}$ which is greater than $\alpha(N) - \beta(N)$ for large enough N , whence Proposition 9.1 follows.

9.3 Proof of Corollary 9.4

9.3.1 Rates of gelation

Recall that we define

$$f_k(t) := \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \lim_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N))$$

where the limit is along a subsequence where $\mathbf{p}^{(N)}$ converges, to some \mathbf{p} , say. Recall that $N\mathbf{p}^{(N)}(r, k)$ is the number of particles in solution at time t with k active arms and r arms in total.

The first question concerns the definition of $f_k(t)$: it is a priori not-clear that it should be well-defined. Rather than writing cumbersome formulas as we just did, we shall work a bit informally, the details being easy but long to write, and quite similar to what we just did. We hope that doing so will be more informative for the reader.

Let us rather start by considering

$$f_{r,k}(t) := \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \lim_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N) | a_1 = r)$$

where we recall that a_1 is the total number of arms of 1. This conditioning has positive probability as soon as $\mu(r) \neq 0$, and we will implicitly only consider such r .

We just saw in Proposition 9.1 that if 1 falls into the gel, this is due (up to terms of order $o(\varepsilon)$) to one (and only one) of its subclusters $\mathcal{C}^{\setminus 1}(i)$ reaching size $\alpha(N) - \beta(N)$, and this event has probability of order ε . The probability that, on top of that, 1 activates one arm has thus order ε^2 . Hence, if $f_{r,k}$ is well-defined, then so is f_k and $f_{r,k} = f_k$ for all $r \geq k$.

Let us check that $f_{r,k}$ is well-defined. So assume that 1 is in solution at time t , has k active arms at that time and r arms in total. On the time interval $[t, t + \varepsilon]$, it may:

- activate one arm: this happens with probability $(r - k)(1 - e^{-\varepsilon})$;
- fall into the gel: this happens with probability $\tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N) | a_1 = r)$;

- activate one arm and fall into the gel;
- activate more than two arms.

We just saw that the probability of the third event is of order ε^2 , and the probability of the fourth is obviously of order ε^2 .

As a consequence, up to terms of order ε^2 , $\mathbf{p}^{(N)}(r, k)$ changes on the interval $[t, t + \varepsilon]$ thanks to three phenomena.

- A particle in solution at time t with k active arms and r in total activates one arm: for each of them, this happens with probability $(r - k)(1 - e^{-\varepsilon})$.
- Such a particle falls into the gel: this happens with probability $\tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N)|a_1 = r)$.
- A particle in solution at time t with $k - 1$ active arms and r in total activates one arm: for each of them, this happens with probability $(r - k + 1)(1 - e^{-\varepsilon})$.

Hence, we may write

$$\begin{aligned} \mathbf{p}_{t+\varepsilon}(r, k) - \mathbf{p}_t(r, k) &= (r - k + 1)(1 - e^{-\varepsilon})\mathbf{p}_t(r, k - 1) - (r - k)(1 - e^{-\varepsilon})\mathbf{p}_t(r, k) \\ &\quad + \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N)|a_1 = r) + o(\varepsilon). \end{aligned} \quad (4.36)$$

According to Proposition 7.1, $\mathbf{p}(n, k)$ is Lipschitz-continuous, and thus is a.e. differentiable. This equation thus implies that

$$\tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| > \alpha(N)|a_1 = r)$$

is a.e. differentiable, and thus $f_{r,k}$, and then f_k , are well-defined a.e.

9.3.2 Relation to the number of active arms

The proof of the following corollary is now easy.

Corollary 9.4. *For all $t \geq 0$, $f_k(t) = k f_1(t)$.*

Proof. Because of Proposition 9.1, we can write

$$\tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_{t+\varepsilon}(1)| \geq \alpha(N)) = \sum_{i \xleftrightarrow{t} 1} \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_t^{\setminus 1}(i)| \geq \alpha(N) - \beta(N)) + o(\varepsilon). \quad (4.37)$$

Let us define, for $i \xleftrightarrow{t} 1$,

$$g(t) = \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \lim_{N \rightarrow +\infty} \tilde{\mathbb{P}}_t^{(k)}(|\mathcal{C}_t^{\setminus 1}(i)| \geq \alpha(N) - \beta(N)).$$

This quantity is readily seen to be well-defined, and it does not depend on i , since the state at time t is a configuration model, and thus all the subclusters attached at 1 have the same law. In particular (4.37) gives

$$f_k(t) = k g(t).$$

Hence $g_1(t) = f_1(t)$ and the result follows. \square

10 Evolution in time

We shall now end our proof. Since the tightness of the quantities we study, $(n^{(N)})$ and $(\pi^{(N)})$, has been proved, and the evolution in time is essentially given in the previous section, it just remains to prove the uniqueness of the limit point of these quantities, which is a far easier matter.

10.1 Evolution of $\mathbf{p}(r, k)$

The evolution of $\mathbf{p}(r, k)$ has been described in Section 9.3.1. From that, it is straightforward to deduce a PDE for its generating function.

Proposition 10.1. *Any limit point \mathbf{p} of $(\mathbf{p}^{(N)})$ enjoys the relation*

$$\frac{d\mathbf{p}_t}{dt}(r, k) = -\mathbf{p}_t(r, k)(kf(t) + (r - k)) + (r - k + 1)\mathbf{p}_t(r, k - 1).$$

In particular, the generating function

$$h_t(x, y) = \sum_{r, k \geq 0} \mathbf{p}_t(r, k) x^k y^r, \quad x, y \in [0, 1),$$

solves the PDE

$$\frac{\partial h_t}{\partial t} = x(1 - f(t) - x) \frac{\partial h_t}{\partial x} + (x - 1)y \frac{\partial h_t}{\partial y}, \quad (4.38)$$

with initial condition $h_0(x, y) = G_\mu(y)$.

Remark 10.2. In this statement, the derivatives have to be understood in the weak sense, or, in other words, one should write the equation in the integral form. However, the Lipschitz-continuity of $\mathbf{p}(r, k)$ ensures that it is the integral of its derivative, so there is no harm in doing so and we rather write derivatives to have clearer notations.

10.2 Solution to the PDE

The solution to this PDE is easy to obtain with the method of characteristics.

Proposition 10.3. *The PDE (4.38) has a unique solution given, for $x, y \in [0, 1)$, by*

$$h_t(x, y) = G_\mu \left(ye^{-t} (1 + x\theta(t)/\theta'(t)) \right),$$

where

$$\theta(t) = \int_0^t \exp - \left(\int_0^s (1 - f(r)) dr \right) ds.$$

Proof. It is easy to check that the given function is actually a solution to the PDE. To see that it is the only one, take h_t a solution to (4.38), and consider

$$\varphi_t^{(1)}(x, y) = \frac{x\theta'(t)}{1 - x\theta(t)}, \quad \varphi_t^{(2)}(x, y) = ye^t(1 - x\theta(t)).$$

Then, one may readily check that $h_t(\varphi_t^{(1)}(x, y), \varphi_t^{(2)}(x, y))$ is constant in t , and hence

$$h_t(\varphi_t^{(1)}(x, y), \varphi_t^{(2)}(x, y)) = G_\mu(y), \quad t \geq 0. \quad (4.39)$$

Now, note that, for each t , $(\varphi_t^{(1)}, \varphi_t^{(2)})$ is a bijection from $[0, 1)^2$ to $[0, 1)^2$ with inverse function

$$(x, y) \mapsto \left(\frac{x}{\theta'(t) + x\theta(t)}, ye^{-t} \left(1 + x \frac{\theta(t)}{\theta'(t)} \right) \right).$$

Plugging this values in (4.39) shows that the only possible solution is the one given in the statement. \square

10.3 Consequences

This formula allows to compute many quantities related to $\mathbf{p}_t(r, k)$, by extracting the coefficients of the representation in power series of h_t . Let us point out at the following, where we denote

$$\sigma(t) = e^{-t} \left(1 + \frac{\theta(t)}{\theta'(t)} \right).$$

- To begin with, we get

$$p_t(r, k) = \mu(r) e^{-rt} \binom{r}{k} \left(\frac{\theta(t)}{\theta'(t)} \right)^k.$$

- The mass at time t is

$$n_t = \sum_{r, k \geq 0} p_t(r, k) = h_t(1, 1) = G_\mu(\sigma(t)).$$

- The probability that a particle has k activated arms, knowing that it is in solution and has r arms in total is

$$\binom{r}{k} \left(\frac{\theta'(t)}{\theta(t) + \theta'(t)} \right)^{r-k} \left(\frac{\theta(t)}{\theta(t) + \theta'(t)} \right)^k.$$

In particular, each arm of a particle in solution is activated independently with probability $\theta(t)/(\theta(t) + \theta'(t))$.

- The sequence $(\pi_t(k))$ has generating function

$$G_{\pi_t}(x) = \frac{1}{n_t} G_\mu(e^{-t}(1 + x\theta(t)/\theta'(t))) = \frac{1}{n_t} G_\mu(\sigma(t)x + e^{-t}(1 - x)).$$

10.4 Recovering the rate of gelation

In the previous statements, everything depends on f , which is, at this point, unknown. However, we may recover it as follows. First, recall from Proposition 8.1 that, for $t > T_{\text{gel}}$, π_t is critical, i.e. $\langle \hat{\pi}_t, k \rangle = 1$. Knowing G_{π_t} , this implies that

$$(\sigma(t) - e^{-t})G'_\nu(\sigma(t)) = G_\nu(\sigma(t)). \quad (4.40)$$

One can readily check that this equation uniquely determines $\sigma(t)$, and so $f(t)$ as well. We may even obtain the following formula

$$f(t) = \frac{\sigma'(t)}{e^{-t} - \sigma(t)}.$$

Hence f , and thus every quantity, is uniquely determined, and so this ends our proof of Theorem 2.11.

