Marcus-Lushnikov processes, Smoluchowski's and Flory's models

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Abstract

The Marcus-Lushnikov process is a finite stochastic particle system in which each particle is entirely characterized by its mass. Each pair of particles with masses x and y merges into a single particle at a given rate $K(x,y)$. We consider a *strongly gelling* kernel behaving as $K(x,y) = x^\alpha y + xy^\alpha$ for some $\alpha \in (0,1]$. In such a case, it is well-known that *gelation* occurs, that is, giant particles emerge. Then two possible models for hydrodynamic limits of the Marcus-Lushnikov process arise: the Smoluchowski equation, in which the giant particles are inert, and the Flory equation, in which the giant particles interact with finite ones.

We show that, when using a suitable cut-off coagulation kernel in the Marcus-Lushnikov process and letting the number of particles increase to infinity, the possible limits solve either the Smoluchowski equation or the Flory equation.

We also study the asymptotic behaviour of the largest particle in the Marcus-Lushnikov process without cut-off and show that there is only one giant particle. This single giant particle represents, asymptotically, the lost mass of the solution to the Flory equation.

Keywords : Marcus-Lushnikov process, Smoluchowski's coagulation equation, Flory's model, gelation.

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1 Introduction

We investigate the connection between a stochastic coalescence model, the Marcus-Lushnikov process, and two deterministic coagulation equations, the Smoluchowski and Flory equations. Recall that the Marcus-Lushnikov process [7, 8] is a finite stochastic system of coalescing particles while the Smoluchowski and Flory equations describe the evolution of the concentration $c(t,x)$ of particles of mass $x \in (0,\infty)$ at time $t \geq 0$ in an infinite system of coalescing particles. Both models depend on a *coagulation kernel* $K(x,y)$ describing the likeliness that two particles with respective masses x and y coalesce. When K increases sufficiently rapidly for large values of x and y, a singular phenomenon known as *gelation* occurs: giant particles (that is, particles with infinite mass) appear in finite time (see Jeon [6], Escobedo-Mischler-Perthame [3]). There is however a strong

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difference between the Smoluchowski and Flory equations: for the former, the giant particles are inert, while for the latter, the giant particles interact with the finite particles.
When $K(x,y)/y \longrightarrow 0$ as $y \to \infty$ for all $x \in (0,\infty)$, it is by now well-known that the Marcus-Lushnikov process converges to the solution of the Smoluchowski equation when the number of particles increases to infinity (see, e.g., Jeon [6] and Norris [9]). On the other hand, it has been shown in [5] that, if $K(x,y)/y \longrightarrow l(x) \in (0,\infty)$ as $y \to \infty$ for all $x \in (0,\infty)$, then the Marcus-Lushnikov process converges to the solution of the Flory equation.
Our aim in this paper is to study more precisely how this transition from the Smoluchowski equation to the Flory equation arises in the Marcus-Lushnikov process. For a coagulation kernel K of the form $K(x,y) \simeq xy^\alpha + x^\alpha y$ for some $\alpha \in (0,1]$, we consider a Marcus-Lushnikov process starting with n particles, with total mass m_n, where coalescence between particles larger than some threshold mass a_n is not allowed. We show that, in the limit of large n, m_n and a_n, this Marcus-Lushnikov process converges, up to extraction of a subsequence, either to the solution of the Flory equation or that of the Smoluchowski equation, according to the behaviour of a_n/m_n for large values of n.
We also study the behaviour of the largest particles in the Marcus-Lushnikov process without cut-off, and show that, in some sense, the total lost mass of the Flory equation is represented by one giant particle in the Marcus-Lushnikov process. Aldous [1] proved other results about giant particles for some similar (but more restrictive) kernels. We in fact obtain a much more precise result about the size of the largest particle after gelation, but we are not able to extend to our class of kernels his result about the largest particle before gelation.

2 Main result

Throughout the paper, a *coagulation kernel* is a function $K : (0,\infty)^2 \mapsto [0,\infty)$ such that $K(x,y) = K(y,x)$ for all $(x,y) \in (0,\infty)^2$. We denote by $\mathcal{M}_f^+$ the set of non-negative finite measures on $(0,\infty)$. Let us first recall the definition of the Marcus-Lushnikov process.

Definition 2.1 *Consider a coagulation kernel K, and an initial state $\mu_0 = m^{-1}\sum_{i=1}^n \delta_{x_i}$, with $n \geq 1$, $(x_1,...,x_n) \in (0,\infty)^n$ and $m = x_1 + ... + x_n$. A càdlàg $\mathcal{M}_f^+$-valued Markov process $(\mu_t)_{t\geq 0}$ is a Marcus-Lushnikov process associated with the pair (K,μ_0) if it a.s. takes its values in*

$$\mathcal{S}(n,m) := \left\{ \frac{1}{m} \sum_{i=1}^{k} \delta_{y_i},\ 1 \leq k \leq n,\ (y_i)_{1\leq i\leq k} \in (0,\infty)^k, \sum_{i=1}^{k} y_i = m \right\} \tag{2.1}$$

and its generator is given by

$$L^{K,\mu_0}\psi(\mu) = \sum_{i\neq j} \left\{ \psi\left[\mu + m^{-1}\left(\delta_{y_i+y_j} - \delta_{y_i} - \delta_{y_j}\right)\right] - \psi\left[\mu\right] \right\} \frac{K(y_i,y_j)}{2m} \tag{2.2}$$

for all measurable functions $\psi : \mathcal{M}_f^+ \mapsto \mathbb{R}$ and all states $\mu = m^{-1}\sum_{i=1}^k \delta_{y_i} \in \mathcal{S}(n,m)$.

This process is known to be well-defined and unique, without any assumption on K, see, e.g., Aldous [2, Section 4] or Norris [9, Section 4].

We now describe the Smoluchowski and Flory coagulation equations and first introduce the class of coagulation kernels to be considered in the sequel. As already mentioned, we will deal with kernels of the form $K(x,y) \simeq x^\alpha y + xy^\alpha$ for some $\alpha \in (0,1]$. More precisely, we assume the following:

Assumption (A_α): The coagulation kernel K is continuous on $(0,\infty)^2$ and there are $\alpha \in (0,1]$, $l \in \mathcal{C}((0,\infty))$, and positive real numbers $0 < c < C < \infty$ such that

$$\lim_{y\to\infty} K(x,y)/y = l(x)\,,$$

and

$$c\ (x^\alpha y + xy^\alpha) \le K(x,y) \le C\ (x^\alpha y + xy^\alpha) \quad \text{and} \quad c\ x^\alpha \le l(x) \le C\ x^\alpha \tag{2.3}$$

for all $(x,y) \in (0,\infty)^2$.

For such coagulation kernels, weak solutions to the Smoluchowski and Flory coagulation equations are then defined as follows:

Definition 2.2 *Consider a coagulation kernel K satisfying (A_α) for some $\alpha \in (0,1]$ and $\mu_0 \in \mathcal{M}_f^+$ such that $\langle \mu_0(dx), 1+x\rangle < \infty$. For $\phi : (0,\infty) \mapsto \mathbb{R}$, set*

$$\Delta\phi(x,y) := \phi(x+y) - \phi(x) - \phi(y)\,. \tag{2.4}$$

A family $(\mu_t)_{t\ge 0} \subset \mathcal{M}_f^+$ such that $t \mapsto \langle \mu_t(dx), x\rangle$ and $t \mapsto \langle \mu_t(dx), 1\rangle$ are non-increasing is a solution to:
(i) the Smoluchowski equation (S) if

$$\langle \mu_t, \phi\rangle = \langle \mu_0, \phi\rangle + \frac{1}{2}\int_0^t \langle \mu_s(dx)\mu_s(dy), K(x,y)\Delta\phi(x,y)\rangle\, ds \tag{2.5}$$

for all $\phi \in C_c([0,\infty))$ and $t \ge 0$;
(ii) the Flory equation (F) if for

$$\begin{aligned}\langle \mu_t, \phi\rangle &= \langle \mu_0, \phi\rangle + \frac{1}{2}\int_0^t \langle \mu_s(dx)\mu_s(dy), K(x,y)\Delta\phi(x,y)\rangle\, ds \\ &\quad - \int_0^t \langle \mu_s(dx), \phi(x)l(x)\rangle\, \langle \mu_0(dx) - \mu_s(dx), x\rangle\, ds \end{aligned}\tag{2.6}$$

for all $\phi \in C_c([0,\infty))$ and $t \ge 0$. Here and below, $C_c([0,\infty))$ denotes the space of continuous functions with compact support in $[0,\infty)$.

Note that the assumptions on K, $(\mu_t)_{t\ge0}$ and ϕ ensure that all the terms in (2.5) and (2.6) make sense.

Applying (2.5) (or (2.6)) with $\phi(x) = x$ (which does not belong to $C_c([0,\infty))$) would clearly give $\Delta\phi = 0$. Hence the total mass $\langle \mu_t(dx), x\rangle$ is *a priori* constant as time evolves. However, for coagulation kernels satisfying (A_α), the gelation phenomenon (that is, the loss of mass in finite time, or, equivalently, the appearance of particles with infinite mass) is known to occur [3, 6], which we recall now, together with other properties.

Proposition 2.3 *Consider a coagulation kernel K satisfying (A_α) for some $\alpha \in (0,1]$ and $\mu_0 \in \mathcal{M}_f^+$ such that $\langle \mu_0(dx), 1\rangle < \infty$ and $\langle \mu_0(dx), x\rangle = 1$. For any solution $(\mu_t)_{t\geq 0}$ of the Smoluchowski or Flory equation, the gelation time*

$$T_{gel} := \inf\{t \geq 0 \ : \ \langle \mu_t(dx), x\rangle < \langle \mu_0(dx), x\rangle\} \tag{2.7}$$

is finite with the following upper estimate (here c is defined in (A_α))

$$T_{gel} \leq \frac{\langle \mu_0(dx), x^{1-\alpha}\rangle}{(1-2^{-\alpha})c}\,.$$

If $(\mu_t)_{t\geq 0}$ solves the Flory equation, then $t \mapsto \langle \mu_t(dx), x\rangle$ is continuous and strictly decreasing on (T_{gel}, ∞),

$$\lim_{t\to\infty} \langle \mu_t(dx), x\rangle = 0 \quad \text{and} \quad \int_{T_{gel}+\varepsilon}^{\infty} \langle \mu_s(dx), x^{1+\alpha}\rangle\, ds < \infty$$

for all $\varepsilon > 0$.

The proof that gelation occurs is easier under (A_α) than the general proof of Escobedo-Mischler-Perthame [3], and we will sketch it in the next section. This result expresses that particles with infinite mass appear in finite time. Observe next that equations (S) and (F) do not differ until gelation. The additional term in equation (F) represents the loss of finite particles with mass x, proportionally to $l(x)$ and to the mass of the giant particles $\langle \mu_0(dx) - \mu_s(dx), x\rangle$.
Note that we are not able, and this is a well-known open problem, to show that $t \mapsto \langle \mu_t(dx), x\rangle$ is continuous at $t = T_{gel}$.
We finally consider a converging sequence of initial data.

