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Approximations to the gelation phase of an aerosol

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Abstract

We investigate a time discretized version of the Smoluchowski coagulation equation. By means of a numerical example we prove its suitability as a basis for the efficient simulation of the transition to gelation.

Key words: Smoluchowski equation, coagulation, gelation, numerical simulation.

MSC classification: 37M05, 82C22

1 Introduction

1.1 A Smoluchowski equation

In the following we denote $\mathbb{N} = \{1, 2, \ldots\}$ and define $\mathcal{L}^1_+$ as the set of sequences $\mathbf{h} = (h_n)_{n\in\mathbb{N}}$ satisfying

$$h_n \geq 0, \quad \|\mathbf{h}\|_1 = \sum_{n=1}^{\infty} h_n < \infty. \tag{1.1}$$

We consider the following Smoluchowski system of equations for $\mathcal{L}^1_+$-valued functions $\tilde{\mathbf{f}} = (\tilde{f}_n)_{n=1}^{\infty}$,

$$\partial_t \tilde{f}_n = \frac{1}{2} \sum_{i=1}^{n-1} i(n-i)\tilde{f}_i \tilde{f}_{n-i} - n\tilde{f}_n \cdot \sum_{i=1}^{\infty} i\tilde{f}_i \tag{1.2}$$

resp. the equivalent version [2] for $\mathbf{f} = (f_n)_{n=1}^{\infty} = (n\tilde{f}_n)_{n=1}^{\infty}$,

$$\partial_t f_n \;=\; \sum_{i=1}^{n-1} i f_i f_{n-i} - n f_n \cdot \rho \tag{1.3}$$

where (for small times t) $\rho(t) = \rho^{\infty}$, defined by

$$\rho^{\infty}[\mathbf{f}(t)] = \sum_{i=1}^{\infty} f_n(t), \tag{1.4}$$

is the total mass in the state space $\mathbb{N}$ at time t. System (1.3) has been frequently investigated in literature (see the review papers [1, 10]). Define the first moment of $\mathbf{f}$ (i.e. the second moment of $\tilde{\mathbf{f}}$)

$$M(t) = M[\mathbf{f}(t)] := \sum_{i=1}^{\infty} n f_n(t) \tag{1.5}$$

and assume $M(0) < \infty$. Then a formal calculation yields the equation for M

$$M' = M^2 \quad , \tag{1.6}$$

(for a rigorous derivation, see Proposition 2.3(c)). Its solution

$$M(t) = \frac{M(0)}{1 - M(0) \cdot t} \tag{1.7}$$

remains finite only for $t < 1/M(0) =: t_{gel}$. t_{gel} is called the *gelation time*.

Define the partial sums

$$\rho^N(t) = \rho^N[\mathbf{f}(t)] := \sum_{i=1}^{N} f_n(t). \tag{1.8}$$

Then ρ^N satisfies

$$\partial_t \rho^N = \sum_{n=1}^{N} n f_n \left(\rho^{N-n} - \rho\right) \leq 0. \tag{1.9}$$

Thus ρ^N and with this also ρ^{∞} are monotonically decreasing. As one can show, $\rho^{\infty}(t)$ remains constant until t_{gel}; after this time, the mass starts to decrease strictly. We can obtain formal mass conservation in our system by adding a new state variable "∞" to the state space $\mathbb{N}$ (writing $\overline{\mathbb{N}} = \mathbb{N} \cup \{\infty\}$) and putting

$$f_{\infty}(t) := \rho^{\infty}(0) - \rho^{\infty}(t). \tag{1.10}$$

We define

$$\overline{\rho} := \rho^{\infty}(t) + f_{\infty}(t) = \rho(0). \tag{1.11}$$

For $t > t_{gel}$, there are two possibilities of choosing ρ in (1.3) depending on the model under consideration. Considering an *active gel*, the mass contained in f_{∞} still contributes to the interaction, while it is removed in the case of a *passive* gel. This means that we have to define

$$\rho(t) = \begin{cases} \overline{\rho} & \text{for an active gel} \\ \rho^{\infty}(t) & \text{for a passive gel} \end{cases} \quad . \tag{1.12}$$