10.5 Passing to the limit

Let us now see what happens when time tends to infinity. First, note that $\sigma(t)$ lies in the compact $[0, 1]$ and, according to (4.40), any of its limit point η solves

$$\eta G'_\nu(\eta) = G_\nu(\eta).$$

This equation has a unique solution, so $\sigma(t)$ converges to this η (which is the same as in Equation (4.4)). In particular, we obtain the results of Corollary 2.12. Let us remind that they allow to recover the results of [44], which was our initial goal.

A Exploration process of a configuration model

A.1 Definition

The exploration process $(V_k, k \geq 0)$ of the configuration model with a fixed degree sequence is defined for example in Chapter 10 of [54]. We shall remind its construction since we need the details in some proofs. We assume that we have n particles¹², particle i begin given d_i arms. Without loss of generality, we may assume that $d_1 \leq \dots \leq d_n$. Define $D_n = (d_1, \dots, d_n)$. Recall that the configuration model $CM(n, D_n)$ is obtained by picking a uniform pairing of the arms.

The vertices of the graph will be revealed through the exploration process. We will thus consider two sets of vertices, namely the set E of already explored vertices, and the set U of unexplored ones. We also define three sets of arms, namely the sleeping half edges S , the awoken ones A and the dead ones K .

Step 0 Initially, $U = \{1, \dots, n\}$, S is the set of all arms, and $V_0 = 0$.

Step 1 When there are no awoken arms (that is $A = \emptyset$, or equivalently (V_m) just reached a new infimum), choose a vertex, say i , uniformly at random in U . Put i in E , and awake all its half edges $\{(i, 1), \dots, (i, d_i)\}$ (that is, put them in A). If we have already recorded m steps of V , then we record the $(m + 1)$ -th by setting

$$V_{m+1} = V_m + d_i - 1.$$

Step 2 When $A \neq \emptyset$, pick one arm uniformly at random in A (say e_{j,n_j}), and pick another arm uniformly at random in $A \cup S$ (say e_{k,n_k}). Kill those two arms. At this juncture, there are two possibilities.

- a) Either the second arm that we just picked was asleep (i.e. $e_{k,n_k} \in A$). Then this means that we are connecting vertex j to vertex k which had not yet been explored. This obviously does not create a cycle. We then put k in E , and awake all its remaining arms. If we have already recorded m steps of V , then we record the $(m + 1)$ -th by setting

$$V_{m+1} = V_m + d_k - 2,$$

and then go on with the exploration.

- b) Or the second arm that we picked was already activated. Then this means that we are connecting vertices j and k , which were already explored at previous steps of the exploration. This also means we just found a cycle in the explored component. We do not need in that case to awake any new arm. But more importantly, we update our path and lower its value by 2. That is, if the last recorded value of V was $V_m = k$ we leave the first $m - 1$ steps of V unchanged, but correct V_m to $k - 2$.

Remark A.1. The important feature of this algorithm is that the size of a connected component corresponds to the number of recorded steps of V between reaching times of new infima. More precisely, the size of the first explored component is the hitting time of -1 by V , the size of the second is the hitting time of -2 , and so on.

¹² n will not necessarily be N !

A.2 Remarks

At any step of the exploration, let $\Delta(U) = (d_i)_{i \in U}$ denote the $\#U$ -uplet of the degrees of the unexplored vertices, so in particular $\Delta(U)$ is ordered.

- Any time we start the exploration of a new component (i.e. perform Step 2.a)), the corresponding increment for V , shifted up by one, is that of a uniformly chosen element in $\Delta(U)$.
- Now, recall that $S = \{(i, j) : i \in U\}$ is the set of sleeping arms. If $U = \{i_1, \dots, i_{\#U}\}$, we define

$$\mathcal{H}(S) = \left(\overbrace{d_{i_1}, \dots, d_{i_1}}^{d_{i_1} \text{ times}}, \dots, \overbrace{d_{i_{\#U}}, \dots, d_{i_{\#U}}}^{d_{i_{\#U}} \text{ times}} \right).$$

Note that this uplet is ordered.

Any step of the exploration process which does not start the exploration of a new component corresponds to choosing uniformly at random a pair of distinct arms in $A \times A \cup S$.

- If we choose the second arm in A we will record no new step for V , and update its current final value by -2 , an event of probability

$$\frac{\#A - 1}{\#A - 1 + \#S}$$

- If we choose the second arm in S , an event of probability

$$\frac{\#S}{\#A - 1 + \#S}$$

we record a new step for V , and this step, shifted up by one, is that of a uniformly chosen element of $\mathcal{H}(S)$.

The exact distribution of the steps of V is quite complicated, because the distribution of the degrees of U evolves as we go along with the exploration. Note, however, that when n is large, this distribution should not vary too much during the first $f(n)$ steps of V , when $f(n)/n$ is small. We exhibit in the next paragraph a natural coupling of V with a path \bar{V} , respectively \underline{V} such that $(V_i, i \leq f(n))$ will be bounded above by $(\bar{V}_i, i \leq f(n))$, respectively below by $(\underline{V}_i, i \leq f(n))$.

A.3 Some notations

Let $p \in \mathbb{N}^*$, $A_p := (d_1 \leq \dots \leq d_p)$ a p -uplet of integers, and the natural stick-breaking of $[0, 1]$ in p parts,

$$I_1 = [0, 1/p), I_2 = [1/p, 2/p) \dots, I_p = [(p-1)/p, 1].$$

For $x \in [0, 1]$, let

$$\phi_{A_p}(x) = \sum_{i=1}^p d_i \mathbb{1}_{\{I_i\}}(x).$$

Definition A.2. For $p, q \in \mathbb{N}^*$, and A_p an ordered p -uplet, B_q and ordered q -uplet of integers, we say $A_p \prec B_q$ if

$$\forall x \in [0, 1] \quad \phi_{A_p}(x) \leq \phi_{B_q}(x).$$

Recall now that $D_n : d_1 \leq \dots \leq d_n$ is the initial sequence of degrees, and fix $f(n) < n$. From D_n , we may define other lists

$$\overline{D}_n := d_{f(n)+1} \leq \dots \leq d_n, \quad \underline{D}_n := d_1 \leq \dots \leq d_{n-f(n)}.$$

In that case, when less than $f(n)$ steps of the exploration process have been performed, we have

$$\underline{D}_n \prec \Delta(U) \prec \overline{D}_n.$$

In the same vein, we may define

$$\overline{H}_n := \left(\overbrace{d_{f(n)+1}, \dots, d_{f(n)+1}}^{d_{f(n)+1} \text{ times}}, \dots, \overbrace{d_n, \dots, d_n}^{d_n \text{ times}} \right)$$

and

$$\underline{H}_n := \left(\overbrace{d_1, \dots, d_1}^{d_1 \text{ times}}, \dots, \overbrace{d_{n-f(n)}, \dots, d_{n-f(n)}}^{d_{n-f(n)} \text{ times}} \right)$$

so that

$$\underline{H}_n \prec \mathcal{H} \prec \overline{H}_n$$

when less than $f(n)$ steps of the exploration process have been taken.

A.4 Reformulation with uniform variables

We may reformulate our algorithm in terms of continuous independent uniform random variables. More precisely, let $(U_{i,j}, U_{i,j,\ell}, i \geq 1, j \geq 1, \ell \geq 1)$ be independent uniform random variables on $[0, 1]$, and proceed as follows.

- a) $V_0 = 0$.
- b) If we have recorded k steps of V , and V_k is at its j th new infimum, then we choose the next explored vertex in such a way that

$$V_{k+1} - V_k = \phi_{\Delta(U)}(U_{1,j}) - 1.$$

- c) If

- we have recorded k steps of V ,
- V is not currently at a new infimum,
- we last recorded the i th step in the j th excursion of V above new infima, and for some $\ell \geq 0$ we just recorded the ℓ th correction of V_k ,

then we choose the pair of arms in $A \times A \cup S$ in such a way that

$$\begin{cases} V_k \text{ is corrected by } -2 & \text{if } U_{i,j,\ell+1} \leq (\#A - 1)/(\#A - 1 + \#S), \\ V_{k+1} - V_k = \phi_{\mathcal{H}(S)}(U_{i+1,j}) & \text{if } U_{i,j,\ell+1} > \#S/(\#A - 1 + \#S). \end{cases}$$

It remains to observe that when less than $f(n)$ vertices have been explored, we must have

$$\bar{g}(n) := 0 \leq (\#A - 1)/(\#A - 1 + \#S) \leq \underline{g}(n) := \frac{\sum_{i=n-f(n)+1}^n d_i}{\sum_{i=1}^{n-f(n)} d_i}.$$

A.5 Lower- and upper-bound

We are now able to define \underline{V} and \bar{V} .

- a. Let $\underline{V}_0 = 0$.
- b. If we have recorded k steps of \underline{V} (resp. \bar{V}), and V_k (resp. \bar{V}_k) is at its j th new infimum, then we let

$$\underline{V}_{k+1} - \underline{V}_k = \phi_{\underline{D}}(U_{1,j}) - 1 \quad \left(\text{resp. } \bar{V}_{k+1} - \bar{V}_k = \phi_{\bar{D}}(U_{1,j}) - 1 \right).$$

c. If

- we have recorded k steps of \underline{V} (resp. \bar{V}),
- it is not currently at a new infimum,
- we last recorded the i th step in the j th excursion of \underline{V} (resp. \bar{V}) above new infima, and then for some $\ell \geq 0$ we just recorded the ℓ -th correction of \underline{V}_k (resp. \bar{V}_k),

then

$$\begin{cases} \underline{V}_k \text{ is corrected to } (\underline{V}_k - 2) \vee (\inf_{m \leq k} \underline{V}_m - 1) & \text{with probability } \underline{g}(n) \\ \underline{V}_{k+1} - \underline{V}_k = \phi_{\underline{H}}(U_{i+1,j}) - 1 & \text{with probability } 1 - \underline{g}(n) \end{cases}$$

and respectively,

$$\bar{V}_{k+1} - \bar{V}_k = \phi_{\bar{H}}(U_{i+1,j}) - 1.$$

Define now $T_j = \inf\{k : V_k = -j\}$, $\bar{T}_j = \inf\{k : \bar{V}_k = -j\}$ and $\underline{T}_j = \inf\{k : \underline{V}_k = -j\}$. We then have the following result, which readily follows from our construction.