Assumption (I): For each $n \in \mathbb{N}\setminus\{0\}$, we are given $\mu_0^n = m_n^{-1}\sum_{i=1}^n \delta_{x_i^n}$ for some $(x_1^n, ..., x_n^n) \in (0,\infty)^n$ and $m_n = x_1^n + ... + x_n^n$. We assume that there exists $\mu_0 \in \mathcal{M}_f^+$ such that $\langle \mu_0(dx), x\rangle = 1$ and $\lim_n \langle \mu_0^n(dx), \phi\rangle = \langle \mu_0(dx), \phi\rangle$ for all $\phi \in C_b([0,\infty))$, $C_b([0,\infty))$ denoting the space of continuous and bounded functions on $[0,\infty)$. In addition,

$$\lim_{\varepsilon\to 0} \sup_n \langle \mu_0^n(dx), \mathbb{1}_{(0,\varepsilon]}\rangle = 0\,.$$

We will actually not use explicitly all the assumptions in (I) and (A_α): some are just needed to apply the results of [5]. We now state a compactness result which follows from [5].

Proposition 2.4 *Consider a coagulation kernel K satisfying (A_α) for some $\alpha \in (0,1]$ and a sequence of initial conditions $(\mu_0^n)_{n\geq 1}$ satisfying (I). For each $a > 0$ and $n \geq 1$, we put $K_a := K\ \mathbb{1}_{(0,a]\times(0,a]}$ and denote by $(\mu_t^{n,a})_{t\geq 0}$ the Marcus-Lushnikov process associated with the pair (K_a, μ_0^n). The family $\{(\mu_t^{n,a})_{t\geq 0}\}_{n\geq 1, a>0}$ is tight in $\mathbb{D}([0,\infty), \mathcal{M}_f^+)$, endowed with the Skorokhod topology associated with the vague topology on $\mathcal{M}_f^+$.*

This proposition is proved in [5, Theorem 2.3-i] (with the choice of the subadditive function $\phi(x) = \sqrt{2C}(1+x)$, for which $K(x,y) \leq \phi(x)\phi(y)$). Actually, it is stated in [5] without the dependence on a, but the extension is straightforward.
Notice here that, if $a \geq m_n$, the Marcus-Lushnikov process $(\mu_t^{n,a})_{t\geq 0}$ reduces to the standard Marcus-Lushikov process associated with (K, μ_0^n).

We may finally state our main results. Recall that we assume the total mass of the system to be initially $\langle \mu_0(dx), x\rangle = 1$.

Theorem 2.5 *Consider a coagulation kernel K satisfying (A_α) for some $\alpha \in (0,1]$ and a sequence of initial conditions $(\mu_0^n)_{n\geq 1}$ satisfying (I). Consider also a sequence $(a_n)_{n\geq 1}$ of positive real numbers such that $\lim_n a_n = \infty$. For each $n \geq 1$, let $(\mu_t^{n,a_n})_{t\geq 0}$ be the Marcus-Lushnikov process associated with the pair (K_{a_n}, μ_0^n) where $K_{a_n} := K\, \mathbb{1}_{(0,a_n]\times(0,a_n]}$ and consider the weak limit $(\mu_t)_{t\geq 0}$ in $\mathbb{D}([0,\infty), \mathcal{M}_f^+)$ of a subsequence $\left\{(\mu_t^{n_k,a_{n_k}})_{t\geq 0}\right\}_{k\geq 1}$. Then $(\mu_t)_{t\geq 0}$ belongs a.s. to $C([0,\infty),\mathcal{M}_f^+)$ and enjoys the following properties:*

1. *Assume that $a_n = m_n$.*
   (i) Then $(\mu_t)_{t\geq 0}$ solves a.s. the Flory equation with coagulation kernel K and initial condition μ_0.
   (ii) Denote by $M_1^n(t) \geq M_2^n(t) \geq ...$ the ordered sizes of the particles in the Marcus-Lushnikov process $(\mu_t^{n,m_n})_{t\geq 0}$, and define the (a priori random) gelation time T_{gel} of $(\mu_t)_{t\geq 0}$ as in (2.7). Then for all $\eta > 0$ and $\beta > 0$,

$$\lim_{k\to\infty} E\left[\sup_{t\in[T_{gel}+\eta,\infty)}\left|\frac{M_1^{n_k}(t)}{m_{n_k}} - (1 - \langle \mu_t(dx), x\rangle)\right|\right] = 0, \tag{2.8}$$

$$\lim_{b\to\infty}\limsup_{k\to\infty} P\left[\left(\int_{T_{gel}}^{\infty}\frac{1}{m_{n_k}}\sum_{i\geq 2} M_i^{n_k}(s)\mathbb{1}_{[b,\infty)}\left(M_i^{n_k}(s)\right)ds\right) \geq \beta\right] = 0. \tag{2.9}$$

   Furthermore, there is a positive constant L depending only on K such that, for all $\eta > 0$ and $b > 1$,

$$\lim_{k\to\infty} E\left[\sup_{t\in[0,T_{gel}-\eta]}\frac{M_1^{n_k}(t)}{m_{n_k}}\right] = 0, \tag{2.10}$$

$$\limsup_{k\to\infty} E\left[\int_0^{T_{gel}}\left\langle \mu_s^{n_k,m_{n_k}}(dx), x\mathbb{1}_{[b,\infty)}(x)\right\rangle^2 ds\right] \leq \frac{L}{b^\alpha}. \tag{2.11}$$

2. *If $a_n/m_n \to 0$ as $n\to\infty$, then $(\mu_t)_{t\geq 0}$ solves a.s. the Smoluchowski equation with coagulation kernel K and initial condition μ_0.*

3. *If $a_n/m_n \to \gamma \in (0,1)$ as $n\to\infty$, then $(\mu_t)_{t\in[0,T_1)}$ solves a.s. the Flory equation with coagulation kernel K and initial condition μ_0 where*

$$T_1 := \inf\{t > 0 \;:\; 1 - \langle \mu_t(dx), x\rangle \geq \gamma\}. \tag{2.12}$$

Point 1-(i) is proved in [5, Theorem 2.3-ii]. Remark that (2.10) is almost obvious while (2.11) gives an estimate on the tail of the mass distribution before gelation. The most interesting estimate is of course (2.8) which shows that, for $t > T_{gel}$, the largest particle in the Marcus-Lushnikov process without cut-off occupates a positive fraction of the total mass of the system with a precise asymptotic. Finally, (2.9) shows that, in some sense, there is only one giant particle after gelation: the other particles are rather small. Other results about the largest particles for the kernel

$$K(x,y) = \frac{2(xy)^{1+\alpha}}{(x+y)^{1+\alpha} - x^{1+\alpha} - y^{1+\alpha}},$$

which satisfies (A_α), were obtained by Aldous [1]. He however did not show that, after gelation, the size of the largest particle is of order εm_n.
Point 2 seems to be new, and quite interesting. Indeed, we allow arbitrary cut-off sequences (a_n) which increase more slowly than (m_n).
Finally, Point 3 can be explained in the following way: assume that $a_n = \gamma m_n$ for all $n \geq 1$ and some $\gamma \in (0,1)$ and that there is only one giant particle in $(\mu_t^{n,m_n})_{t\geq 0}$. In that situation, we then clearly have $(\mu_t^{n,\gamma m_n})_{t\in[0,T_1^n]} = (\mu_t^{n,m_n})_{t\in[0,T_1^n]}$, where T_1^n is the first time at which the giant particle has a size greater than γm_n, i.e., it occupates a fraction γ of the total mass of the system. Thus, $(\mu_t^{n,\gamma m_n})_{t\in[0,T_1^n]}$ should converge to $(\mu_t)_{t\in[0,T_1]}$, where μ solves the Flory equation, and T_1 is the first time for which the giant particle occupates a fraction γ of the total mass in the Flory model.
The proof of Theorem 2.5 is given in Section 4, after establishing some properties of solutions to the Smoluchowski and Flory coagulation equations in the next section. The final section of the paper is devoted to numerical illustrations.

3 Properties of solutions to (S) and (F)

Throughout this section, K is a coagulation kernel satisfying (A_α) for some $\alpha \in (0,1]$ and μ_0 belongs to $\mathcal{M}_f^+$ with total mass $\langle \mu_0(dx), x\rangle = 1$.

Proof of Proposition 2.3. Let $(\mu_t)_{t\geq 0}$ be a solution to the Smoluchowski equation (S) or the Flory equation (F), and define $T_{gel} \in (0,\infty]$ by (2.7). Classical approximation arguments allow us to use (2.5) and (2.6) with $\phi(x) = x^{1-\alpha}$. Indeed, it suffices to approximate ϕ by a sequence of functions in $C_c([0,\infty))$ and to pass to the limit, using the first inequality in

$$\min(x,y)^{1-\alpha} \geq x^{1-\alpha} + y^{1-\alpha} - (x+y)^{1-\alpha} \geq (2 - 2^{1-\alpha})\ \min(x,y)^{1-\alpha}\,, \tag{3.1}$$

which warrants that $K(x,y)|\Delta\phi(x,y)| \leq 2Cxy$ by (A_α). We deduce from (2.5), (2.6), and the second inequality in (3.1) that, for all $t \geq 0$,

$$\begin{aligned} \left\langle \mu_t(dx), x^{1-\alpha}\right\rangle \ &\leq\ \left\langle \mu_0(dx), x^{1-\alpha}\right\rangle \\ &\quad -\frac{2-2^{1-\alpha}}{2}\int_0^t \left\langle \mu_s(dx)\mu_s(dy), K(x,y)\min(x,y)^{1-\alpha}\right\rangle\ ds. \end{aligned}$$

By virtue of (2.3), $K(x,y)\min(x,y)^{1-\alpha} \geq cxy$, whence

$$(1-2^{-\alpha})c\int_0^t \langle \mu_s(dx), x\rangle^2\ ds \leq \left\langle \mu_0(dx), x^{1-\alpha}\right\rangle \tag{3.2}$$

for all $t \geq 0$. Since $\langle \mu_s(dx), x\rangle = \langle \mu_0(dx), x\rangle = 1$ for all $s \in [0, T_{gel})$, we realize that T_{gel} has to be finite for (3.2) to hold true. A further consequence of (3.2) is that

$$(1-2^{-\alpha})cT_{gel} = (1-2^{-\alpha})c\int_0^{T_{gel}} \langle \mu_s(dx), x\rangle^2\ ds \leq \left\langle \mu_0(dx), x^{1-\alpha}\right\rangle,$$

whence

$$T_{gel} \leq \frac{\left\langle \mu_0(dx), x^{1-\alpha}\right\rangle}{(1-2^{-\alpha})c}\,.$$

It also follows from (3.2) that $t \longmapsto \langle \mu_t(dx), x \rangle$ belongs to $L^2(0,\infty)$ which, together with the monotonicity and non-negativity of $t \longmapsto \langle \mu_t(dx), x \rangle$ implies that $\langle \mu_t(dx), x \rangle \longrightarrow 0$ as $t \to \infty$.