Independent of this choice there is a simple way to formally construct solutions as functions of ρ. Given $\mathbf{f}(t_0)$, define $\mathbf{g}(t) = (g_n(t))_{n\in\mathbb{N}}$ for $t \geq 0$ by

$$g_n(t) = f_n(t_0 + t) \cdot \exp\left(n \int_{t_0}^{t_0+t} \rho(\tau) d\tau\right) \quad . \tag{1.13}$$

Then $\mathbf{g}$ is uniquely given by the recursive system

$$\mathbf{g}(0) = \mathbf{f}(t_0), \tag{1.14}$$

$$g_1(t) = g_1(0) = \text{const}, \quad \text{and for} \quad n > 1, \tag{1.15}$$

$$\partial_t g_n = \sum_{i=1}^{n-1} i g_i g_{n-i} \tag{1.16}$$

which is bounded by the solution $\mathbf{a} = (a_n)$ of

$$\partial_t a_n = \sum_{i=1}^{n-1} i a_i a_{n-i}, \quad a_n(0) = 1. \tag{1.17}$$

We easily check that

$$a_n(t) = 1 + t \cdot \phi_n(t) \tag{1.18}$$

with a continuous monotonically increasing function ϕ_n defined on $\mathbb{R}_+ = [0, \infty)$, and $\phi_n(0) = n(n-1)/2$. In the case of an active gel, $\rho(t) = \overline{\rho}$ is a given constant. Solutions $\mathbf{f}^p$ of a passive and $\mathbf{f}^a$ of an active gel with given initial condition $\mathbf{f}(0)$ can be transformed into one another using the formula

$$f_n^p(t) \cdot \exp\left(n \int_0^t \rho^{\infty}(\tau) d\tau\right) = f_n^a(t) \cdot \exp(n\overline{\rho}t) \quad . \tag{1.19}$$

From now on we restrict to the case of an *active* gel. Its solution is uniquely given from (1.13)... (1.16) and satisfies the inequalities

$$f_n(t_0) \cdot \exp(-\overline{\rho} n t) \leq f_n(t_0 + t) \leq [f_n(t_0) + \phi_n(t) \cdot t] \cdot \exp(-\overline{\rho} n t) \tag{1.20}$$

with ϕ_n defined by (1.17), (1.18).

1.2 Objectives and outline of the paper

The Smoluchowski system introduced above is at present of high scientific interest in combination with diffusion in physical space. The corresponding system for $\mathbf{f}(t, x)$ ($x \in \mathbb{R}^d$ the space coordinate) reads

$$\partial_t f_n = D_n \Delta_x f_n + \sum_{i=1}^{n-1} i f_i f_{n-i} - n \rho f_n \quad . \tag{1.21}$$

One particular point of interest is the mutual dependence of diffusion and gelation. For the numerical solution of (1.21) it is important to dispose of an algorithm which is numerically efficient and capable of resolving the phase transition to gelation. In [2, 5, 6], stochastic particle schemes for the Smoluchowski system have been proposed which have been applied to diffusion problems e.g. in [3, 4, 7]. All these schemes are based on the version (1.3) proposed in [2] rather than on the original version (1.2) which turns out to be less efficient for numerical purposes. While the schemes in [5, 6, 7] use exponentially distributed random times, those of [2, 3, 4] are based on a fixed time discretization. It is the objective of the present paper to use this latter approach for the derivation of a completely deterministic approximation scheme for system (1.3).

The plan of the paper is as follows. In section 2 we introduce an order relation on $\mathcal{L}^1_+$ which is crucial since it controls the whole approximation process. Monotonicity properties for solutions of the Smoluchowski equation are proven in terms of a Markov jump process. In section 3 we piecewise linearize the Smoluchowski equation, establish an approximation scheme and prove its convergence. Section 4 is devoted to a numerical scheme capable of passing through the gelation time. As the most promising variant turns out a version which locally in time is supplemented with a small stochastic system simulating the first transition to gel.