Proposition A.3. *As long as less than $f(n)$ steps of the exploration process have been taken, we have:*

$$\underline{V}_{\bar{T}_j+k} \leq V_{T_j+k} \quad \forall k \in \{0, \dots, (\underline{T}_{j+1} - \underline{T}_j) \wedge (f(n) - T_j)\}, \quad (4.41)$$

$$V_{T_j+k} \leq \bar{V}_{\bar{T}_j+k} \quad \forall k \in \{0, \dots, (T_{j+1} - T_j) \wedge (f(n) - T_j)\}. \quad (4.42)$$

This implies in particular that for j fixed, if the j th connected component of the graph is fully explored once $f(n)$ vertices have been explored, its size is bounded above by $\bar{T}_{j+1} - \bar{T}_j$ (resp. it is bounded below by $\underline{T}_{j+1} - \underline{T}_j$).

Also, the size of the component of a uniformly chosen vertex is bounded above by \bar{T}_1 , and below by $f(n) \wedge \underline{T}_1$.

B Tightness

B.1 Reminder of the results

In this appendix, we prove Proposition 7.1. We start with the tightness of $(n^{(N)})$, from which the tightness of the other quantities follows easily. Our two main tools are Propositions 7.2 and 7.3 proved in Section 7, which we recall now.

Proposition B.1. *For every compact subset K of $(0, +\infty)$, there is a constant C , depending only on this compact and μ , such that, for all $t \in K$ and $s \leq 1$,*

$$\limsup_{N \rightarrow +\infty} \mathbb{E} \left(\frac{n_t^{(N)} - n_{t+s}^{(N)}}{n_t^{(N)}} \right) \leq Cs.$$

Proposition B.2. *For any $t, s \geq 0$,*

$$\limsup_{N \rightarrow +\infty} \mathbb{V} \left(n_t^{(N)} - n_{t+s}^{(N)} \right) = 0.$$

These results are enough to prove the expected result, as we shall now see.

B.2 Proof of the tightness of $(n^{(N)})$

Fix $\eta > 0$ and let $0 < \delta < T_{\text{gel}} \wedge 1$, $0 < T^- < T_{\text{gel}}$ and $T^+ > T_{\text{gel}} + 1$. Consider the partition $t_0 = 0 < t_1 = T^- < t_2 < \dots < t_n = T^+$ of $[0, T^+]$, with $t_{i+1} = t_i + \delta/2$ for $1 \leq i < n-1$ and $\delta/2 \leq t_n - t_{n-1} < \delta$. Define also

$$w_N(u, v) = \sup_{t, s \in [u, v]} \left| n_t^{(N)} - n_s^{(N)} \right|.$$

Now, Propositions B.1 and B.2 ensure that we can find N_0 such that, for $N \geq N_0$ and $1 \leq i < n$,

$$\mathbb{E} \left(n_{t_i}^{(N)} - n_{t_i+\delta}^{(N)} \right) \leq \mathbb{E} \left(\frac{n_{t_i}^{(N)} - n_{t_i+\delta}^{(N)}}{n_{t_i}^{(N)}} \right) \leq (K+1)\delta$$

and

$$\mathbb{V} \left(n_{t_i}^{(N)} - n_{t_i+\delta}^{(N)} \right) \leq \delta^2$$

and thus Chebyshev's inequality yields, for $C = (K+2)/\eta^2$,

$$\mathbb{P} \left(n_{t_i}^{(N)} - n_{t_i+\delta}^{(N)} > \eta \right) \leq C\delta^2.$$

Now, note that, when $s, t \in [t_i, t_{i+1}]$ for some i and $s < t$, since $n^{(N)}$ is decreasing, the inequality

$$n_s^{(N)} - n_t^{(N)} \leq n_{t_i}^{(N)} - n_{t_{i+1}}^{(N)}$$

holds. Note also that, by Proposition 4.1, since $T^- < T_{\text{gel}}$,

$$\limsup_{N \rightarrow +\infty} \mathbb{P}(n_0^{(N)} - n_{T^-}^{(N)} > \eta) = 0,$$

and hence, for large enough N ,

$$\mathbb{P}(n_0^{(N)} - n_{T^-}^{(N)} > \eta) \leq \delta$$

We can finally deduce that, for large N ,

$$\mathbb{P}\left(\max_{i=0\dots n-1} w_N(t_i, t_{i+1}) > \eta\right) \leq C\delta^2 \times n + \delta \leq (C+1)\delta$$

and thus

$$\limsup_{N \rightarrow +\infty} \mathbb{P}\left(\max_{i=0\dots n-1} w_N(t_i, t_{i+1}) > \eta\right) \leq (C+1)\delta.$$

Having $\delta \rightarrow 0$ finally ensures that, along with the tightness at fixed t of Lemma 5.3, the usual tightness criterion in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$ (see [6]) is verified, whence the tightness follows. Finally, Propositions B.1 and B.2 and dominated convergence readily ensure the regularity of the trajectories.

B.3 Proof of the tightness of the other processes

Let us write \mathcal{M}_1^w for the space \mathcal{M}_1 endowed with the tight topology (of convergence in law), and \mathcal{M}_1^v when it is endowed with the vague topology. Let us recall a useful result of tightness in $D(\mathbb{R}^+, \mathcal{M}_1^w)$ (see [50]): a sequence $(\lambda^{(N)})$ of \mathcal{M}_1 -valued processes is tight in $D(\mathbb{R}^+, \mathcal{M}_1^w)$ if and only if

- (i) it is tight in $D(\mathbb{R}^+, \mathcal{M}_1^v)$, i.e. $(\lambda^{(N)}(k))$ is tight in $D(\mathbb{R}^+, \mathbb{R})$ for every $k \in \mathbb{N}$,
- (ii) for any vague limit point λ of $\lambda^{(N)}$ in $D(\mathbb{R}^+, \mathcal{M}_1^v)$, λ has mass 1.

Let us first introduce an easy result. Denote

$$Na_t^{(N)} = \sum_{i=1}^N A_t(i)$$

the total number of activated arms at time t . Recall that $K_N = N\langle \mu^{(N)}, k \rangle$ is the total number of arms at time 0, and that $K_N/N \rightarrow \langle \mu, k \rangle$ by Assumption 2.1. Then $K_N - Na_t^{(N)}$ is a pure-death process with rate 1, so it is easy to check that $(K_N - a_t^{(N)})/N$ converges in $\mathbb{D}(\mathbb{R}^+, \mathbb{R}^+)$ to $\langle \mu, k \rangle e^{-t}$, so

$$\frac{a_t^{(N)}}{N} \xrightarrow{N \rightarrow \infty} \langle \mu, k \rangle (1 - e^{-t})$$

in $\mathbb{D}([0, +\infty), \mathbb{R}^+)$.

We will now check item (i). Take $\alpha > 0$ and recall that convergence to a continuous limit for Skorokhod's topology is equivalent to uniform convergence on every compact. Fix a compact $K \subset [0, +\infty)$ and assume, with the above result and the tightness of $(n^{(N)})$, that (up to some subsequence which we do not write) $(n^{(N)})$ and $(a^{(N)})$ converge. Then, uniform convergence ensures that there is a $\varepsilon > 0$ such that, w.h.p.,

$$\sup_{t \in K} \sup_{s \leq \varepsilon} |a_t^{(N)} - a_{t+s}^{(N)}| < \alpha, \quad \sup_{t \in K} \sup_{s \leq \varepsilon} |n_t^{(N)} - n_{t+s}^{(N)}| < \alpha$$

so it is easy to see, along with (4.18), that there is a constant C such that w.h.p.,

$$\sup_{t \in K} \sup_{s \leq \varepsilon} |\pi_t^{(N)}(k) - \pi_{t+s}^{(N)}(k)| < C\alpha. \quad (4.43)$$

From this inequality, tightness in $D(\mathbb{R}^+, \mathcal{M}_1^v)$ follows readily.

Item (ii) is readily deduced from (4.19), which finally shows the tightness of $(\pi^{(N)})$ in $D(\mathbb{R}^+, \mathcal{M}_1^w)$.

Finally, Hölder's inequality and (4.19) ensure that similar inequalities as (4.43) can be obtained for the other quantities, whence their tightness follows. The continuity of the limit points also stems directly from these bounds.

C Mass in Smoluchowski's and Flory's models

The goal of this appendix is to prove Lemma 7.5, which we remind now.

Lemma C.1. *For every $i \in I_S(t)$, one has*

$$\mathbb{P}\left(|\mathcal{C}^{(N)}(i)| > \alpha(N) \mid I_S(t), \xi_{t,t-h}^{(N)}\right) \leq \mathbb{P}\left(|\tilde{G}^{(N)}(i)| > \alpha(N) \mid I_S(t), \xi_{t,t-h}^{(N)}\right).$$

In particular

$$\mathbb{E}\left(\mathcal{C}_{\text{gel}}^{(N)} \mid I_S(t), \xi_{t,t-h}^{(N)}\right) \leq \mathbb{E}\left(\tilde{G}_{\text{gel}}^{(N)} \mid I_S(t), \xi_{t,t-h}^{(N)}\right)$$

and

$$\mathbb{E}\left(\frac{1}{N_t} \mathcal{C}_{\text{gel}}^{(N)}\right) \leq \mathbb{E}\left(\frac{1}{N_t} \tilde{G}_{\text{gel}}^{(N)}\right).$$

Recall it concerns two graphs $\mathcal{C}^{(N)}$ and $\tilde{G}^{(N)}$ that are constructed as follows.