We now assume that $(\mu_t)_{t\geq 0}$ solves the Flory equation (F) and prove that, for all $\varepsilon > 0$,

$$\int_{T_{gel}+\varepsilon}^{\infty} \left\langle \mu_t(dx), x^{1+\alpha} \right\rangle \, dt < \infty. \tag{3.3}$$

To do so, we apply (2.6) with the choice $\phi_A(x) = \min(x, A)$ for some positive real number A. Since $\Delta\phi_A$ is non-positive, we get

$$\langle \mu_t, \phi_A \rangle \leq \langle \mu_0, \phi_A \rangle - \int_0^t \langle \mu_s(dx), \min(x,A) l(x) \rangle \left(1 - \langle \mu_s(dx), x \rangle\right) \, ds.$$

Since $1 = \langle \mu_0(dx), x \rangle \geq \langle \mu_s(dx), x \rangle$ we may let $A \to \infty$ and $t \to \infty$ in the above inequality and use the Fatou lemma to deduce that

$$\int_0^{\infty} \langle \mu_s(dx), x l(x) \rangle \left(1 - \langle \mu_s(dx), x \rangle\right) \, ds \leq 1. \tag{3.4}$$

Let $\varepsilon > 0$. On the one hand, putting

$$\delta_\varepsilon := \inf_{t \geq T_{gel}+\varepsilon} \left\{1 - \langle \mu_t(dx), x \rangle\right\},$$

it follows from the definition (2.7) of T_{gel} that $\delta_\varepsilon > 0$. On the other hand, $x l(x) \geq c x^{1+\alpha}$ by (A_α). We therefore infer from (3.4) that

$$\int_{T_{gel}+\varepsilon}^{\infty} \left\langle \mu_s(dx), x^{1+\alpha} \right\rangle ds \leq \frac{1}{c\delta_\varepsilon},$$

whence (3.3).

We now check that $t \mapsto \langle \mu_t(dx), x \rangle$ is continuous on (T_{gel}, ∞). Using once more (2.6) with the choice $\phi_A(x) = \min(x, A)$, we obtain for $T_{gel} < s < t$

$$\begin{aligned} \langle \mu_t - \mu_s, \phi_A \rangle &= \frac{1}{2} \int_s^t \langle \mu_\tau(dx) \mu_\tau(dy), K(x,y) \Delta\phi_A(x,y) \rangle \, d\tau \\ &\quad - \int_s^t \langle \mu_\tau(dx), \phi_A(x) l(x) \rangle \left(1 - \langle \mu_\tau(dx), x \rangle\right) d\tau. \end{aligned}$$

Clearly $\Delta\phi_A(x,y) \to 0$ as $A \to \infty$ for all $(x,y) \in (0,\infty)^2$ while (A_α) warrants that

$$K(x,y) |\Delta\phi_A(x,y)| \leq C(x^\alpha y + x y^\alpha) \min(x,y) \leq C(x^{1+\alpha} y + x y^{1+\alpha}).$$

Using (3.3) and the Lebesgue dominated convergence theorem, we obtain

$$\langle \mu_t(dx) - \mu_s(dx), x \rangle = -\int_s^t \langle \mu_\tau(dx), x l(x) \rangle \left(1 - \langle \mu_\tau(dx), x \rangle\right) d\tau\,. \tag{3.5}$$

Using again (3.3) and that $\left| \langle \mu_\tau(dx), x l(x) \rangle \left(1 - \langle \mu_\tau(dx), x \rangle\right) \right| \leq C \left\langle \mu_\tau(dx), x^{1+\alpha} \right\rangle$ by (A_α), we conclude that $t \longmapsto \langle \mu_t(dx), x \rangle$ is continuous on (T_{gel}, ∞).

It remains to check that $t \longmapsto \langle \mu_t(dx), x \rangle$ is strictly decreasing on $t \in (T_{gel}, \infty)$. According to (3.5) this is true as long as $\mu_\tau \neq 0$ for $T_{gel} < s < \tau < t$: it thus suffices to show that $\mu_t \neq 0$ for all $t \geq 0$. For that purpose, we take $\phi(x) = x 1\!\!1_{(0,A]}(x)$ in (2.6) where $A > 0$ is chosen so that $\left\langle \mu_0(dx), x 1\!\!1_{(0,A]}(x) \right\rangle > 0$ (such an A always exists as $\langle \mu_0(dx), x \rangle = 1$). Thanks to (2.3), we get

$$\begin{aligned} \frac{d}{dt} \langle \mu_t, \phi \rangle &\geq - \left\langle \mu_t(dx)\mu_t(dy), K(x,y) x 1\!\!1_{(0,A]}(x) \right\rangle - \left\langle \mu_t(dx), x l(x) 1\!\!1_{(0,A]}(x) \right\rangle \\ &\geq -C \left\langle \mu_t(dx)\mu_t(dy), (x^{\alpha+1}y + x^2 y^\alpha) 1\!\!1_{(0,A]}(x) \right\rangle - C \left\langle \mu_t(dx), x^{1+\alpha} 1\!\!1_{(0,A]}(x) \right\rangle \\ &\geq -C(A^\alpha + A) \langle \mu_t, \phi \rangle \langle \mu_t(dx), x + x^\alpha \rangle - C A^\alpha \langle \mu_t, \phi \rangle . \end{aligned} \tag{3.6}$$

Since $t \longmapsto \langle \mu_t(dx), x \rangle$ and $t \longmapsto \langle \mu_t(dx), x^\alpha \rangle$ are non-increasing and $\langle \mu_0(dx), x + x^\alpha \rangle < \infty$, we conclude that

$$\frac{d}{dt} \langle \mu_t, \phi \rangle \geq -C_A \left(1 + \langle \mu_0(dx), x^\alpha \rangle\right) \langle \mu_t, \phi \rangle \geq -C_{A,\mu_0} \langle \mu_t, \phi \rangle$$

for all $t \geq 0$ for some constant $C_{A,\mu_0} > 0$. Consequently, $\langle \mu_t, \phi \rangle > 0$ for all $t \geq 0$ as the choice of A warrants that $\langle \mu_0, \phi \rangle > 0$, and the proof of Proposition 2.3 is complete. □

Next, as a preliminary step towards the proof of Theorem 2.5 Point 1-(ii), we show that solutions to the Smoluchowski and Flory coagulation equations do not coincide after the gelation time.

Corollary 3.1 *Let $(\mu_t)_{t\geq 0}$ and $(\nu_t)_{t\geq 0}$ be solutions to the Smoluchowski equation (S) and the Flory equation (F), respectively, (with the same coagulation kernel K and initial condition μ_0), and assume further their respective gelation times coincide, that is,*

$$T_{gel} := \inf\{t \geq 0 \ : \ \langle \mu_t(dx), x \rangle < \langle \mu_0(dx), x \rangle\} = \inf\{t \geq 0 \ : \ \langle \nu_t(dx), x \rangle < \langle \mu_0(dx), x \rangle\} .$$

Then, for each $\varepsilon > 0$, there exists $s_\varepsilon \in (T_{gel}, T_{gel} + \varepsilon)$ such that $\mu_{s_\varepsilon} \neq \nu_{s_\varepsilon}$.

Proof. Consider $\varepsilon > 0$.
Either $t \longmapsto \left\langle \mu_t(dx), x^{1+\alpha} \right\rangle$ does not belong to $L^1\left(T_{gel} + (\varepsilon/2), T_{gel} + \varepsilon\right)$ and μ_t cannot coincide with ν_t on $(T_{gel} + (\varepsilon/2), T_{gel} + \varepsilon)$ since $t \longmapsto \left\langle \nu_t(dx), x^{1+\alpha} \right\rangle$ belongs to $L^1\left(T_{gel} + (\varepsilon/2), T_{gel} + \varepsilon\right)$ by Proposition 2.3.
Or $t \longmapsto \left\langle \mu_t(dx), x^{1+\alpha} \right\rangle$ belongs to $L^1\left(T_{gel} + (\varepsilon/2), T_{gel} + \varepsilon\right)$ and it is not difficult to check that this property and (2.5) entail that $\langle \mu_t(dx), x \rangle = \left\langle \mu_{T_{gel}+(\varepsilon/2)}(dx), x \right\rangle$ for $t \in [T_{gel} + (\varepsilon/2), T_{gel} + \varepsilon]$: indeed, take $\phi_A(x) = \min(x, A)$ in (2.5) and pass to the limit as $A \to \infty$ using that $\Delta\phi_A(x,y) \to 0$ and the time integrability of $t \longmapsto \left\langle \mu_t(dx), x^{1+\alpha} \right\rangle$. Owing to the strict monotonicity of $t \longmapsto \langle \nu_t(dx), x \rangle$ established in Proposition 2.3, the previous property of μ_t excludes that $\mu_t = \nu_t$ for all $t \in (T_{gel} + (\varepsilon/2), T_{gel} + \varepsilon)$ and completes the proof of Corollary 3.1. □

4 Proof of the main results

We fix a coagulation kernel K satisfying (A_α) for some $\alpha \in (0,1]$ and a sequence of initial data $(\mu_0^n)_{n\geq 1}$ satisfying (I). Next, for $a > 0$ and $n \geq 1$, we put $K_a(x,y) := K(x,y) 1\!\!1_{(0,a]\times(0,a]}$ and denote by $(\mu_t^{n,a})_{t\geq 0}$ the Marcus-Lushnikov process associated with the pair (K_a, μ_0^n). According to Definition 2.1 we may write

$$\mu_t^{n,a} = \frac{1}{m_n} \sum_i \delta_{M_i^{n,a}(t)} \quad \text{with} \quad M_1^{n,a}(t) \geq M_2^{n,a}(t) \geq M_3^{n,a}(t) \geq ... \tag{4.1}$$

for all $t \geq 0$, $n \geq 1$, and $a > 0$.
We next recall that the space $\mathbb{D}([0,\infty), \mathcal{M}_f^+)$ is endowed with the Skorokhod topology associated with the vague convergence topology on $\mathcal{M}_f^+$ (see Ethier-Kurtz [4] for further information), and denote by d a distance on $\mathcal{M}_f^+$ metrizing the vague convergence topology.
Marcus-Lushnikov processes have some martingale properties, which are immediately obtained from (2.2), see also [9, Section 4].

Lemma 4.1 *For all $\phi \in L^\infty_{loc}(0,\infty)$ and $t \geq 0$, we have*

$$\begin{aligned}
\langle \mu_t^{n,a}, \phi\rangle &= \langle \mu_0^n, \phi\rangle + O_t^{n,a}(\phi) \\
&\quad + \frac{1}{2m_n^2} \int_0^t \sum_{i\neq j} K_a(M_i^{n,a}(s), M_j^{n,a}(s)) \Delta\phi(M_i^{n,a}(s), M_j^{n,a}(s))\ ds \\
&= \langle \mu_0^n, \phi\rangle + O_t^{n,a}(\phi) + \frac{1}{2}\int_0^t \langle \mu_s^{n,a}(dx)\mu_s^{n,a}(dy), K_a(x,y)\Delta\phi(x,y)\rangle\ ds \\
&\quad - \frac{1}{2m_n}\int_0^t \langle \mu_s^{n,a}(dx), K_a(x,x)\Delta\phi(x,x)\rangle\ ds
\end{aligned} \tag{4.2}$$

where $\Delta\phi$ is defined in (2.4), and $O^{n,a}(\phi)$ is a martingale starting from 0 with (predictable) quadratic variation

$$\begin{aligned}
\langle O^{n,a}(\phi)\rangle_t &= \frac{1}{2m_n}\int_0^t \left\langle \mu_s^{n,a}(dx)\mu_s^{n,a}(dy), K_a(x,y)\left[\Delta\phi(x,y)\right]^2\right\rangle\ ds \\
&\quad - \frac{1}{2m_n^2}\int_0^t \left\langle \mu_s^{n,a}(dx), K_a(x,x)\left[\Delta\phi(x,x)\right]^2\right\rangle\ ds.
\end{aligned}$$

Furthermore, if $\phi : (0,\infty) \to \mathbb{R}$ is a subadditive function, that is, $\phi(x+y) \leq \phi(x) + \phi(y)$ for $(x,y) \in (0,\infty)^2$, then $t \mapsto \langle \mu_t^{n,a}, \phi\rangle$ is a.s. a non-increasing function.