2 Preliminaries

2.1 An order relation on $\mathcal{L}^1_+$

We make use of the sequence spaces $\mathcal{L}^1_+$ as defined above and of

$$\mathcal{L}^\infty_+ \quad = \quad \{\mathbf{h} = (h_n)_{n\in\mathbb{N}} \quad | \quad h_n \geq 0, \quad \|\mathbf{h}\|_\infty = \sup_{n\in\mathbb{N}} |h_n| < \infty\}. \tag{2.1}$$

For $\gamma = (\gamma_n) \in \mathcal{L}^1_+$ and $N \in \overline{\mathbb{N}} = \mathbb{N} \cup \{\infty\}$ we define the partial sums

$$\rho^N[\gamma] := \sum_{n=1}^{N} \gamma_n \quad \text{for} \quad N \in \overline{\mathbb{N}} = \mathbb{N} \cup \{\infty\}, \tag{2.2}$$

and the first moment

$$M[\gamma] := \sum_{n=1}^{\infty} n\gamma_n \in \overline{\mathbb{R}} = [0, \infty] \quad . \tag{2.3}$$

On $\mathcal{L}^1_+$ we define the mapping $\mathbf{P}$ by

$$(\mathbf{P}\gamma)_n := \rho^N[\gamma] \tag{2.4}$$

and denote its range as $\mathcal{R}(\mathbf{P})$. Obviously, $\mathcal{R}(\mathbf{P}) \subset \mathcal{L}^\infty_+$, and $\mathbf{P} : \mathcal{L}^1_+ \to \mathcal{R}(\mathbf{P})$ is a bijection with its inverse given by

$$(\mathbf{P}^{-1}\sigma)_n = \begin{cases} \sigma_1 & \text{for} \quad n = 1 \\ \sigma_n - \sigma_{n-1} & \text{else} \quad . \end{cases} \tag{2.5}$$

Furthermore, $\sigma \in \mathcal{R}(\mathbf{P})$ iff σ_n is increasing.

The central concept of our convergence analysis is now presented. On $\mathcal{L}^1_+$ we introduce the partial ordering "$\preceq$" by

$$\gamma \preceq \xi \Leftrightarrow_{def} \rho^N[\gamma] \leq \rho^N[\xi] \quad \text{for all} \quad n \in \mathbb{N}. \tag{2.6}$$

The componentwise ordering "$\leq_c$" (on $\mathcal{L}^1_+$ and $\mathcal{L}^\infty_+$) is defined by

$$\gamma \leq_c \xi \Leftrightarrow_{def} \gamma_n \leq \xi_n \quad \text{for all} \quad n \in \mathbb{N}. \tag{2.7}$$

Componentwise convergence of sequences is indicated by "$\rightarrow_c$".

Some simple but useful results are

2.1 Lemma: (a) If $\gamma, \xi \in \mathcal{L}^1_+$ satisfy $\rho^\infty[\gamma] = \rho^\infty[\xi]$ and $\gamma \preceq \xi$ then $M[\gamma] \geq M[\xi]$.
(b) For γ, $\gamma^{(k)} \in \mathcal{L}^1_+$,

$$\gamma^{(k)} \rightarrow_c \gamma \Leftrightarrow \mathbf{P}\gamma^{(k)} \rightarrow_c \mathbf{P}\gamma \tag{2.8}$$

(c) If the sequence $\gamma^{(k)}$ in $\mathcal{L}^1_+$ is monotone and bounded with respect to $\preceq$, then $\gamma := \lim_{k\rightarrow\infty} \gamma^{(k)}$ exists in $\mathcal{L}^1_+$.
(d) Suppose given $\gamma, \gamma^{(k)} \in \mathcal{L}^1_+$, $k \in \mathbb{N}$, satisfying

$$\rho^\infty[\gamma^{(k)}] = \rho^\infty[\gamma], \tag{2.9}$$

$$\gamma^{(k)} \succeq \gamma, \tag{2.10}$$

$$\lim_{k\rightarrow\infty} M[\gamma^{(k)}] = M[\gamma] < \infty. \tag{2.11}$$

Then $\gamma^{(k)} \rightarrow_c \gamma$.

Proof: (a) follows from

$$\sum_{k=n}^\infty \gamma_k = \rho^\infty[\gamma] - \rho^{n-1}[\gamma] \geq \rho^\infty[\xi] - \rho^{n-1}[\xi] = \sum_{k=n}^\infty \xi_k \tag{2.12}$$

and

$$M[\gamma] = \sum_{n=1}^\infty \sum_{k=n}^\infty \gamma_k \geq \sum_{n=1}^\infty \sum_{k=n}^\infty \xi_k = M[\xi]. \tag{2.13}$$

(b) follows from (2.2) and (2.5).
(c) $\mathbf{P}\gamma^{(k)}$ is a monotone and bounded sequence in $\mathcal{R}(\mathbf{P}) \subset \mathcal{L}^\infty_+$ and thus converges

componentwise to some element in $\mathcal{R}(\mathbf{P})$.