- Condition on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$.
- Pick a uniform pairing of the O arms, conditioned on having no giant cluster.
- To get $\mathcal{C}^{(N)}$, activate each Y arm independently after an exponential time with parameter 1. Bind the first two activated arms *in solution*, then the two next, and so on (do not forget that clusters fall into the gel in the meantime), until time $s + h$.
- To get $\tilde{G}^{(N)}$, activate each Y arm independently after an exponential time with parameter 1. Bind the first two activated arms *in solution or not*, then the two next, and so on, until time $s + h$.

Then Lemma C.1 informally states that a typical component of $\mathcal{C}^{(N)}$ is larger than a typical component of $\tilde{G}^{(N)}$, from which it is easy to deduce that there is more gel in $\mathcal{C}^{(N)}$ than in $\tilde{G}^{(N)}$. Indeed, we may write

$$\mathcal{C}_{\text{gel}}^{(N)} = \sum_{i \in I_S(t)} \mathbb{1}_{\{|\mathcal{C}^{(N)}(i)| > \alpha(N)\}}$$

and the second part of the lemma is obtained by taking conditional expectations, whereas the last is deduced by taking expectations and noticing that N_t is $I_S(t)$ -measurable.

To keep things as simple as possible, notice that the only difference between the two graphs $\mathcal{C}^{(N)}$ and $\tilde{G}^{(N)}$ lies in the algorithm used to bind the Y arms. So we may condition on $I_S(t)$, $\xi_{t,t-h}^{(N)}$, the initial pairing of O arms and the activated Y arms on the time-interval $[0, s + h]$. This being done, all the randomness is given by a uniform ordering of the Y arms. We can thus consider the following problem.

- Start from a certain configuration of (not giant) clusters, each with a certain number of available arms. Say there are K particles $1, \dots, K$ in all.
- Choose a uniform ordering of these available arms.
- Bind the first two activated arms *in solution*, then the two next, and so on (again, clusters of size $\alpha(N)$ fall into the gel in the meantime), to get a configuration \mathcal{D} .
- Bind the first two activated arms *in solution or not*, then the two next, and so on, to get a configuration $\tilde{\mathcal{D}}$.

In particular, we can construct \mathcal{D} and $\tilde{\mathcal{D}}$ by discrete-time algorithms. Now, all we have to prove is the following.

Lemma C.2. *For every $p \in [K]$,*

$$\mathbb{P}(|\mathcal{D}(p)| > \alpha(N)) \leq \mathbb{P}(|\tilde{\mathcal{D}}(p)| > \alpha(N)).$$

Proof. To make it easier to study, it is interesting to change the algorithm a little.

- For \mathcal{D} , pick at each step an arm uniformly at random, and pair it to another one chosen uniformly at random in the same substrate (i.e. in solution if the first arm is in solution, or in the gel if the first arm is in the gel).
- For $\tilde{\mathcal{D}}$, pick at each step an arm uniformly at random, and bind it to another one chosen uniformly at random. Note that it is just a uniform pairing of the arms, or, in other words, a configuration model.

It is obvious that the final states in solution (but we are not interested in the configuration of the gel) of these two constructions have the same law as \mathcal{D} and $\tilde{\mathcal{D}}$. We will call the first algorithm Smoluchowski's algorithm, the second Flory's.

Now, fix a particle $p \in [K]$. To get the result, we shall couple these two processes so as to provide two cluster configurations \mathcal{D}_0 and $\tilde{\mathcal{D}}_0$ enjoying the following properties.

1. The configuration \mathcal{D}_0 and \mathcal{D} have the same law.
2. The cluster $\tilde{\mathcal{D}}_0(p)$ has the law as $\tilde{\mathcal{D}}(p)$.
3. If $|\mathcal{D}_0(p)| > \alpha(N)$, then $|\tilde{\mathcal{D}}_0(p)| > \alpha(N)$.

This will prove that $\mathbb{P}(|\mathcal{D}(p)| > \alpha(N)) \leq \mathbb{P}(|\tilde{\mathcal{D}}(p)| > \alpha(N))$, thus proving the result.

Let us now define this coupling. It will be clear from the construction that this can be done on a large enough probability space. Recall that we say that an arm is free if it is not linked. Let σ be a uniform ordering of the arms. Pick, say, $\sigma = \sigma_0$, which is fixed in all the steps of the two following algorithms. In the sequel, the phrase "the smallest arm" will thus refer to the smallest for that ordering.

1. At step 0, all the arms are active.
2. At each step, consider the smallest active arm. If it is in solution, bind it to another arm chosen uniformly in solution and deactivate the both. If it is in the gel, choose another arm uniformly in the gel, and deactivate the both.

We thus get a cluster configuration \mathcal{D}_0 , defined on $\{\sigma = \sigma_0\}$.

Now, let us condition on the steps of this algorithm, and let us construct another cluster configuration $\tilde{\mathcal{D}}_0$. At each step k of the first algorithm, we let S_k the set of active arms in solution, G_k the set of active arms in the gel, s_k and g_k their cardinality. We will construct the cluster of p as follows.

1. As long as $|\tilde{\mathcal{D}}_0(p)| \leq \alpha(N)$, consider the smallest free arm b of $\tilde{\mathcal{D}}_0(p)$. Since $\tilde{\mathcal{D}}_0(p)$ has size less than $\alpha(N)$, then b is deactivated at a step k in the above algorithm, and it is in solution at that time¹³. Hence, b is linked to an arm $b' \in S_k$. Then

¹³If b is deactivated at this step and is in the gel, then this gel has been created by earlier steps of the first algorithm. Earlier steps of the current algorithm should hence also have created some gel.

- (i) with probability $(s_k - 1)/(s_k + g_k - 1)$, link b and b' ;
 - (ii) with probability $g_k/(s_k + g_k - 1)$, link b to an arm chosen uniformly in G_k .
2. Proceed until no more arms are available (i.e. the whole cluster is formed) or $\tilde{\mathcal{D}}_0(p) > \alpha(N)$. When, if ever, $\tilde{\mathcal{D}}_0(p)$ reaches a size greater than $\alpha(N)$, proceed as for the configuration model: chose at each step an arm of the cluster (say the smallest) and pair it to another arm chosen uniformly among all free arms, until the whole cluster is constructed.

Note first that this algorithm is well-defined. There is indeed no problem if we always create the same configuration as in the first algorithm, i.e. we only perform step (i). Now, say that we do perform step (ii), linking an arm b of $\tilde{\mathcal{D}}_0(p)$, deactivated at step k of the first algorithm, and an arm $b' \in G_k$. An issue would arise if, when continuing the algorithm, we had to link an arm to b' . But, as long as $\tilde{\mathcal{D}}_0(p)$ has not reached size $\alpha(N)$, the steps of the first algorithm that we consider are steps when b' is not in the gel. Hence, we can only bind either to a particle bound at a step before k of the first algorithm, and which is thus not b' ; or to a particle in the gel at a step before k , which is not b' as we just said. However, this stops holding when $|\tilde{\mathcal{D}}_0(p)| > \alpha(N)$, and that is why the algorithm is modified should this occur.

It is easy to see that if $|\mathcal{D}_0(p)| \geq \alpha(N)$, then $|\tilde{\mathcal{D}}_0(p)| \geq \alpha(N)$. Note indeed that, when constructing $\tilde{\mathcal{D}}_0(p)$, then either we create the exact same cluster as $\mathcal{D}_0(p)$ and so this is obvious. Or, at some step, we pick an arm in the gel. Then, continuing the algorithm with the particle to which this arm is attached, we see by recurrence that it can only grow to a size greater than $\alpha(N)$.

Let us now prove that $\tilde{\mathcal{D}}_0(p)$ has indeed the law of the cluster of p in a configuration model. Note that, in our second algorithm, an arm which is deactivated at step k of the first algorithm is bound to another arm chosen uniformly at random among all available arms at that step. So take b the smallest arm of p . If b is the smallest arm of all, then it is deactivated at step 1 of the first algorithm, and is thus bound to another arm chosen uniformly among all. Now, say it is the second smallest arm of all, and the smallest is b_1 . Then

- with probability $1/(2K - 1)$, b_1 chooses b at step 1 of the first algorithm, so b is bound to b_1 with that probability. If b_1 chooses another than b , then b will not bind to b_1 . So b and b_1 are bound with probability $1/(2K - 1)$.
- For $b' \neq b, b_1$, with probability $(2K - 3)/(2K - 1)$, b_1 chooses an arm different from b and b' . Then, at step 2 of the first algorithm, b choses b' (which is available) with probability $1/(2K - 3)$. So the probability that b is linked to b' is

$$\frac{2K - 3}{2K - 1} \frac{1}{2K - 3} = \frac{1}{2K - 1}.$$

It is easy to continue according to the order of b . Then, upon conditioning on this first link, it is not difficult but a tad tedious to check that each other arm chooses another arm uniformly at random among all available arms. This reasoning holds until $\tilde{\mathcal{D}}_0(p)$ reaches size $\alpha(N)$, after what each arm still chooses another one uniformly in solution by construction.

We have thus shown that, on $\{\sigma = \sigma_0\}$, if $|\mathcal{D}_0(p)| > \alpha(N)$ then $|\tilde{\mathcal{D}}_0(p)| > \alpha(N)$, and that $\tilde{\mathcal{D}}_0(p)$ has the same law as $\tilde{\mathcal{D}}(p)$. Obviously, this is still true after integrating over σ .

Finally, upon integrating over σ , it is easy that the cluster $\mathcal{D}_0(p)$ we get is a typical cluster for Smoluchowski's evolution, i.e. $\mathcal{D}_0(p) \stackrel{(d)}{=} \mathcal{D}(p)$. Indeed, after integration, at each step, an arm is chosen uniformly and bound to another arm chosen uniformly in the same substrate. This is precisely Smoluchowski's algorithm.