We carry on with some easy facts.

Lemma 4.2 *Let $(a_n)_{n\geq 1}$ be a sequence of positive numbers. Then any weak limit $(\mu_t)_{t\geq 0}$ of the sequence $\{(\mu_t^{n,a_n})_{t\geq 0}\}_{n\geq 1}$ belongs a.s. to $C([0,\infty), \mathcal{M}_f^+)$, and both $t \mapsto \langle \mu_t(dx), x\rangle$ and $t \mapsto \langle \mu_t(dx), 1\rangle$ are a.s. non-increasing functions. Furthermore,*

$$\sup_{n\geq 1}\sup_{t\geq 0} \langle \mu_t^{n,a_n}(dx), 1+x\rangle = \kappa := \sup_n \langle \mu_0^n(dx), 1+x\rangle < \infty, \tag{4.3}$$

and for all $\phi \in C_c([0,\infty))$ and $T > 0$,

$$\lim_{n\to\infty} E\left[\sup_{t\in[0,T]} \left| \frac{1}{2m_n}\int_0^t \langle \mu_t^{n,a_n}(dx), K_{a_n}(x,x)\Delta\phi(x,x)\rangle\ ds \right|\right] = 0, \tag{4.4}$$

$$\lim_{n\to\infty} E\left[\sup_{t\in[0,T]} \left(O_t^{n,a_n}(\phi)\right)^2\right] = 0. \tag{4.5}$$

Proof. First, if $\phi \in C_b([0,\infty))$, the jumps of $\langle \mu_t^{n,a_n}, \phi\rangle$ are of the form $m_n^{-1}\Delta\phi(x,y)$ and clearly converge to zero as $n\to\infty$ since $m_n\to\infty$. Hence any weak limit $(\mu_t)_{t\geq 0}$ belongs to $C([0,\infty),\mathcal{M}_f^+)$ a.s.

Consider next a family $(\mathcal{X}_b)_{b>0}$ of continuous non-increasing functions on $(0,\infty)$ such that $\mathcal{X}_b(x)=1$ for $x\leq b$ and $\mathcal{X}_b(x)=0$ for $x\geq b+1$, and a non-negative subadditive function ϕ. Then, on the one hand, $\phi\mathcal{X}_b$ is also subadditive and Lemma 4.1 ensures that $t\longmapsto \langle \mu_t^{n,a_n}, \phi\mathcal{X}_b\rangle$ is a.s. non-increasing for all $n\geq 1$. On the other hand, since $\phi\mathcal{X}_b\in C_c([0,\infty))$, it follows from the definition of $(\mu_t)_{t\geq 0}$ that there is a subsequence $(n_k)_{k\geq 1}$, $n_k\to\infty$, such that $\left\{\left(\left\langle \mu_t^{n_k,a_{n_k}}, \phi\mathcal{X}_b\right\rangle\right)_{t\geq 0}\right\}_{k\geq 1}$ converges in law towards $(\langle \mu_t,\phi\mathcal{X}_b\rangle)_{t\geq 0}$ for each fixed $b>0$ as $k\to\infty$. Therefore, $t\longmapsto\langle\mu_t,\phi\mathcal{X}_b\rangle$ is a.s. non-increasing for each $b>0$. Since $(\langle\mu_t,\phi\mathcal{X}_b\rangle)_{b>0}$ converges to $\langle\mu_t,\phi\rangle$ as $b\to\infty$ for each $t\geq 0$, we conclude that $t\mapsto\langle\mu_t,\phi\rangle$ is a.s. non-increasing. Applying this result to $\phi(x)=1$ and $\phi(x)=x$, we obtain that both $t\mapsto\langle\mu_t(dx),x\rangle$ and $t\mapsto\langle\mu_t(dx),1\rangle$ are a.s. non-increasing functions of time.

Next, since $x\mapsto 1+x$ is subadditive, Lemma 4.1 implies that we have a.s. $\langle\mu_t^{n,a_n}(dx),1+x\rangle\leq\langle\mu_0^n(dx),1+x\rangle$ for $n\geq 1$ and $t\geq 0$, and $\langle\mu_0^n(dx),1+x\rangle$ is bounded uniformly with respect to n by assumption (I).

Consider finally $\phi\in C_c([0,\infty))$ with support included in $[0,R]$ for some $R>0$. By (2.3), $|K_{a_n}(x,x)\Delta\phi(x,x)|\leq 6C\|\phi\|_{L^\infty}\ R^{1+\alpha}$, whence

$$|\langle\mu_t^{n,a_n}(dx),K_{a_n}(x,x)\Delta\phi(x,x)\rangle|\leq 6C\kappa\|\phi\|_{L^\infty}\ R^{1+\alpha}\quad \text{a.s.}$$

by (4.3), from which (4.4) readily follows since $m_n\to\infty$. By a similar argument, we establish that $E\left[\langle O^{n,a_n}(\phi)\rangle_t\right]\longrightarrow 0$ as $n\to\infty$, which implies (4.5) by Doob's inequality. □

We now prove a fundamental estimate which provides a control on the large masses contained in $\mu_t^{n,a}$.

Lemma 4.3 *There exists a positive real number L depending only on c and α in (A_α) such that*

$$E\left[\int_0^\infty \frac{1}{m_n^2}\sum_{i\neq j} M_i^{n,a}(s)M_j^{n,a}(s)1\!\!1_{[b,a]}\left(M_i^{n,a}(s)\right)1\!\!1_{[b,a]}\left(M_j^{n,a}(s)\right)\ ds\right]\leq\frac{L}{b^\alpha} \tag{4.6}$$

for all $n\geq 1$, $a>0$, and $b\in(0,a)$, the $M_i^{n,a}$ being defined in (4.1).

Proof. To prove this estimate, we use (4.2) with $\phi(x)=x^{1-\alpha}\min(x,b)^\alpha$ for some $b\in(0,a)$. We first notice that $\langle\mu_0^n,\phi\rangle\leq\langle\mu_0^n(dx),x\rangle=1$ and $\langle\mu_t^{n,a},\phi\rangle\geq 0$ for all $t\geq 0$ and $n\geq 1$. In addition, ϕ is subadditive so that $\Delta\phi(x,y)$ is always non-positive and we infer from (2.3) and (3.1) that

$$K_a(x,y)\Delta\phi(x,y)\leq -(2-2^{1-\alpha})cb^\alpha xy1\!\!1_{[b,a]}(x)1\!\!1_{[b,a]}(y)$$

for $(x,y)\in(0,\infty)^2$. Taking expectations in (4.2) and using the above inequalities, we obtain

$$0\leq 1-\frac{b^\alpha}{L}E\left[\int_0^t \frac{1}{m_n^2}\sum_{i\neq j} M_i^{n,a}(s)M_j^{n,a}(s)1\!\!1_{[b,a]}\left(M_i^{n,a}(s)\right)1\!\!1_{[b,a]}\left(M_j^{n,a}(s)\right)\ ds\right]$$

for all $t \geq 0$, with $1/L := c(1-2^{-\alpha})$. We conclude the proof by letting $t \to \infty$ in the previous inequality. □

We now turn to the proof of Theorem 2.5 and first recall that Point 1-(i) is included in [5, Theorem 2.3-ii] as $(\mu_t^{n,m_n})_{t\geq 0}$ is the standard Marcus-Lushnikov process associated with (K, μ_0^n).

Proof of Point 2 of Theorem 2.5. Let $(a_n)_{n\geq 1}$ be a sequence of positive real numbers satisfying $a_n \to \infty$ and $a_n/m_n \to 0$ as $n \to \infty$. We consider the limit $(\mu_t)_{t\geq 0}$ of a subsequence $\left\{(\mu_t^{n_k,a_{n_k}})_{t\geq 0}\right\}_{k\geq 1}$ in the sense that a.s.

$$\lim_{k\to\infty} \sup_{[0,T]} d(\mu_t^{n_k,a_{n_k}}, \mu_t) = 0 \quad \text{for all} \quad T > 0\,, \tag{4.7}$$

the existence of such a limit being guaranteed by Proposition 2.4, Lemma 4.2 (which ensures the time continuity of μ_t) and the Skorokhod representation theorem. We now aim at showing that $(\mu_t)_{t\geq 0}$ solves a.s. the Smoluchowski equation (S) and proceed in two steps.
Step 1. We first deduce from Lemma 4.3 that

$$E\left[\int_0^T \left\langle \mu_t^{n,a_n}(dx), x\mathbb{1}_{[b,a_n]}(x)\right\rangle^2\, dt\right] \leq \left(\frac{L}{b^\alpha} + T\frac{a_n}{m_n}\right) \tag{4.8}$$

for all $b > 0$, $n \geq 1$, and $T > 0$. Indeed, we have a.s., for all $t \geq 0$,

$$\begin{aligned}
&\frac{1}{m_n^2}\sum_{i\neq j} M_i^{n,a_n}(t)M_j^{n,a_n}(t)\mathbb{1}_{[b,a_n]}\left(M_i^{n,a_n}(t)\right)\mathbb{1}_{[b,a_n]}\left(M_j^{n,a_n}(t)\right)\\
&= \left\langle \mu_t^{n,a_n}(dx)\mu_t^{n,a_n}(dy), xy\mathbb{1}_{[b,a_n]}(x)\mathbb{1}_{[b,a_n]}(y)\right\rangle - \frac{1}{m_n}\left\langle \mu_t^{n,a_n}(dx), x^2\mathbb{1}_{[b,a_n]}(x)\right\rangle\\
&\geq \left\langle \mu_t^{n,a_n}(dx), x\mathbb{1}_{[b,a_n]}(x)\right\rangle^2 - \frac{a_n}{m_n}\left\langle \mu_t^{n,a_n}(dx), x\right\rangle\\
&\geq \left\langle \mu_t^{n,a_n}(dx), x\mathbb{1}_{[b,a_n]}(x)\right\rangle^2 - \frac{a_n}{m_n}\,,
\end{aligned} \tag{4.9}$$

hence (4.8) after integrating over $(0,T)$, taking expectation, and using Lemma 4.3 (with $a = a_n$).
Step 2. By Lemma 4.2, we already know that $t \mapsto \langle \mu_t(dx), x\rangle$ and $t \mapsto \langle \mu_t(dx), 1\rangle$ are a.s. non-increasing functions. Consider now $\phi \in C_c([0,\infty))$. The convergence (4.7) and the assumption (I) ensure that $\left\langle \mu_t^{n_k,a_{n_k}}, \phi\right\rangle \longrightarrow \langle \mu_t, \phi\rangle$ a.s. for all $t \geq 0$ and $\langle \mu_0^{n_k}, \phi\rangle \longrightarrow \langle \mu_0, \phi\rangle$ as $k \to \infty$. Recalling (4.2), (4.4), and (4.5), we realize that $(\mu_t)_{t\geq 0}$ solves (2.5) provided that we check that $B_k(t) \longrightarrow B(t)$ (for instance in L^1) as $k \to \infty$ for all $t \geq 0$, where

$$\begin{aligned}
B_k(t) &:= \int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx)\mu_s^{n_k,a_{n_k}}(dy), K_{a_{n_k}}(x,y)\Delta\phi(x,y)\right\rangle\, ds,\\
B(t) &:= \int_0^t \langle \mu_s(dx)\mu_s(dy), K(x,y)\Delta\phi(x,y)\rangle\, ds.
\end{aligned}$$