(d) This follows from

$$\begin{aligned}\sum_{n=1}^{\infty}(\rho[\gamma]-\rho^{n-1}[\gamma]) &= \sum_{n=1}^{\infty}\sum_{\ell=n}^{\infty}\gamma_\ell = \sum_{\ell=1}^{\infty}\ell\gamma_\ell = M[\gamma] = \lim_{k\to\infty} M[\gamma^{(k)}] \\ &= \cdots = \lim_{k\to\infty}\sum_{n=1}^{\infty}(\rho[\gamma^{(k)}]-\rho^{n-1}[\gamma^{(k)}]) \qquad (2.14)\end{aligned}$$

and

$$\rho^{\infty}[\gamma]-\rho^{n-1}[\gamma] \geq \rho^{\infty}[\gamma^{(k)}]-\rho^{n-1}[\gamma^{(k)}] \qquad \square \qquad (2.15)$$

combined with (b). $\square$

The next results concerns monotonous $\mathcal{L}^1_+$-valued functions.

2.2 Lemma: Suppose $\mathbf{f} : [0,T] \to \mathcal{L}^1_+$ satisfies

$$\mathbf{f}(t) \succeq \mathbf{f}(t') \qquad \text{for} \quad 0 \leq t < t' \leq T \qquad (2.16)$$

$$\rho^{\infty}[\mathbf{f}(t)] = \rho^{\infty}[\mathbf{f}(0)] =: \rho \qquad \text{for} \quad 0 \leq t \leq T \qquad (2.17)$$

$$M[\mathbf{f}(T)] < \infty \qquad . \qquad (2.18)$$

Suppose further there exists a sequence of functions $\mathbf{f}^{(k)} : [0,T] \to \mathcal{L}^1_+$ satisfying

$$\mathbf{f}^{(k)}(t) \succeq \mathbf{f}(t) \qquad \text{for} \quad 0 \leq t \leq T, \quad k \in \mathbb{N} \qquad (2.19)$$

$$\rho^{\infty}[\mathbf{f}^{(k)}(t)] = \rho \qquad \text{for} \quad 0 \leq t \leq T \qquad (2.20)$$

$$M[\mathbf{f}^{(k)}(T)] \nearrow M[\mathbf{f}(T)] \qquad \text{for} \quad k \to \infty. \qquad (2.21)$$

Then $\mathbf{f}^{(k)}$ converges in the following sense.

(a) For any $\epsilon > 0$ there exists $N_0 \in \mathbb{N}$ such that for all $k \in \mathbb{N}$ and $t \in [0,T]$

$$\sum_{N=N_0+1}^{\infty} |\rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)]| \leq \epsilon \qquad (2.22)$$

(b) For any $\epsilon > 0$, $n, N \in \mathbb{N}$ and $t \in [0,T]$ there exists $K \in \mathbb{N}$ such that for $k \geq K$

$$|\rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)]| \leq \epsilon \qquad (2.23)$$

$$|f_n^{(k)}(t) - f_n(t)| \leq \epsilon \qquad (2.24)$$

Proof: (a) From Lemma 2.1(a) follows

$$M[\mathbf{f}^{(k)}(t)] \le M[\mathbf{f}(t)] \le M[\mathbf{f}(T)] \quad \text{for} \quad k \in \mathbb{N}, \quad t \in [0,T]. \tag{2.25}$$

For any $\mathbf{g} \in \mathcal{L}^1_+$ with finite moment $M[\mathbf{g}]$,

$$M[\mathbf{g}] = \sum_{\ell=1}^{\infty} \sum_{n=\ell}^{\infty} g_n = \rho^{\infty}[\mathbf{g}] + \sum_{N=1}^{\infty} (\rho^{\infty}[\mathbf{g}] - \rho^N[\mathbf{g}]) \tag{2.26}$$

Thus

$$0 \le M[\mathbf{f}(t)] - M[\mathbf{f}^{(k)}(t)] = \sum_{N=1}^{\infty} (\rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)]) \tag{2.27}$$