We have thus verified the three conditions we requires above, so this ends our proof. \square

D Exploration process of a two-type uniform pairing

D.1 Goal

Our aim in this appendix is to prove Lemma 7.8 and show the properties of the measures γ_N , γ_N^O and γ_N^Y stated in Section 7.3.1. To this end, similarly to the construction of an upper-bound for the exploration process of a configuration model described in Appendix A, we provide here a construction of a process bounding the exploration process of a two-type uniform pairing. It is actually the exploration process of a two-type Galton-Watson tree.

We shall first condition on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$ in the notations of Lemma 5.7. Then, we will build a graph and its exploration process, as well as an upper-bound for it. The latter depends only on measures γ_N , γ_N^O and γ_N^Y which are deterministic functions of $\#I_S(t)$ and $\xi_{t,t-h}^{(N)}$.

Hence, in the first part of our construction, we fix $Z \in \mathbb{N}^*$ particles, whose empirical distribution of O and Y arms is denoted ξ , i.e. there are $\xi(i, j)Z$ particles with i O arms and j Y arms. We construct a graph G by activating each Y arms independently with probability $1 - e^{-(s+h)}$, and then picking independently a uniform pairing of the O arms and a uniform pairing of the Y arms.

D.2 Construction of a typical component

Let us now explain how to build a typical component of G , that is, we chose a vertex i uniformly at random and build a connected component which has the same law as the connected component of i in G . We define in the same time the exploration process (V_k) of that component. To avoid introducing irrelevant random variables, we also take an arbitrary ordering of the arms, so in the following, "the smallest arm" corresponds to the smallest arm for that order.

We may explore and construct a typical component of the graph G as follows. The (O or Y) arms may be in three states : S(leeping), D(ead) or A(wake). At each step, we denote

- S^O and S^Y the sets of sleeping O and Y arms, $S = S^O \cup S^Y$ is the set of sleeping arms.
- A^O and A^Y are the set of awaken O and Y arms, $A = A^O \cup A^Y$ is the set of awaken arms.
- D is the set of dead arms.
- g is the generation (i.e. the graph-distance to i), and the index g means "at generation g ", so for instance A_g is the set of active arms at generation g .

Step 0 $g = 0$, $k = 0$, S^O (resp. S^Y) is the set of all the O (resp Y) arms, $A_i = \emptyset$ for all $i \geq 0$, $D = \emptyset$, and $k = 0$, $V_k = V_0 = 0$.

Step 1 Chose a vertex i uniformly in $[Z]$. We then perform the following steps of the algorithm.

- With probability $1 - e^{-(s+h)}$, each arm of $[i]^Y$ goes independently into A_0^Y , say there are p of them.
- $A_0^O \leftarrow [i]^O$, $A_0 \leftarrow A_0^O \cup A_0^Y$, $A \leftarrow A_0$.
- $S^O \leftarrow S^O \setminus [i]^O$, $S^Y \leftarrow S^Y \setminus [i]^Y$, $S \leftarrow S^O \cup S^Y$.
- $g \leftarrow g + 1 = 1$.
- $k \leftarrow k + 1 = 1$.

Then, we let

$$V_k = V_1 = V_0 + d_i^O + p - 1 = d_i^O + p - 1.$$

Step 2 As long as $A \neq \emptyset$, or equivalently, as long as V has not reached -1 , we perform the following steps.

- As long as $A_g^O \neq \emptyset$, consider b the smallest arm in A_g^O and perform the following.
 1. Chose b' uniformly in $A^O \cup S^O \setminus \{b\}$, link b and b' , and let $D \leftarrow D \cup \{b, b'\}$.
 2. If $b' \in A_g^O$, we let $A_g^O \leftarrow A_g^O \setminus \{b, b'\}$, $A_g \leftarrow A_g^O \cup A_g^Y$, $A = \cup_{r \geq 0} A_r$, and update V_k to $V_k - 2$.
 3. If $b' \in A_{g+1}^O$, we let $A_g^O \leftarrow A_g^O \setminus \{b\}$, $A_g = A_g^O \cup A_g^Y$, $A_{g+1}^O \leftarrow A_{g+1}^O \setminus \{b'\}$, $A_{g+1} \leftarrow A_{g+1}^O \cup A_{g+1}^Y$, $A = \cup_{r \geq 0} A_r$, and update V_k to $V_k - 2$.
 4. If $b' \in S^O$, we let j the vertex to which it is attached, and perform the following.
 - $A_{g+1}^O \leftarrow A_{g+1}^O \cup [j]^O$.
 - With probability $1 - e^{-(s+h)}$, each arm of $[j]^Y$ goes into A_{g+1}^Y , say there are p of them.
 - $A_{g+1} \leftarrow A_{g+1}^O \cup A_{g+1}^Y$, $A \leftarrow \cup_{r \geq 1} A_r$.
 - $S^O \leftarrow S^O \setminus [j]^O$, $S^Y \leftarrow S^Y \setminus [j]^Y$, $S \leftarrow S^O \cup S^Y$.
 - We take another step of the exploration process

$$V_{k+1} = V_k + d_j^O + p - 2.$$

- Finally, $k \leftarrow k + 1$.
- As long as $A_g^Y \neq \emptyset$, proceed as above with obvious changes.
- If $A_g = 0$, we let $g \leftarrow g + 1$.

Step 3 When $A = \emptyset$, the algorithm stops.

Note that at each step, only A_g and A_{g+1} for some g can be non-empty. Moreover, it is easy to check that at the end of the algorithm, we have constructed a typical component of G . More precisely, we have constructed it generation by generation : first, we take a vertex uniformly at random. Then, we add the neighbors of that vertex (which form the first generation), then their non yet explored neighbors (the second generation), and so on.

D.2.1 Construction using uniform variables

Let us first introduce some notations. We call an increasing list a matrix \mathcal{H} with three rows such that $\mathcal{H}_{2,i} + \mathcal{H}_{3,i}$ is increasing in i .

Consider a set of arms $B = \{b_1 = (i_1, j_1), \dots, b_k = (i_k, j_k)\}$. We can associate to B an increasing list

$$\mathcal{H}(B) = \begin{pmatrix} b_1 & \dots & b_k \\ d_{i_1}^O & \dots & d_{i_k}^O \\ d_{i_1}^Y & \dots & d_{i_k}^Y \end{pmatrix},$$

implicitly assuming that B is labeled in such a way that $d_{i_1} \leq \dots \leq d_{i_k}$, i.e. such that the list is increasing. For the sake of definiteness, if ties occur, we order the columns in the order of arms.

Now, consider an increasing list \mathcal{H} of length k , and let, for $1 \leq r < k$, $I_r = [(r-1)/r, i/r)$, and $I_k = [(k-1)/k, 1]$. Then, we define

$$\begin{aligned} \phi_{\mathcal{H}}^1(x) &= \sum_{r=1}^k \mathcal{H}_{1,r} \mathbf{1}_{I_r}(x), \\ \phi_{\mathcal{H}}^2(x) &= \sum_{r=1}^k (\mathcal{H}_{2,r}, \mathcal{H}_{3,r}) \mathbf{1}_{I_r}(x), \\ \phi_{\mathcal{H}}^+(x) &= \sum_{r=1}^k (\mathcal{H}_{2,r} + \mathcal{H}_{3,r}) \mathbf{1}_{I_r}(x), \quad x \in [0, 1]. \end{aligned}$$

We will simply write ϕ_B^1 for $\phi_{\mathcal{H}(B)}^1$, and so on.

For a set $S = \{i_1, \dots, i_k\}$ of vertices, we also denote

$$\mathcal{H}(S) = \begin{pmatrix} i_1 & \dots & i_k \\ d_{i_1}^O & \dots & d_{i_k}^O \\ d_{i_1}^Y & \dots & d_{i_k}^Y \end{pmatrix},$$

implicitly assuming again that this list is ordered. We will also write e.g. ϕ_S^1 for $\phi_{\mathcal{H}(S)}^1$.

Remark D.1. The interest of these notations is that, when U is a uniform random variable on $[0, 1]$, $\phi_B^1(U)$, is a uniformly chosen arm in B , say j , $\phi_B^O(U)$ and $\phi_B^Y(U)$ are the number of O and Y arms of j , and $\phi_B^+(U)$ the total number of arms of j , whereas $\phi_S^1(U)$ is a uniformly chosen vertex in S .

Now, we will precise how we chose the arms or vertices at random, in order to bound by above the exploration process (V_k) . To this end, we take $U_0, U_{g,r}, U_{g,r}^O, U_{g,r}^Y, U'_{g,r,l}$ with $g, r, l \geq 1$ independent uniform r.v. on $[0, 1]$.

Step 0 We initialize the variables as before, and let $r = 1$ and $l = 1$.

Step 1 Choose $i = \phi_{[Z]}^1(U_0)$. Call $b_1, \dots, b_{d_i^Y}$ the d_i^Y Y ordered arms of i . Then, b_p goes in A_0^Y if and only if $U_{1,p} \leq 1 - e^{-(s+h)}$. We then update the other variables as above and let

$$V_1 = d_i^O + \sum_{p=1}^{d_i^Y} \mathbf{1}_{\{U_{1,p} \leq 1 - e^{-s}\}} - 1.$$

Step 2 As long as $A \neq \emptyset$, or equivalently, as long as V has not reached -1 , we perform the following steps.

1. If $A_g^O \neq \emptyset$, we chose the smallest $b \in A_g^O$ and proceed as follows.
 - If $U'_{g,r,l} \leq (\#A_g^O - 1) / (\#A^O + \#S^O - 1)$, then we take $b' = \phi_{A_g^O \setminus \{b\}}(U_{g,r}) \in A_g^O$, update the variables accordingly and V_k to $V_k - 2$. Also $l \leftarrow l + 1$.