For that purpose, we consider a family $(\mathcal{X}_b)_{b>0}$ of continuous non-increasing functions on $[0,\infty)$ such that $\mathcal{X}_b(x)=1$ for $x\in(0,b]$ and $\mathcal{X}_b(x)=0$ for $x\in[b+1,\infty)$, and put

$$\begin{aligned} B_k(t,b) &:= \int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx)\mu_s^{n_k,a_{n_k}}(dy), K_{a_{n_k}}(x,y)\Delta\phi(x,y)\mathcal{X}_b(x)\mathcal{X}_b(y)\right\rangle\ ds,\\ B(t,b) &:= \int_0^t \langle \mu_s(dx)\mu_s(dy), K(x,y)\Delta\phi(x,y)\mathcal{X}_b(x)\mathcal{X}_b(y)\rangle\ ds. \end{aligned}$$

On the one hand, it follows from (A_α), the boundedness of ϕ, the bounds $x^\alpha\le 1+x$ and

$$\sup_{t\ge 0}\langle \mu_t(dx),1+x\rangle = \langle \mu_0(dx),1+x\rangle\ ,$$

and the Lebesgue dominated convergence theorem that

$$\lim_{b\to\infty} E[|B(t,b)-B(t)|]=0. \tag{4.10}$$

On the other hand, for each $b\in(0,\infty)$, we have $K_{a_{n_k}}(x,y)\mathcal{X}_b(x)\mathcal{X}_b(y)=K(x,y)\mathcal{X}_b(x)\mathcal{X}_b(y)$ for all $(x,y)\in(0,\infty)^2$ as soon as $b+1\le a_{n_k}$, the latter being true for k sufficiently large. Consequently, since $(x,y)\longmapsto K(x,y)\mathcal{X}_b(x)\mathcal{X}_b(y)\Delta\phi(x,y)$ belongs to $C_c([0,\infty)^2)$, the convergence (4.7) entails that $B_k(t,b)\longrightarrow B(t,b)$ for all $t\ge 0$ a.s. as $k\to\infty$. Thanks to (A_α) and (4.3) we may apply the Lebesgue dominated convergence theorem to obtain that

$$\lim_{k\to\infty} E[|B_k(t,b)-B(t,b)|]=0 \quad \text{for each} \quad b>0\,. \tag{4.11}$$

Finally, owing to (2.3), we have for k sufficiently large (such that $a_{n_k}\ge b$)

$$\begin{aligned} & E[|B_k(t,b)-B_k(t)|]\\ &\le 3\|\phi\|_{L^\infty} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx)\mu_s^{n_k,a_{n_k}}(dy), K_{a_{n_k}}(x,y)\,(1-\mathcal{X}_b(x)\mathcal{X}_b(y))\right\rangle\ ds\right]\\ &\le 6C\|\phi\|_{L^\infty} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx)\mu_s^{n_k,a_{n_k}}(dy), x^\alpha y \mathbb{1}_{(0,a_{n_k}]}(x)\mathbb{1}_{(0,a_{n_k}]}(y)\,(1-\mathcal{X}_b(x)\mathcal{X}_b(y))\right\rangle\ ds\right]\\ &\le 6C\|\phi\|_{L^\infty} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx)\mu_s^{n_k,a_{n_k}}(dy), x^\alpha y \mathbb{1}_{(0,a_{n_k}]}(x)\mathbb{1}_{[b,a_{n_k}]}(y)\right\rangle\ ds\right]\\ &+6C\|\phi\|_{L^\infty} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx)\mu_s^{n_k,a_{n_k}}(dy), x^\alpha y \mathbb{1}_{[b,a_{n_k}]}(x)\mathbb{1}_{(0,a_{n_k}]}(y)\right\rangle\ ds\right]\\ &\le 6C\|\phi\|_{L^\infty} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx),(1+x)\mathbb{1}_{(0,a_{n_k}]}(x)\right\rangle \left\langle \mu_s^{n_k,a_{n_k}}(dy), y\mathbb{1}_{[b,a_{n_k}]}(y)\right\rangle\ ds\right]\\ &+\frac{6C\|\phi\|_{L^\infty}}{b^{1-\alpha}} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx), x\mathbb{1}_{[b,a_{n_k}]}(x)\right\rangle \left\langle \mu_s^{n_k,a_{n_k}}(dy), y\mathbb{1}_{(0,a_{n_k}]}(y)\right\rangle\ ds\right]. \end{aligned}$$

We then infer from (4.3), the Cauchy-Schwarz inequality, and (4.8) that, for $b \geq 1$,

$$\begin{aligned} E[|B_k(t,b) - B_k(t)|] &\leq 12\kappa C\|\phi\|_{L^\infty} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx), x1\!\!1_{[b,a_{n_k}]}(x)\right\rangle\, ds\right] \\ &\leq 12\kappa C\|\phi\|_{L^\infty} t^{1/2} E\left[\int_0^t \left\langle \mu_s^{n_k,a_{n_k}}(dx), x1\!\!1_{[b,a_{n_k}]}(x)\right\rangle^2\, ds\right]^{1/2} \\ &\leq 12\kappa C\|\phi\|_{L^\infty} t^{1/2}\left(\frac{L}{b^\alpha} + t\frac{a_{n_k}}{m_{n_k}}\right)^{1/2}. \end{aligned}$$

Since $a_n/m_n \to 0$ as $n \to \infty$, we may first let $k \to \infty$ and then $b \to \infty$ in the previous inequality to conclude that

$$\lim_{b\to\infty} \limsup_{k\to\infty} E[|B_k(t,b) - B_k(t)|] = 0. \tag{4.12}$$

Combining (4.10), (4.11), and (4.12) ends the proof. □

We next complete the proof of Point 1 of Theorem 2.5.

Proof of Point 1-(ii) of Theorem 2.5. Recall that we are in the situation where $a_n = m_n$, so that $(\mu_t^{n,a_n})_{t\geq 0} = (\mu_t^{n,m_n})_{t\geq 0}$ is the classical Marcus-Lushnikov process associated with (K,μ_0^n) for each $n \geq 1$. Let $(\mu_t)_{t\geq 0}$ be the limit of a subsequence $\left\{(\mu_t^{n_k,m_{n_k}})_{t\geq 0}\right\}_{k\geq 1}$ in the sense that a.s.

$$\lim_{k\to\infty} \sup_{[0,T]} d(\mu_t^{n_k,m_{n_k}}, \mu_t) = 0 \quad \text{for all} \quad T > 0\,, \tag{4.13}$$

the existence of such a limit following by the same arguments as (4.7).
We already know from [5] that $(\mu_t)_{t\geq 0}$ solves a.s. the Flory equation. We define the (*a priori* random) gelling time T_{gel} of $(\mu_t)_{t\geq 0}$ by (2.7) and write

$$\mu_t^{n,m_n} = \frac{1}{m_n}\sum_i \delta_{M_i^n(t)} \quad \text{with} \quad M_1^n(t) \geq M_2^n(t) \geq M_3^n(t) \geq ... \tag{4.14}$$

for all $t \geq 0$ and $n \geq 1$.

As before we denote by $(\mathcal{X}_b)_{b>0}$ a family of continuous non-increasing functions such that $\mathcal{X}_b(x) = 1$ for $x \in [0,b]$ and $\mathcal{X}_b(x) = 0$ for $x \geq b+1$. We start with the proof of (2.10) which is almost immediate. Since $t \mapsto M_1^n(t)/m_n$ is a.s. non-decreasing and bounded by 1, it suffices to check that $P[M_1^{n_k}(T_{gel} - \eta) \geq \delta m_{n_k}] \longrightarrow 0$ as $k \to \infty$ for all $\eta > 0$ and $\delta > 0$. For that purpose, fix $b > 0$. Since $1 - \mathcal{X}_b \geq 0$, it follows from (4.14) that

$$\begin{aligned} \left\langle \mu_t^{n_k,m_{n_k}}(dx), x(1-\mathcal{X}_b(x))\right\rangle &\geq \frac{M_1^{n_k,m_{n_k}}(t)\left(1 - \mathcal{X}_b\left(M_1^{n_k,m_{n_k}}(t)\right)\right)}{m_{n_k}}, \\ 1 - \left\langle \mu_t^{n_k,m_{n_k}}(dx), x\mathcal{X}_b(x)\right\rangle &\geq \frac{M_1^{n_k,m_{n_k}}(t)}{m_{n_k}} 1\!\!1_{[b+1,\infty)}\left(M_1^{n_k,m_{n_k}}(t)\right). \end{aligned}$$

For k large enough we have $\delta m_{n_k} \geq b+1$ and thus

$$\begin{aligned} E\left[1-\left\langle \mu_{T_{gel}-\eta}^{n_k,m_{n_k}}(dx), x\mathcal{X}_b(x)\right\rangle\right] &\geq E\left[\frac{M_1^{n_k,m_{n_k}}(T_{gel}-\eta)}{m_{n_k}} \mathbb{1}_{[\delta m_{n_k},\infty)}\left(M_1^{n_k,m_{n_k}}(T_{gel}-\eta)\right)\right] \\ &\geq \delta P\left[M_1^{n_k}(T_{gel}-\eta) \geq \delta m_{n_k}\right] . \end{aligned}$$

Now, thanks to the compactness of the support of $\mathcal{X}_b$ and (4.13), the sequence

$$\left(\left\langle \mu_{T_{gel}-\eta}^{n_k,m_{n_k}}(dx), x\mathcal{X}_b(x)\right\rangle\right)_{k\geq 1}$$

converges a.s. to $\langle \mu_{T_{gel}-\eta}(dx), x\mathcal{X}_b(x)\rangle$ and is bounded by (4.3). We may then let $k\to\infty$ in the above inequality to obtain

$$E\left[1-\langle \mu_{T_{gel}-\eta}(dx), x\mathcal{X}_b(x)\rangle\right] \geq \delta \limsup_{k\to\infty} P\left[M_1^{n_k}(T_{gel}-\eta)\geq \delta m_{n_k}\right] .$$

Next, owing to the definition of T_{gel}, the sequence $\left(\langle \mu_{T_{gel}-\eta}(dx), x\mathcal{X}_b(x)\rangle\right)_{b>0}$ converges towards $\langle \mu_{T_{gel}-\eta}(dx), x\rangle = 1$ as $b\to\infty$ and is bounded by 1. Passing to the limit as $b\to\infty$ in the previous inequality entails that $P\left[M_1^{n_k}(T_{gel}-\eta)\geq \delta m_{n_k}\right] \longrightarrow 0$ as $k\to\infty$, which is the claimed result. The limit (2.10) then follows.

We now turn to the proof of (2.11). Let $b>0$. Since $a_n = m_n \to \infty$ as $n\to\infty$, we have $a_{n_k} > b$ for k large enough and it follows from Lemma 4.3 (with $a=a_n$) by an argument similar to (4.9) that (recall that all the particles represented in $\mu_t^{n_k,m_{n_k}}$ are smaller than m_{n_k} by construction, see Definition 2.1)

$$\begin{aligned} & E\left[\int_0^{T_{gel}} \left\langle \mu_t^{n_k,m_{n_k}}(dx), x\mathbb{1}_{[b,\infty)}(x)\right\rangle^2 \, dt\right] \\ & \leq \frac{L}{b^\alpha} + E\left[\int_0^{T_{gel}} \frac{1}{m_{n_k}} \left\langle \mu_t^{n_k,m_{n_k}}(dx), x^2\mathbb{1}_{[b,\infty)}(x)\right\rangle \, dt\right] \\ & \leq \frac{L}{b^\alpha} + E\left[\int_0^{T_{gel}} \frac{M_1^{n_k}(t)}{m_{n_k}} \left\langle \mu_t^{n_k,m_{n_k}}(dx), x\right\rangle \, dt\right] \\ & \leq \frac{L}{b^\alpha} + E\left[\int_0^{T_{gel}} \frac{M_1^{n_k}(t)}{m_{n_k}} \, dt\right] . \end{aligned} \tag{4.15}$$

Since $M_1^{n_k}(t)\leq m_{n_k}$ and T_{gel} is a bounded random variable by Proposition 2.3, we easily deduce from (2.10) that $E\left[\int_0^{T_{gel}} (M_1^{n_k}(t)/m_{n_k}) \, dt\right] \longrightarrow 0$ as $k\to\infty$. Thus (2.11) follows from (4.15).

We next establish (2.8) and (2.9) and split the proof into five steps. In the first two steps we show that, for $t>T_{gel}$, at least one particle has a size of order δm_n for some $\delta>0$. Since such a particle is very attractive, we deduce in Step 3 that no other large particle can exist and obtain (2.9). We then conclude in the last two steps that, for $t>T_{gel}$, this single giant particle is solely responsible for the loss of mass and obtain (2.8).

Step 1. Let $(\alpha_n)_{n\geq 1}$ be any sequence of positive numbers such that $\alpha_n/m_n \to 0$ as $n\to\infty$. The aim of this step is to show that

$$\lim_{k\to\infty} P[M_1^{n_k}(T_{gel}+\varepsilon) > \alpha_{n_k}] = 1 \quad \text{for all} \quad \varepsilon > 0\,. \tag{4.16}$$

For that purpose, we introduce the stopping time

$$\tau_k := \inf\left\{t\geq 0 \ : \ M_1^{n_k}(t) > \alpha_{n_k}\right\}\,,$$

and notice that we may assume that $\alpha_n\to\infty$ as $n\to\infty$ without loss of generality. Owing to the time monotonicity of M_1^n, it suffices to prove that $P[\tau_k \geq T_{gel}+\varepsilon] \longrightarrow 0$ as $k\to\infty$ for all $\varepsilon>0$ to establish (4.16). Assume thus for contradiction that there is $\varepsilon>0$ such that $\delta := \limsup_k P[\tau_k \geq T_{gel}+\varepsilon] > 0$. Then, on the one hand, we have $P[\tilde\Omega]\geq\delta>0$ with $\tilde\Omega := \{\limsup_k \tau_k \geq T_{gel}+\varepsilon\}$. On the other hand, for each $k\geq 1$, it is clearly possible to couple the Marcus-Lushnikov processes $(\mu_t^{n_k,m_{n_k}})_{t\geq 0}$ and $(\mu_t^{n_k,\alpha_{n_k}})_{t\geq 0}$ in such a way that they coincide on $[0,\tau_k)$. Hence, a.s. on $\tilde\Omega$, up to extraction of a subsequence,

$$\lim_{k\to\infty}\sup_{[0,T_{gel}+\varepsilon/2]} d(\mu_t^{n_k,m_{n_k}},\mu_t) = \lim_{k\to\infty}\sup_{[0,T_{gel}+\varepsilon/2]} d(\mu_t^{n_k,\alpha_{n_k}},\mu_t) = 0.$$

By Theorem 2.5 Points 1-(i) and 2, we deduce that the limit $(\mu_t)_{t\geq 0}$ solves simultaneously the Flory and Smoluchowski equations on $[0,T_{gel}+\varepsilon/2)$ with positive probability, which contradicts Corollary 3.1.

Step 2. We now deduce from Step 1 that

$$\lim_{\delta\to 0}\liminf_{k} P\left[M_1^{n_k}(T_{gel}+\varepsilon) > \delta m_{n_k}\right] = 1 \quad \text{for all} \quad \varepsilon>0\,. \tag{4.17}$$

Assume for contradiction that there is $\varepsilon>0$ for which (4.17) fails to be true. Then there exists $\gamma\in[0,1)$ such that $\liminf_k P\left[M_1^{n_k}(T_{gel}+\varepsilon) > \delta m_{n_k}\right] < \gamma$ for all $\delta>0$. We may thus find a strictly increasing sequence $(k_l)_{l\geq 1}$ such that $P\left[M_1^{n_{k_l}}(T_{gel}+\varepsilon) > m_{n_{k_l}}/l\right] \leq \gamma$ for every $l\geq 1$. We then put $\alpha_{n_{k_l}} = m_{n_{k_l}}/l$ for $l\geq 1$ (and e.g. $\alpha_n = m_n^{1/2}$ if $n\notin\{n_{k_l} : l\geq 1\}$). Then $\alpha_n/m_n\to 0$ as $n\to\infty$ and the assertion (4.16) established in Step 1 warrants that $P\left[M_1^{n_k}(T_{gel}+\varepsilon) > \alpha_{n_k}\right] \longrightarrow 1$ as $k\to\infty$. But $P\left[M_1^{n_{k_l}}(T_{gel}+\varepsilon) > \alpha_{n_{k_l}}\right] \leq \gamma < 1$ for all $l\geq 1$, hence a contradiction.

Step 3. We are now in a position to prove (2.9) which somehow means that the other particles are small in the sense that

$$\lim_{b\to\infty}\limsup_{k\to\infty} P\left[\left(\int_{T_{gel}}^{\infty} X^{n_k}(s,b)\,ds\right) \geq \beta\right] = 0 \tag{4.18}$$

for all $\beta>0$ with the notation

$$X^n(s,b) := \frac{1}{m_n}\sum_{i\geq 2} M_i^n(s)\mathbb{1}_{[b,\infty)}\left(M_i^n(s)\right)\,.$$

First note that a.s., for all $s\geq 0$ and $n\geq 1$, we have

$$\frac{1}{m_n^2}\sum_{i\neq j} M_i^n(s)M_j^n(s)\mathbb{1}_{[b,m_n]}\left(M_i^n(s)\right)\mathbb{1}_{[b,m_n]}\left(M_j^n(s)\right) \geq \frac{M_1^n(s)}{m_n}\mathbb{1}_{[b,\infty)}\left(M_1^n(s)\right)\ X^n(s,b)\,,$$

since $M_i^n(s) \le m_n$ for all $s \ge 0$ and $i \ge 1$. By Lemma 4.3 (with $a = m_n$), we obtain

$$E\left[\int_{T_{gel}}^{\infty} \frac{M_1^n(s)}{m_n} 1\!\!1_{[b,\infty)}\left(M_1^n(s)\right)\ X^n(s,b)\ ds\right] \le \frac{L}{b^{\alpha}}. \tag{4.19}$$

We next fix $\beta > 0$, $\eta > 0$, and $b > 0$. By (4.17) there is $\delta > 0$ such that $\liminf_k P[M_1^{n_k}(T_{gel}+\beta/2) \ge \delta m_{n_k}] \ge 1-\eta$. Recalling that $t \mapsto M_1^n(t)$ is a.s. non-decreasing and $X^n(s,b) \le 1$ for all $s \ge 0$ a.s., we have for k sufficiently large such that $\delta m_{n_k} > b$,

$$\begin{aligned}
& P\left[\left(\int_{T_{gel}}^{\infty} X^{n_k}(s,b)\ ds\right) \ge \beta\right] \le P\left[\left(\int_{T_{gel}+\beta/2}^{\infty} X^{n_k}(s,b)\ ds\right) \ge \beta/2\right] \\
& \le P\left[M_1^{n_k}(T_{gel}+\beta/2) \le \delta m_{n_k}\right] \\
& + P\left[\left(\int_{T_{gel}+\beta/2}^{\infty} \frac{M_1^{n_k}(T_{gel}+\beta/2)}{\delta m_{n_k}} 1\!\!1_{[b,\infty)}\left(M_1^{n_k}(T_{gel}+\beta/2)\right) X^{n_k}(s,b)\ ds\right) \ge \beta/2\right] \\
& \le 1 - P\left[M_1^{n_k}(T_{gel}+\beta/2) > \delta m_{n_k}\right] \\
& + P\left[\left(\int_{T_{gel}+\beta/2}^{\infty} \frac{M_1^{n_k}(s)}{m_{n_k}} 1\!\!1_{[b,\infty)}\left(M_1^{n_k}(s)\right) X^{n_k}(s,b)\ ds\right) \ge \beta\delta/2\right] \\
& \le 1 - P\left[M_1^{n_k}(T_{gel}+\beta/2) > \delta m_{n_k}\right] + \frac{2L}{b^{\alpha}\beta\delta}\,,
\end{aligned}$$

the last inequality being a consequence of (4.19). Letting $k \to \infty$ in the above inequality, we obtain, thanks to the choice of δ,

$$\limsup_{k\to\infty} P\left[\left(\int_{T_{gel}}^{\infty} X^{n_k}(s,b)\ ds\right) \ge \beta\right] \le \eta + \frac{2L}{b^{\alpha}\beta\delta}\,.$$

Now, we first pass to the limit as $b \to \infty$ and then as $\eta \to 0$ in the above inequality to obtain (4.18), i.e. (2.9).