- If $(\#A_g^O - 1)/(\#A_g^O + \#S^O - 1) < U'_{g,r,l} \leq (\#A^O - 1)/(\#A^O + \#S^O - 1)$, then we take $b' = \phi_{A_{g+1}^O}(U_{g,r}^O) \in A_{g+1}^O$, update the variables accordingly and V_k to $V_k - 2$. Also $l \leftarrow l + 1$.
- Else, we take $b' = \phi_{S^O}(U_{g,r}^O) \in S^O$. Call j the vertex to which b' is attached, and denote $b_1, \dots, b_{d_j^Y}$ its ordered Y arms. Then, b_p goes in A_{g+1}^Y if and only if $U_{g+1,p} \leq 1 - e^{-(s+h)}$. We then update the variables as above and let

$$V_{k+1} = V_k + d_j^O + \sum_{p=1}^{d_j^Y} \mathbb{1}_{\{U_{g+1,p} \leq 1 - e^{-(s+h)}\}} - 2.$$

Also $r \leftarrow r + 1$, $k \leftarrow k + 1$.

- If now $A_g^O = \emptyset$, then we let $r \leftarrow 1$ and start over Step 2 again.
2. If $A_g^O = \emptyset$, $A_g^Y \neq \emptyset$, we proceed as above with obvious changes.
 3. If $A_g = 0$, we let $g \leftarrow g + 1$, $r \leftarrow 1$, $l \leftarrow 1$.

Step 3 When $A = \emptyset$, the algorithm stops.

At a given generation g , the variables r and l account respectively for the number of new vertices visited and the number of corrections we observe.

D.2.2 Upper-bound for the exploration process

We will now define an upper-bound (\bar{V}) for the exploration process. We assume here that our set of Z particles is actually a subset of our initial N particles. This allows us to control the total number of arms on the particles.

Construction of new lists Consider the initial number of arms on the N particles a_1, \dots, a_N and let \mathcal{L} be the list of the a_i 's ranked in the increasing order (which is thus constituted of $N\mu^N(0)$ times 0, $N\mu^N(1)$ times 1, and so on). Consider as well the size-biased list \mathcal{L}' of size $K_N = N\langle\mu^{(N)}, k\rangle$ created by writing a_1 times a_1 , then a_2 times a_2 , etc., and reorder it in the increasing order.

- From the initial increasing list $\mathcal{H}([Z])$, we may consider another list \mathcal{H} created by replacing its second and third rows by the Z last elements of \mathcal{L} , to get a list called \mathcal{H} .
- From $\mathcal{H}(S^O)$, we may construct another increasing list \mathcal{H}^O as follows.
 - Replace the third row of $\mathcal{H}(S^O)$ by the $\#S^O$ last elements of \mathcal{L}' ;
 - delete its $\alpha(N)$ first columns (if there are that many, else keep only the last column);
 - subtract 1 to each term of its second row.
- From $\mathcal{H}(S^Y)$, we may construct another increasing list \mathcal{H}^Y as follows.
 - Replace the second and third rows of $\mathcal{H}(S^Y)$ by the $\#S^Y$ last elements of \mathcal{L}' ;
 - delete its $\alpha(N)$ first columns (if there are that many, else keep only the last column);
 - subtract 1 to each term of its third row.

Remark D.2. The key in the construction of these lists is that, when U is a uniform variable on $[0, 1]$, $(d^O, d^Y) = \phi_{[Z]}^2(U)$ and $(\delta^O, \delta^Y) = \phi_{\mathfrak{H}}^2(U)$ then $d^O \leq \delta^O$ and $d^Y \leq \delta^Y$. If now $(d^O, d^Y) = \phi_{\mathfrak{S}^O}^2(U)$ and $(\delta^O, \delta^Y) = \phi_{\mathfrak{H}^O}^2(U)$, then $d^O - 1 \leq \delta^O$ and $d^Y \leq \delta^Y$ as long as less than $\alpha(N)$ steps of the exploration process have been performed. If finally $(d^O, d^Y) = \phi_{\mathfrak{S}^Y}^2(U)$ and $(\delta^O, \delta^Y) = \phi_{\mathfrak{H}^Y}^2(U)$, then $d^O \leq \delta^O$ and $d^Y - 1 \leq \delta^Y$ as long as less than $\alpha(N)$ steps of the exploration process have been performed. This allows us to bound the exploration process defined above, and makes the proof of Lemma 7.8 below quite obvious.

Construction of the upper-bound We now define our upper-bound exploration process as follows.

Step 0 We first let $\bar{V}_0 = 0$, $g = 1$, $d_r^O = d_r^Y = 0$ for all $r \geq 1$.

Step 1 We then chose $(d^O, d_0^Y) = \phi_{\mathfrak{H}}^2(U_0)$. We let $d_1^O \leftarrow d^O$,

$$d^Y \leftarrow \sum_{p=1}^{d_0^Y} \mathbb{1}_{\{U_{1,p} \leq 1 - e^{-(s+h)}\}},$$

$$d_1^Y \leftarrow d_1^Y + d^Y = d^Y, \text{ and}$$

$$\bar{V}_1 = d^O + d^Y - 1.$$

Step 2 As long as \bar{V} has not reached -1 , or equivalently, as long as $d_g^O + d_g^Y \neq 0$ just after an update of g , we perform the following steps.

1. As long as $d_g^O \neq 0$, we let $(d^O, d_0^Y) = \phi_{\mathfrak{H}^O}^2(U_{g,r}^O)$, $d_{g+1}^O \leftarrow d_{g+1}^O + d^O$,

$$d^Y \leftarrow \sum_{p=1}^{d_0^Y} \mathbb{1}_{\{U_{g+1,p} \leq 1 - e^{-(s+h)}\}},$$

$$d_{g+1}^Y \leftarrow d_{g+1}^Y + d^Y, \text{ and}$$

$$\bar{V}_{k+1} = \bar{V}_k + d^O + d^Y - 1$$

and $r \leftarrow r + 1$, $k \leftarrow k + 1$.

2. When $d_g^O = 0$, we let $r \leftarrow 1$, and as long as $d_g^Y \neq 0$, we proceed as above with obvious change.
3. When $d_g^Y = 0$, we let $g \leftarrow g + 1$.

Step 3 When $\bar{V}_k = -1$, or equivalently, when $d_g^O + d_g^Y = 0$ just after an update of g , the algorithm stops.

Three measures and a Galton-Watson process For U a uniform r.v. on $[0, 1]$, let $(d^O, d^Y) = \phi_{\mathfrak{H}^O}^2(U)$ (resp. $(d^O, d^Y) = \phi_{\mathfrak{H}^Y}^2(U)$) and γ^O (resp. γ^Y) be the law of

$$\left(d^O, \sum_{p=1}^{d^Y} \mathbb{1}_{\{U_{1,p} \leq 1 - e^{-(s+h)}\}} \right).$$

For $(d^O, d^Y) = \phi_{\mathfrak{H}}^2(U)$ let also γ be the law of (d^O, d^Y) . Then, it is important to notice that \bar{V} is actually the walk associated to a two-type (O and Y) Galton-Watson tree

- visited by breadth-first-search, where O individuals are visited, at each generation, before Y individuals,
- with reproduction law γ for the ancestor,
- with reproduction laws γ^O and γ^Y for the O and Y individuals.

In particular, this implies that the equivalence “ $V_k = -1 \Leftrightarrow d_g^O + d_g^Y = 0$ just after an update of g ” holds.

Now, denote T the hitting time of -1 by V and \bar{T} the hitting time of -1 by \bar{V} . Let G_{gel} the number of particles in giant clusters in G . It is then easy to check the following.

Lemma D.3. *For $k \leq T \wedge \alpha(N)$, $V_k \leq \bar{V}_k$. In particular, $T \wedge \alpha(N) \leq \bar{T} \wedge \alpha(N)$, and*

$$\frac{1}{Z} \mathbb{E}(G_{\text{gel}}) \leq \mathbb{P}(\bar{T} > \alpha(N)).$$

Proof. The comparison between V and \bar{V} is easy but tedious. Using Remark D.2, we may indeed check that, at each step of the algorithm, the increments we add are bigger for \bar{V} than for V (when less than $\alpha(N)$ steps have been taken), and that we take (at least) more steps for \bar{V} than for V .

This result obviously implies that $T \wedge \alpha(N) \leq \bar{T} \wedge \alpha(N)$. Finally, note that T is precisely the size of the connected component of the vertex chosen at Step 0 of the algorithm, so $T > \alpha(N)$ if and only if this vertex is in the gel. Since this vertex is chosen uniformly at random, then

$$\mathbb{E} \left(\frac{1}{Z} G_{\text{gel}} \right) = \mathbb{P}(T > \alpha(N))$$

whence the result follows. \square

This shows precisely Lemma 7.8. What it remains to do now is to check the properties of the measures given in Section 7.3.1.

D.3 Convergence of the measures

We will now translate the previous result in the random setting we are interested in. Recall that we have fixed $t > 0$, $s > 0$, a small $h \in (0, t)$ and assume, by tightness, that $n_t^{(N)}$ and $\xi_{t,t-h}^{(N)}$ converge sharply to some n_t and $\xi_{t,t-h}$, in the notations of Lemma 5.7. We are thus dealing with

- a number $N_t = n_t^{(N)} N$ of particles,
- with empirical distribution of O and Y arms given by $\xi_{t,t-h}^{(N)}$,
- with empirical distribution of O arms given by $\pi_{t,t-h}^{(N)}(\cdot) = \sum_{j \geq 0} \xi_{t,t-h}^{(N)}(\cdot, j)$ (which is obviously the same as in Lemma 5.6).

Recall that $K_N = \langle \mu^{(N)}, k \rangle N$ is the total number of arms at time 0, and define

- $O_{t,t-h} = \langle \xi_{t,t-h}^{(N)}, i \rangle N_t$ the number of O arms, $o_{t,t-h}^{(N)} = O_{t,t-h} / K_N$,
- $Y_{t,t-h} = \langle \xi_{t,t-h}^{(N)}, j \rangle N_t$ the number of Y arms, $y_{t,t-h}^{(N)} = Y_{t,t-h} / K_N$.