Step 4. Set $\gamma_t := 1 - \left\langle \mu_{T_{gel}+t}(dx), x\right\rangle$ and $B_k(t) := M_1^{n_k}(T_{gel}+t)/m_{n_k}$ for $t \ge 0$ and $k \ge 1$. Our aim in this step is to prove that

$$\lim_{k\to\infty} E\left[\int_0^T |B_k(t) - \gamma_t|\ dt\right] = 0 \quad \text{for all} \quad T > 0\,. \tag{4.20}$$

As before let $(\mathcal{X}_b)_{b>0}$ be a family of continuous non-increasing functions such that $\mathcal{X}_b(x) = 1$ for $x \in [0,b]$ and $\mathcal{X}_b(x) = 0$ for $x \ge b+1$. We then put

$$\gamma_t^b := 1 - \left\langle \mu_{T_{gel}+t}(dx), x\mathcal{X}_b(x)\right\rangle \quad \text{and} \quad \gamma_t^{b,k} := 1 - \left\langle \mu_{T_{gel}+t}^{n_k,m_{n_k}}(dx), x\mathcal{X}_b(x)\right\rangle$$

for $b > 0$, $k \ge 1$, and $t \ge 0$. On the one hand, we have a.s. that $\gamma_t^b \longrightarrow \gamma_t$ as $b \to \infty$ for all $t \ge 0$. Since $|\gamma_t| + |\gamma_t^b| \le 2$, we deduce from the Lebesgue dominated convergence theorem that

$$\lim_{b\to\infty} E\left[\int_0^T |\gamma_t - \gamma_t^b|\ dt\right] = 0\,. \tag{4.21}$$

On the other hand, owing to the compactness of the support of $\mathcal{X}_b$, we infer from (4.13) that $\gamma_t^{b,k} \longrightarrow \gamma_t^b$ a.s. for all $b > 0$ and $t \geq 0$. As $|\gamma_t^b| + |\gamma_t^{b,k}| \leq 2$, we use again the Lebesgue dominated convergence theorem to obtain that

$$\lim_{k\to\infty} E\left[\int_0^T |\gamma_t^b - \gamma_t^{b,k}|\ dt\right] = 0 \quad \text{for each} \quad b > 0\,. \tag{4.22}$$

But $\gamma_t^{b,k} = A_{b,k}(t) + B_k(t) - C_{b,k}(t)$ a.s., where

$$\begin{aligned} A_{b,k}(t) &:= \frac{1}{m_{n_k}} \sum_{i\geq 2} M_i^{n_k,m_{n_k}}(T_{gel}+t)\left(1 - \mathcal{X}_b(M_i^{n_k,m_{n_k}}(T_{gel}+t))\right), \\ C_{b,k}(t) &:= \frac{M_1^{n_k,m_{n_k}}(T_{gel}+t)}{m_{n_k}} \mathcal{X}_b(M_1^{n_k,m_{n_k}}(T_{gel}+t)) \leq \frac{b+1}{m_{n_k}}. \end{aligned}$$

Clearly,

$$\lim_{k\to\infty} E\left[\int_0^T C_{b,k}(t)\ dt\right] = 0 \quad \text{for each} \quad b > 0\,, \tag{4.23}$$

while, since $0 \leq A_{b,k}(t) \leq X^{n_k}(T_{gel}+t, b) \leq 1$ a.s.,

$$\lim_{b\to\infty} \limsup_{k\to\infty} E\left[\int_0^T A_{b,k}(t)\ dt\right] = 0 \tag{4.24}$$

by (4.18) and the Lebesgue dominated convergence theorem.
Now, since $B_k(t) - \gamma_t = C_{b,k}(t) - A_{b,k}(t) + (\gamma_t^{b,k} - \gamma_t^b) + (\gamma_t^b - \gamma_t)$ for $b > 0$, $k \geq 1$, and $t \geq 0$, it follows from (4.22) and (4.23) that

$$\limsup_{k\to\infty} E\left[\int_0^T |B_k(t) - \gamma_t|\ dt\right] \leq \limsup_{k\to\infty} E\left[\int_0^T A_{b,k}(t)\ dt\right] + E\left[\int_0^T |\gamma_t - \gamma_t^b|\ dt\right]$$

for all $b > 0$. Letting $b \to \infty$ and using (4.21) and (4.24) give (4.20).
Step 5. To complete the proof of (2.8), it remains to show that, for all $\varepsilon > 0$ and $\eta > 0$, we have

$$\lim_{k\to\infty} P\left[\sup_{t\in[\eta,\infty)} |B_k(t) - \gamma_t| \geq \varepsilon\right] = 0. \tag{4.25}$$

Indeed, (4.25) clearly implies (2.8) by the Lebesgue dominated convergence theorem since $B_k(t) \leq 1$ and $\gamma_t \leq 1$ for $t \geq 0$ a.s.
We thus fix $\varepsilon > 0$ and $\eta > 0$. Since $(\mu_t)_{t\geq 0}$ solves the Flory equation (F) by Theorem 2.5 Point 1-(i), it follows from Proposition 2.3 that $t \longmapsto \gamma_t$ is a.s. increasing and continuous on $[\eta, \infty)$ and $\gamma_t \longrightarrow 1$ as $t \to \infty$ a.s. It is also straightforward to check that $t \mapsto B_k(t)$ is a.s. non-decreasing on $[\eta, \infty)$ with $B_k(t) \longrightarrow 1$ as $t \to \infty$ a.s. As a consequence of the a.s. monotonicity and boundedness of $t \longmapsto \gamma_t$ and $B_k(t)$, we have for $t \geq T$

$$\begin{aligned} |B_k(t) - \gamma_t| &= \max\{B_k(t) - \gamma_T + \gamma_T - \gamma_t, \gamma_t - \gamma_T + \gamma_T - B_k(T) + B_k(T) - B_k(t)\} \\ &\leq \max\{1 - \gamma_T, 1 - \gamma_T + \gamma_T - B_k(T)\} \\ &\leq 1 - \gamma_T + \max\{0, \gamma_T - B_k(T)\}\,, \end{aligned}$$

hence

$$\sup_{t\in[T,\infty)} |B_k(t)-\gamma_t| \le 1-\gamma_T + |B_k(T)-\gamma_T| \quad \text{for all} \quad T>0\,. \tag{4.26}$$

To go further we will use the following result which resembles Dini's theorem.

Lemma 4.4 *Let $T>0$ and $f\in C([0,T])$ be a non-decreasing function. If $(f_k)_{k\ge 1}$ is a sequence of non-decreasing functions on $(0,T)$ such that $f_k \longrightarrow f$ in $L^1(0,T)$ as $k\to\infty$, then $f_k\longrightarrow f$ in $L^\infty(\delta,T-\delta)$ as $k\to\infty$ for every $\delta\in(0,T/2)$.*

Let $T>\eta$. By (4.20) and Proposition 2.3, $(B_k)_{k\ge1}$ is a sequence of non-decreasing functions that converges to the continuous and non-decreasing function $t\mapsto\gamma_t$ in $L^1(0,T+\eta)$ a.s. and we use Lemma 4.4 to conclude that

$$\lim_{k\to\infty} P\left[\sup_{t\in[\eta,T]} |B_k(t)-\gamma_t| \ge \varepsilon/2\right] = 0. \tag{4.27}$$

We now infer from (4.26) and (4.27) that

$$\begin{aligned}
P\left[\sup_{t\in[\eta,\infty)} |B_k(t)-\gamma_t| \ge \varepsilon\right] &\le P\left[\sup_{t\in[\eta,T]} |B_k(t)-\gamma_t| + |1-\gamma_T| \ge \varepsilon\right]\\
&\le P\left[1-\gamma_T\ge\varepsilon/2\right] + P\left[\sup_{t\in[\eta,T]} |B_k(t)-\gamma_t| \ge \varepsilon/2\right],\\
\limsup_{k\to\infty} P\left[\sup_{t\in[\eta,\infty)} |B_k(t)-\gamma_t| \ge \varepsilon\right] &\le P\left[1-\gamma_T\ge\varepsilon/2\right].
\end{aligned}$$

The above inequality being valid for any $T>\eta$, we may let $T\to\infty$ to deduce (4.25) since $\gamma_T\longrightarrow 1$ as $T\to\infty$ a.s. □

We finally turn to the proof of the last statement of Theorem 2.5.

Proof of Point 3 of Theorem 2.5. Here $(a_n)_{n\ge1}$ is a sequence of positive real numbers such that $a_n\to\infty$ and $a_n/m_n\to\gamma\in(0,1)$ as $n\to\infty$. We consider the limit $(\mu_t)_{t\ge0}$ of a subsequence $\left\{(\mu_t^{n_k,a_{n_k}})_{t\ge0}\right\}_{k\ge1}$ in the sense that a.s.

$$\lim_{k\to\infty} \sup_{[0,T]} d(\mu_t^{n_k,a_{n_k}},\mu_t) = 0 \quad \text{for all} \quad T\ge 0\,, \tag{4.28}$$

the existence of such a limit following by the same arguments as (4.7).
We then introduce

$$T_1 := \inf\left\{t\ge0\ :\ \langle \mu_0(dx)-\mu_t(dx), x\rangle \ge \gamma\right\},$$

and aim at showing that $(\mu_t)_{t\in[0,T_1)}$ solves a.s. the Flory equation (F).
For $n\ge1$, we set

$$T_1^n := \inf\{t\ge0\ ;\ \left\langle \mu_t^{n,a_n}, \mathbb{1}_{(a_n,\infty)}\right\rangle > 0\}\,,$$

which represents the first time that a particle of size exceeding a_n appears in the Marcus-Lushnikov process $(\mu_t^{n,a_n})_{t\ge0}$. For each $n\ge1$, it is clearly possible to build a classical Marcus-Lushnikov process $(\mu_t^{n,m_n})_{t\geq 0}$ (i.e. without cut-off) such that $\mu_t^{n,m_n} = \mu_t^{n,a_n}$ for $t \in [0, T_1^n]$ a.s. In particular we have also $T_1^n = \inf\{t \geq 0 \ : \ \langle \mu_t^{n,m_n}(dx), \mathbb{1}_{(a_n,\infty)}\rangle > 0\}$ a.s. Denoting by $M_1^n(t)$ the size of the largest particle at time t in the process $(\mu_t^{n,m_n})_{t\geq 0}$, we clearly have $T_1^n = \inf\{t \geq 0 \ : \ M_1^n(t) > a_n\}$ a.s.

By the tightness result of Proposition 2.4, we may assume that, up to extracting a further subsequence (not relabeled), $(\mu_t^{n_k,m_{n_k}})_{t\geq 0}$ converges to $(\nu_t)_{t\geq 0}$ in $\mathbb{D}([0,\infty), \mathcal{M}_f^+)$. By the Skorokhod representation theorem and Lemma 4.2, we may assume that a.s.

$$\lim_{k\to\infty} \sup_{[0,T]} d(\mu_t^{n_k,m_{n_k}}, \nu_t) = 0 \quad \text{for all} \quad T > 0\,. \tag{4.29}$$

By Theorem 2.5 Point 1-(i), $(\nu_t)_{t\geq 0}$ solves a.s. the Flory equation (F). Introducing

$$S_1 := \inf\left\{t \geq 0 \ : \ \langle \mu_0(dx) - \nu_t(dx), x\rangle \geq \gamma\right\}\,,$$

we claim that

$$\lim_{k\to\infty} P[|S_1 - T_1^{n_k}| > \varepsilon] = 0 \quad \text{for all} \quad \varepsilon > 0\,. \tag{4.30}$$

Taking (4.30) for granted, we deduce that $\mu_t = \nu_t$ for $t \in [0, S_1)$ a.s. since $\mu_t^{n,m_n} = \mu_t^{n,a_n}$ for $t \in [0, T_1^n]$ a.s. for all $n \geq 1$. This implies that $S_1 = T_1$ a.s., because the subset $\{\pi \in \mathcal{M}_f^+;\ \langle \mu_0(dx) - \pi(dx), x\rangle \geq \gamma\} = \{\pi \in \mathcal{M}_f^+;\ \langle \pi(dx), x\rangle \leq 1-\gamma\}$ is closed in $\mathcal{M}_f^+$ endowed with the vague topology, and both $t \mapsto \mu_t$ and $t \mapsto \nu_t$ are a.s. continuous for that topology by Lemma 4.2. Therefore, $(\mu_t)_{t\in[0,T_1)}$ solves a.s. the Flory equation.