Conditionally on $I_S(t)$ and $\xi_{t,t-h}^{(N)}$, the construction from the previous Section D.2.2 can be carried out. More precisely, for each instance of $I_S(t)$ and $\xi_{t,t-h}^{(N)}$, we obtain a random graph G , and, as above, laws γ , γ^O and γ^Y , which are deterministic functions of $I_S(t)$ (or merely $N_t = \#I_S(t)$) and $\xi_{t,t-h}^{(N)}$, and which we rather call γ_N , γ_N^O and γ_N^Y to insist on the dependence on N . We will first study their limit.

Recall from Lemma 5.1 that, uniformly for t in the compact sets of $(0, +\infty)$, there is a constant $c > 0$ such that, with high probability, $n_t^{(N)} > c$ and $y_{t,t-h}^{(N)} > c$.

An easy reasoning, as in the proof of Lemma 5.6 ensures that the particles of $I_S(t)$ have, at time $t-h > 0$, a number of active arms of order N , or, in other words, that there is a constant, say $c > 0$ again (which can also be chosen to be uniform for $t-h$ and t in the compacts of $(0, +\infty)$), such that $o_{t,t-h}^{(N)} > c$ w.h.p

Finally, the sharp convergence of $\xi_{t,t-h}^{(N)}$ ensures the convergence of the sequences $o_{t,t-h}^{(N)}$ and $y_{t,t-h}^{(N)}$ to some (random) $o_{t,t-h} \geq c$ and $y_{t,t-h} \geq c$, and the convergence of $\pi_{t,t-h}^{(N)}$ to some $\pi_{t,t-h}$.

Convergence of γ_N To construct a random variable with law γ_N , we may proceed as follows. First, consider M_N the CDF of the law of the initial number of arms of a particle picked uniformly at random in $[N]$ (i.e. M_N is the CDF of $\mu^{(N)}$). Take U a uniform r.v. on $[0, 1]$ independent from all the variables and (e_i) . Then

$$\left(M_N^{-1}(1 - n_t^{(N)} + n_t^{(N)}U), M_N^{-1}(1 - n_t^{(N)} + n_t^{(N)}U) \right)$$

has law γ_N , where M_N^{-1} is the generalized right-inverse of M_N .

Now, M_N converges a.e. to M , the CDF of μ . Hence (see e.g. the proof of Skorokhod's theorem in [6, Th. 25.6, p. 333]), $M_N^{-1} \rightarrow M^{-1}$ a.e. Then, it is easy to check, since U is uniform and $c_t > 0$, that

$$M_N^{-1}(1 - n_t^{(N)} + n_t^{(N)}U) \rightarrow M^{-1}(1 - n_t + n_tU), \quad \text{a.e.}$$

so, if γ denotes the law of $(M^{-1}(1 - n_t + n_tU), M^{-1}(1 - n_t + n_tU))$, then $\gamma_N \rightarrow \gamma$ weakly.

Convergence of γ_N^O Now, take (e_i) an independent family of exponential with parameter 1 random variables, independent from all the variables, and denote

$$B(k) = \sum_{i=1}^k \mathbb{1}_{\{e_i \leq s+h\}},$$

so $B(k)$ has a binomial $(k, 1 - e^{-(s+h)})$ distribution. Call R_N the (random) CDF of the law of $\hat{\pi}_{t,t-h}^{(N)}$. Let also Q_N be the CDF of a uniformly chosen element in \mathcal{L}' , that is of $\hat{\mu}^{(N)}$. Then, the variable

$$\left(R_N^{-1} \left(\frac{\alpha(N)}{N} + \left(1 - \frac{\alpha(N)}{N} \right) U \right), B \left(Q_N^{-1} \left(1 - o_{t,t-h}^{(N)} + \frac{\alpha(N)}{K_N} + \left(o_{t,t-h}^{(N)} - \frac{\alpha(N)}{K_N} \right) U \right) \right) \right)$$

has the same law¹⁴ (as random probability) as γ_N^O . The reasoning above can be easily adapted to take into account random measures, using e.g. Skorokhod's representation theorem, so γ_N^O converges to a measure γ^O which is the law of

$$\left(R^{-1}(U), B \left(Q^{-1}(1 - o_{t,t-h} + o_{t,t-h}U) \right) \right),$$

¹⁴except in the cases where $o_{t,t-h}^{(N)} < \alpha(N)/N$, but since $o_{t,t-h}^{(N)}$ is of order N , this obviously does not make any difference. To avoid cumbersome notations, we ignore this issue.

where $Q = \lim Q_N$ is the CDF of ν and $R = \lim R_N$ is the CDF of $\hat{\pi}_{t,t-h}$.

Convergence of γ_N^Y By still the same reasoning, γ_N^Y converges to γ^Y , the law of

$$\left(Q^{-1}(1 - y_{t,t-h} + y_{t,t-h}U), B\left(Q^{-1}(1 - y_{t,t-h} + y_{t,t-h}U)\right)\right).$$

This all shows the first properties of γ_N , γ_N^O and γ_N^Y of Section 7.3.1.

D.4 Bounds on the laws

To end the proof of the other properties of these measures, all it remains to do is to prove the bounds on the means of γ , γ^O and γ^Y . Note that the inequalities we write below are a.s., though, for notational simplicity, this will remain implicit.

Now, recall from Section D.3 that there is a constant $c > 0$ (depending only on the compact and μ) such that, with high probability $n_t^{(N)} > c$, $o_{t,t-h}^{(N)} > c$ and $y_{t,t-h}^{(N)} > c$. Now, by another reasoning analogous to the proof of Lemma 5.6 and using the fact that $\mu([3, +\infty)) > 0$ (because we assume that gelation occurs), there is a constant, say $c > 0$ again, such that $\pi_{t,t-h}^{(N)}(2) > c$. Once again, c is uniform for $t - h, t$ in the compacts of $(0, +\infty)$.

The marginals of a law λ are denoted λ^1 and λ^2 . We also take a constant M greater than $\langle \mu, k \rangle / c$ and $\langle \nu, k \rangle / c$.

- The mean of $M_N^{-1}(1 - n_t^{(N)} + n_t^{(N)}U)$ is less than $\langle \mu^{(N)}, k \rangle / n_t^{(N)}$, so, by Fatou's lemma,

$$\langle M^{-1}(1 - n_t + n_t U), k \rangle \leq \frac{1}{c} \langle \mu, k \rangle \leq M$$

and thus

$$\langle \gamma^1, k \rangle = \langle \gamma^2, k \rangle = \frac{\partial \phi}{\partial x}(1, 1) = \frac{\partial \phi}{\partial y}(1, 1) \leq M.$$

- The first marginal of γ_O is $\hat{\pi}_{t,t-h}$, so by Proposition 6.1 and Lemma 5.6,

$$\langle \gamma_O^1, k \rangle = \frac{\partial \phi_O}{\partial x}(1, 1) \leq 1.$$

- By easy computations

$$\langle \gamma_O^2, k \rangle = \frac{\partial \phi_O}{\partial y}(1, 1) \leq \frac{\langle \nu, k \rangle}{o_{t,t-h}}(1 - e^{-(s+h)}) \leq M(s+h).$$

- By the same reasoning

$$\langle \gamma_Y^1, k \rangle = \frac{\partial \phi_Y}{\partial x}(1, 1) \leq \frac{\langle \nu, k \rangle}{y_{t,t-h}} \leq M$$

and

$$\langle \gamma_Y^2, k \rangle = \frac{\partial \phi_Y}{\partial y}(1, 1) \leq \frac{\langle \nu, k \rangle}{y_{t,t-h}} s \leq M(s+h).$$

- We also need a lower-bound on

$$\frac{\partial^2 \phi_O}{\partial x^2}(0, 0) = 2\gamma_O(2, 0).$$

First, note that $\mathbb{P}(R^{-1}(U) = 2) = \hat{\pi}_{t,t-h}(2) \geq c > 0$. Now, conditionally on $R^{-1}(U) = 2$, we have, by definition and using Jensen's inequality, that

$$\begin{aligned} & \mathbb{P}(B(Q^{-1}(1 - o_{t,t-h} + o_{t,t-h}U)) = 0 | \gamma_O^1 = 2) \\ &= \mathbb{E} \left(\exp \left(-(s+h)Q^{-1}(1 - o_{t,t-h} + o_{t,t-h}U) \right) | R^{-1}(U) = 2 \right) \\ &\geq \exp - \left((s+h)\mathbb{E} \left(Q^{-1}(1 - o_{t,t-h} + o_{t,t-h}U) | R^{-1}(U) = 2 \right) \right) \\ &\geq \exp - \left((s+h)\mathbb{E} \left(Q^{-1}(1 - o_{t,t-h} + o_{t,t-h}U) \right) / \mathbb{P}(R^{-1}(U) = 2) \right) \\ &\geq \exp - ((s+h)\langle \nu, k \rangle / (o_{t,t-h}c)) \end{aligned}$$

and hence, since $s \leq 1$,

$$\frac{\partial^2 \phi_O}{\partial x^2}(0, 0) \geq \hat{\pi}_{t,t-h}(2) \exp - (\langle \nu, k \rangle / (o_{t,t-h}c)) \geq c \exp - (M/c) := a.$$

We have thus proved all the properties of γ_N , γ_N^O and γ_N^Y given in Section 7.3.1.

Bibliography

- [1] D. Aldous. Deterministic and stochastic models for coalescence (aggregation and coagulation): a review of the mean-field theory for probabilists. *Bernoulli*, 5(1):3–48, 1999.
- [2] J. Bertoin. *Random fragmentation and coagulation processes*, volume 102 of *Cambridge Studies in Advanced Mathematics*. Cambridge University Press, Cambridge, 2006.
- [3] J. Bertoin. Two solvable systems of coagulation equations with limited aggregations. *Ann. Inst. H. Poincaré Anal. Non Linéaire*, 26(6):2073–2089, 2009.
- [4] J. Bertoin and V. Sidoravicius. The structure of typical clusters in large sparse random configurations. *J. Stat. Phys.*, 135(1):87–105, 2009.
- [5] J. Bertoin, V. Sidoravicius, and M. E. Vares. A system of grabbing particles related to Galton-Watson trees. *Random Structures Algorithms*, 36(4):477–487, 2010.
- [6] P. Billingsley. *Probability and measure*. Wiley Series in Probability and Mathematical Statistics. John Wiley & Sons Inc., New York, 3rd edition, 1995.
- [7] M. Deaconu and E. Tanré. Smoluchowski’s coagulation equation: probabilistic interpretation of solutions for constant, additive and multiplicative kernels. *Ann. Scuola Norm. Sup. Pisa Cl. Sci. (4)*, 29(3):549–579, 2000.
- [8] P. B. Dubovskii and I. W. Stewart. Existence, uniqueness and mass conservation for the coagulation-fragmentation equation. *Math. Methods Appl. Sci.*, 19(7):571–591, 1996.
- [9] M. Dwass. The total progeny in a branching process and a related random walk. *J. Appl. Probability*, 6:682–686, 1969.
- [10] P. Erdős and A. Rényi. On the evolution of random graphs. *Magyar Tud. Akad. Mat. Kutató Int. Közl.*, 5:17–61, 1960.
- [11] P. Erdős and A. Rényi. On the evolution of random graphs. *Bull. Inst. Internat. Statist.*, 38:343–347, 1961.
- [12] M. H. Ernst, E. M. Hendriks, and R. M. Ziff. Kinetics of gelation and universality. *J. Phys. A*, 16(10):2293–2320, 1983.
- [13] M. H. Ernst, E. M. Hendriks, and R. M. Ziff. Coagulation processes with a phase transition. *J. Colloid Interface Sci.*, 97:266–277, 1984.

- [14] M. Escobedo, Ph. Laurençot, S. Mischler, and B. Perthame. Gelation and mass conservation in coagulation-fragmentation models. *J. Differential Equations*, 195(1):143–174, 2003.
- [15] M. Escobedo, S. Mischler, and B. Perthame. Gelation in coagulation and fragmentation models. *Comm. Math. Phys.*, 231(1):157–188, 2002.
- [16] S. N. Ethier and T. G. Kurtz. *Markov processes*. Wiley Series in Probability and Mathematical Statistics: Probability and Mathematical Statistics. John Wiley & Sons Inc., New York, 2005. Characterization and convergence.
- [17] N. Fournier and J.-S. Giet. Convergence of the Marcus-Lushnikov process. *Methodol. Comput. Appl. Probab.*, 6(2):219–231, 2004.
- [18] N. Fournier and Ph. Laurençot. Well-posedness of Smoluchowski’s coagulation equation for a class of homogeneous kernels. *J. Funct. Anal.*, 233(2):351–379, 2006.
- [19] N. Fournier and Ph. Laurençot. Marcus-Lushnikov processes, Smoluchowski’s and Flory’s models. *Stochastic Process. Appl.*, 119(1):167–189, 2009.
- [20] A. M. Golovin. The solution of the coagulation equation for cloud droplets in a rising air current. *Izv. Geophys. Ser.*, 5:482–487, 1963.
- [21] I. J. Good. Generalizations to several variables of Lagrange’s expansion, with applications to stochastic processes. *Proc. Cambridge Philos. Soc.*, 56:367–380, 1960.
- [22] A. Hammond and F. Rezakhanlou. Kinetic limit for a system of coagulating planar Brownian particles. *J. Stat. Phys.*, 124(2-4):997–1040, 2006.
- [23] A. Hammond and F. Rezakhanlou. The kinetic limit of a system of coagulating Brownian particles. *Arch. Ration. Mech. Anal.*, 185(1):1–67, 2007.
- [24] Th. E. Harris. *The theory of branching processes*. Dover Publications Inc., Mineola, NY, 2002.
- [25] S. Janson. The largest component in a subcritical random graph with a power law degree distribution. *Ann. Appl. Probab.*, 18(4):1651–1668, 2008.
- [26] S. Janson and M. Luczak. A new approach to the giant component problem. *Random Structures Algorithms*, 34(2):197–216, 2009.
- [27] I. Jeon. Existence of gelling solutions for coagulation-fragmentation equations. *Comm. Math. Phys.*, 194(3):541–567, 1998.
- [28] A. Joffe and M. Métivier. Weak convergence of sequences of semimartingales with applications to multitype branching processes. *Adv. in Appl. Probab.*, 18(1):20–65, 1986.
- [29] N. J. Kokholm. On Smoluchowski’s coagulation equation. *J. Phys. A*, 21(3):839–842, 1988.
- [30] V. Korolyuk and A. Swishchuk. *Semi-Markov random evolutions*, volume 308 of *Mathematics and its Applications*. Kluwer Academic Publishers, Dordrecht, 1995.

- [31] S. G. Krantz and H. R. Parks. *A primer of real analytic functions*. Birkhäuser Advanced Texts: Basler Lehrbücher. [Birkhäuser Advanced Texts: Basel Textbooks]. Birkhäuser Boston Inc., Boston, MA, second edition, 2002.
- [32] R. Lang and X.-X. Nguyen. Smoluchowski's theory of coagulation in colloids holds rigorously in the Boltzmann-Grad-limit. *Z. Wahrsch. Verw. Gebiete*, 54(3):227–280, 1980.
- [33] P. Laurençot and S. Mischler. On coalescence equations and related models. In *Degond, P., Pareschi, L. and Russo, G. (eds) : Modeling and computational methods for kinetic equations*. Birkhäuser, pages 321–356, 2004.
- [34] Ph. Laurençot. Global solutions to the discrete coagulation equations. *Mathematika*, 46(2):433–442, 1999.
- [35] Ph. Laurençot. On a class of continuous coagulation-fragmentation equations. *J. Differential Equations*, 167(2):245–274, 2000.
- [36] F. Leyvraz and H. R. Tschudi. Singularities in the kinetics of coagulation processes. *J. Phys. A*, 14(12):3389–3405, 1981.
- [37] A. A. Lushnikov. Certain New Aspects of the Coagulation Theory. *Izv. Atmos. Ocean Phys.*, 14:738–743, 1978.
- [38] A. H. Marcus. Stochastic coalescence. *Technometrics*, 10:133–143, 1968.
- [39] J. B. McLeod. On an infinite set of non-linear differential equations. *Quart. J. Math. Oxford Ser. (2)*, 13:119–128, 1962.
- [40] G. Menon and R. L. Pego. Approach to self-similarity in Smoluchowski's coagulation equations. *Comm. Pure Appl. Math.*, 57(9):1197–1232, 2004.
- [41] M. Merle and R. Normand. Self-organized criticality in a microscopic model for smoluchowski's equation (tentative title). In preparation.
- [42] M. Molloy and B. Reed. A critical point for random graphs with a given degree sequence. In *Proceedings of the Sixth International Seminar on Random Graphs and Probabilistic Methods in Combinatorics and Computer Science, "Random Graphs '93" (Poznań, 1993)*, volume 6, pages 161–179, 1995.
- [43] R. Normand. A model for coagulation with mating. *J. Stat. Phys.*, 137(2):343–371, 2009.
- [44] R. Normand and L. Zambotti. Uniqueness of post-gelation solutions of a class of coagulation equations. *Ann. Inst. H. Poincaré Anal. Non Linéaire*, 28(2):189–215, 2011.
- [45] J. R. Norris. Smoluchowski's coagulation equation: uniqueness, nonuniqueness and a hydrodynamic limit for the stochastic coalescent. *Ann. Appl. Probab.*, 9(1):78–109, 1999.
- [46] J. R. Norris. Cluster coagulation. *Comm. Math. Phys.*, 209(2):407–435, 2000.
- [47] B. Ráth and B. Tóth. Erdős-Rényi random graphs + forest fires = self-organized criticality. *Electron. J. Probab.*, 14:no. 45, 1290–1327, 2009.

-
- [48] S. Redner and F. Leyvraz. Non-universality and breakdown of scaling in a two-component coagulation model. *Phys. Rev. Lett.*, 57(2):163–166, 1986. Erratum : 57, 3123.
- [49] S. Redner and F. Leyvraz. Non-universal behavior and breakdown of scaling in two-species aggregation. *Phys. Rev. A*, 36(8):4033 – 4049, 1987.
- [50] S. Roelly-Coppoletta. A criterion of convergence of measure-valued processes: application to measure branching processes. *Stochastics*, 17(1-2):43–65, 1986.
- [51] D. Serre. *Matrices*, volume 216 of *Graduate Texts in Mathematics*. Springer, New York, 2nd edition, 2010.
- [52] F. Spitzer. *Principles of random walks*, volume 34. Springer-Verlag, New York, 2nd edition, 1976. Graduate Texts in Mathematics.
- [53] G. Stell and R.M. Ziff. Kinetics of polymer gelation. *J. Chem. Phys.*, 73:3492–3499, 1980.
- [54] R. van der Hofstad. Random graphs and complex networks. Available at <http://www.win.tue.nl/~rhofstad/NotesRGCN2010.pdf>.
- [55] H. J. van Roessel and M. Shirvani. Some results on the coagulation equation. *Non-linear Anal.*, 43(5, Ser. A: Theory Methods):563–573, 2001.
- [56] H. J. van Roessel and M. Shirvani. A formula for the post-gelation mass of a coagulation equation with a separable bilinear kernel. *Phys. D*, 222(1-2):29–36, 2006.
- [57] M. von Smoluchowski. Drei vortrage über diffusion, brownsche molekularbewegung und koagulation von kolloidteilchen. *Phys. Z.*, 17:557–571 and 585–599, 1916.
- [58] H. S. Wilf. *Generatingfunctionology*. Academic Press, 1994. Also available online at <http://www.math.upenn.edu/~wilf/gfology2.pdf>.
- [59] R. M. Ziff. Kinetics of polymerization. *J. Statist. Phys.*, 23(2):241–263, 1980.