We are left with the proof of (4.30). To this end we will use (2.8) and (2.10) (with the weak limit $(\nu_t)_{t\geq 0}$ of the classical Marcus-Lushnikov process $\left(\mu_t^{n_k,m_{n_k}}\right)_{k\geq 1}$). Introducing the (random) gelling time S_{gel} of $(\nu_t)_{t\geq 0}$ given by

$$S_{gel} := \inf\{t \geq 0 \ : \ \langle \nu_t(dx), x\rangle < \langle \mu_0(dx), x\rangle\}\,,$$

we recall that a.s. the map $t \longmapsto \langle \nu_t(dx), x\rangle$ is constant and equal to 1 on $[0, S_{gel})$ and continuous and decreasing on (S_{gel}, ∞) by Proposition 2.3. In the proof of (4.30), we have to handle separately the events $S_{gel} < S_1$ and $S_{gel} = S_1$, the latter being not ruled out *a priori* due to the possible discontinuity of $t \longmapsto \langle \nu_t(dx), x\rangle$ at $t = S_{gel}$.

Fix $\varepsilon > 0$ and write

$$P[|S_1 - T_1^{n_k}| > \varepsilon] = P[U_k] + P[V_k] + P[W_k]$$

with

$$\begin{aligned} U_k &:= \left\{S_{gel} \leq S_1 \leq S_{gel} + \varepsilon/2\,,\ T_1^{n_k} < S_1 - \varepsilon\right\}\,, \\ V_k &:= \left\{S_{gel} + \varepsilon/2 < S_1\,,\ T_1^{n_k} < S_1 - \varepsilon\right\}\,, \\ W_k &:= \left\{S_{gel} \leq S_1\,,\ T_1^{n_k} > S_1 + \varepsilon\right\}\,. \end{aligned}$$

First, on U_k, we have $a_{n_k} \leq M_1^{n_k}(S_1 - \varepsilon) \leq M_1^{n_k}(S_{gel} - \varepsilon/2)$, so that

$$P[U_k] \leq P\left[M_1^{n_k}(S_{gel} - \varepsilon/2) \geq a_{n_k}\right] \leq P\left[\frac{M_1^{n_k}(S_{gel} - \varepsilon/2)}{m_{n_k}} \geq \frac{a_{n_k}}{m_{n_k}}\right] \underset{k\to\infty}{\longrightarrow} 0$$

by (2.10) since $a_n/m_n \to \gamma > 0$ as $n \to \infty$.

Next, introducing $\tau := S_1 - \varepsilon/4$ and $Z := \langle \nu_\tau(dx), x\rangle - 1 + \gamma$, it follows from the a.s. strict monotonicity of $t \longmapsto \langle \nu_t(dx), x\rangle$ on (S_{gel}, ∞) and the definitions of T_1^n and S_1 that

$$Z > 0 \quad \text{a.s. and} \quad V_k \subset \left\{ \frac{M_1^{n_k}(\tau)}{m_{n_k}} - (1 - \langle \nu_\tau(dx), x\rangle) \geq \frac{a_{n_k}}{m_{n_k}} - \gamma + Z \right\} .$$

Let $\eta > 0$. For k large enough we have $|\gamma - a_{n_k}/m_{n_k}| \leq \eta/2$ and since $\tau > S_{gel} + \varepsilon/4$ a.s.

$$\begin{aligned}
& E\left[\sup_{t \in [S_{gel}+\varepsilon/4, \infty)} \left\{ \frac{M_1^{n_k}(t)}{m_{n_k}} - (1 - \langle \nu_t(dx), x\rangle) \right\} \right] \\
\geq \; & E\left[\mathbb{1}_{V_k} \left(\frac{M_1^{n_k}(\tau)}{m_{n_k}} - (1 - \langle \nu_\tau(dx), x\rangle) \right) \right] \geq E\left[\mathbb{1}_{V_k} \left(Z + \frac{a_{n_k}}{m_{n_k}} - \gamma \right) \right] \geq E\left[\mathbb{1}_{V_k} (Z - \eta/2) \right] \\
\geq \; & \frac{\eta}{2} \, E\left[\mathbb{1}_{V_k} \, \mathbb{1}_{[\eta,\infty)}(Z) \right] - \frac{\eta}{2} \, E\left[\mathbb{1}_{(0,\eta)}(Z) \right] \geq \frac{\eta}{2} \, P[V_k] - \eta P[Z \in (0,\eta)] .
\end{aligned}$$

Multiplying the above inequality by $2/\eta$ and letting $k \to \infty$ with the help of (2.8) give

$$\limsup_{k \to \infty} P[V_k] \leq 2P[Z \in (0,\eta)] \quad \text{for all } \eta > 0 .$$

As $Z > 0$ a.s., the right hand side of the above inequality converges to zero as $\eta \to 0$. Consequently, $P[V_k] \longrightarrow 0$ as $k \to \infty$.
Similarly, introducing $\sigma := S_1 + \varepsilon$ and $Y := 1 - \langle \nu_\sigma(dx), x\rangle - \gamma$, the a.s. strict monotonicity of $t \longmapsto \langle \nu_t(dx), x\rangle$ on (S_{gel}, ∞) and the definitions of T_1^n and S_1 warrant that

$$Y > 0 \quad \text{a.s. and} \quad W_k \subset \left\{ (1 - \langle \nu_\sigma(dx), x\rangle) - \frac{M_1^{n_k}(\sigma)}{m_{n_k}} \geq \gamma - \frac{a_{n_k}}{m_{n_k}} + Y \right\} .$$

Arguing as for V_k, we have for all $\eta > 0$ and k large enough

$$E\left[\sup_{t \in [S_{gel}+\varepsilon, \infty)} \left\{ \frac{M_1^{n_k}(t)}{m_{n_k}} - (1 - \langle \nu_t(dx), x\rangle) \right\} \right] \geq \frac{\eta}{2} \, P[W_k] - \eta P[Y \in (0,\eta)] .$$

We then proceed as before to deduce from (2.8) and the a.s. positivity of Y that $P[W_k] \longrightarrow 0$ as $k \to \infty$ and thus complete the proof of (4.30). □

5 Numerical illustrations

We consider the *monodisperse* initial condition $\mu_0 = \delta_1$ and the *multiplicative kernel* $K(x,y) = xy$. Under these conditions, there is an explicit solution to the Smoluchowski equation (S) given by

$$\hat{\mu}_t(dx) := \sum_{k \geq 1} \hat{c}(t,k) \delta_k(dx) \quad \text{with} \quad \hat{c}(t,k) := \begin{cases} \dfrac{k^{k-2}}{k!} t^{k-1} e^{-kt} & \text{for} \quad t \in [0,1] , \\[2ex] \dfrac{k^{k-2}}{k!} t^{-1} e^{-k} & \text{for} \quad t \geq 1 . \end{cases}$$

For the same initial condition, the Flory equation (F) has also an explicit solution given by

$$\mu_t(dx) := \sum_{k\geq 1} c(t,k)\delta_k(dx) \quad \text{with} \quad c(t,k) := \frac{k^{k-2}}{k!} t^{k-1} e^{-kt} \quad \text{for} \quad t \geq 0\,.$$

Before proceeding to simulations, let us point out that $\langle \mu_t(dx), x\rangle = 1$ for $t \in [0,1]$, while $\langle \mu_t(dx), x\rangle = t^*/t$ for $t > 1$, where $t^* \in (0,1)$ is the unique solution to $t^* e^{-t^*} = te^{-t}$ in $(0,1)$. Easy computations show that

$$T_1(\gamma) := \inf\{t \geq 0;\ \langle \mu_0(dx) - \mu_t(dx), x\rangle \geq \gamma\} = -\frac{\ln(1-\gamma)}{\gamma} \quad \text{for} \quad \gamma \in (0,1)\,.$$

In Figures 1 to 5, the plain, dashed, and dotted lines represent $\mu_t^{n,a_n}(\{2\})$, $c(t,2)$, and $\hat{c}(t,2)$, respectively, as functions of t. We observe that, as explained by Theorem 2.5,

(i) for $a_n \ll m_n$, μ_t^{n,a_n} approximates the solution to the Smoluchowski equation, see Figure 1,

(ii) for $a_n = m_n$, μ_t^{n,a_n} approximates the solution to the Flory equation, see Figure 2,

(iii) for $a_n = \gamma m_n$ with $\gamma \in (0,1)$, μ_t^{n,a_n} approximates the solution to the Flory equation until the time $T_1(\gamma)$, and then changes its behaviour: see Figure 3 ($\gamma = 0.5$, $T_1(0.5) = 1.386$), Figure 4 ($\gamma = 0.8$, $T_1(0.8) = 2.012$) and Figure 5 ($\gamma = 0.33$, $T_1(0.33) = 1.21$). Note that Figure 5 shows that the behaviour of μ_t^{n,a_n} *bifurcates* at least twice on $t \in [0,3]$. The second bifurcation certainly corresponds to the time where a second *giant* particle with size 10^5 appears.

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Figure 1: $n = m_n = 10^4$, $a_n = 10^2$

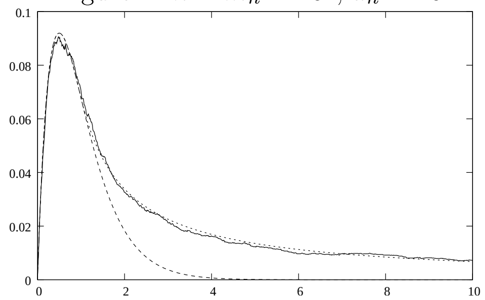

Figure 2: $n = m_n = 10^4$, $a_n = 10^4$

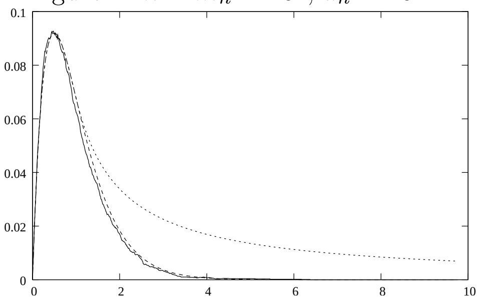

Figure 3: $n = m_n = 10^4$, $a_n = 5.10^3$

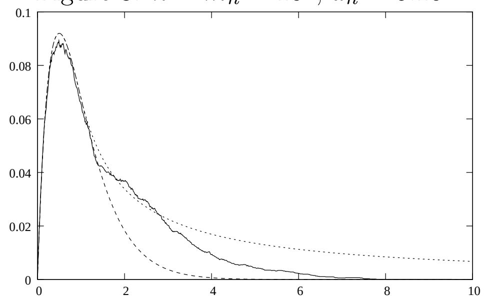

Figure 4: $n = m_n = 10^4$, $a_n = 8.10^3$

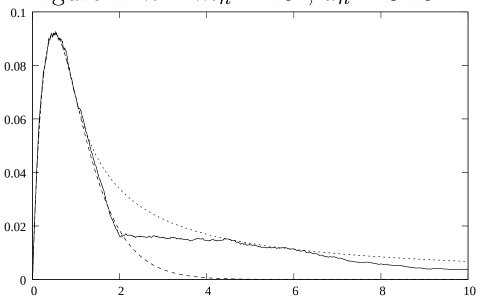

Figure 5: $n = m_n = 3.10^5$, $a_n = 10^5$.