From (2.19) and (2.21) follows for $N \in \mathbb{N}$, $t \in [0,T]$

$$\rho^N[\mathbf{f}^{(k)}(t)] \ge \rho^N[\mathbf{f}(t)] \tag{2.28}$$

$$\rho^N[\mathbf{f}^{(k)}(t)] \to \rho^N[\mathbf{f}(t)] \qquad \text{for} \quad k \to \infty \tag{2.29}$$

Now choose $N_0 \in \mathbb{N}$ such that

$$\sum_{N=N_0+1}^{\infty} (\rho - \rho^N[\mathbf{f}(T)]) \le \epsilon \tag{2.30}$$

(2.16) implies

$$\sum_{N=N_0+1}^{\infty} (\rho - \rho^N[\mathbf{f}(t)]) \le \epsilon \qquad \text{for} \quad t \in [0,T] \tag{2.31}$$

$$\sum_{N=N_0+1}^{\infty} (\rho - \rho^N[\mathbf{f}^{(k)}(t)]) \le \epsilon \qquad \text{for} \quad t \in [0,T], \quad k \in \mathbb{N} \tag{2.32}$$

and thus

$$0 \le \sum_{N=N_0+1}^{\infty} \rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)] \le \epsilon \quad \text{for} \quad t \in [0,T], \quad k \in \mathbb{N}, \quad N \ge N_0 \tag{2.33}$$

(b) Choose N_0 as in (a). Then for all $N > N_0$,

$$0 \le \rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)] \le \sum_{N=N_0+1}^{\infty} \rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)] \le \epsilon \tag{2.34}$$

and because of (2.29) we find $K \in \mathbb{N}$ such that for all $k \geq K$ and $N \leq N_0$

$$0 \leq \rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)] \leq \epsilon \tag{2.35}$$

From this and

$$0 \leq \rho^{N+1}[\mathbf{f}^{(k)}(t)] - \rho^{N+1}[\mathbf{f}(t)] = f_N^{(k)}(t) - f_N(t) + \rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}(t)] \leq \epsilon \tag{2.36}$$

we conclude

$$|f_N^{(k)}(t) - f_N(t)| \leq \epsilon \qquad \square \tag{2.37}$$

A first link between the above ordering and the Smoluchowski equation is given by the first part of Proposition 2.3 for the following *linear* system.

Given a sufficiently regular function $\mathbf{b} : [0,T] \to \mathcal{L}^1_+$ with

$$0 < \sum_{n=1}^{\infty} b_n(t) \leq \overline{\rho} \quad \text{for all} \quad t \geq 0, \tag{2.38}$$

consider the linear Smoluchowski equation

$$\partial_t f_n \;=\; \sum_{i=1}^{n-1} \kappa(i) f_i b_{n-i} - \kappa(n) f_n \cdot \overline{\rho} \quad \text{for} \quad n \in \mathbb{N}, \quad \text{for} \quad t \geq 0, \tag{2.39}$$

with given $\kappa : \mathbb{N} \to \mathbb{R}_+$ satisfying

$$1 \leq \kappa(n) \leq n. \tag{2.40}$$

The unique solution $\mathbf{f}$ to a corresponding IVP can be readily constructed using the method described in section 1.1 for

$$g_n(t) = f_n(0) \cdot \exp(\kappa(n)\overline{\rho}t). \tag{2.41}$$

We easiliy find

2.3 Proposition: (a) For $0 \leq t \leq t'$,

$$\mathbf{f}(t') \preceq \mathbf{f}(t). \tag{2.42}$$

In particular, $\rho^\infty[\mathbf{f}(t)]$ is monotonically decreasing.

(b) Suppose

$$\sup_{t\in[0,T]} M[\mathbf{b}(t)] < \infty, \quad \text{and} \quad M[\mathbf{f}(0)] = M_0 < \infty. \tag{2.43}$$

Define $m(t)$ as the solution of the IVP

$$m'(t) = M[\mathbf{b}(t)] \cdot m(t), \quad m(0) = M_0. \tag{2.44}$$

Then

$$M[\mathbf{f}(t)] \le m(t) \quad \text{for} \quad t \in [0,T]. \tag{2.45}$$

If in addition $\|\mathbf{b}(t)\|_1 = \overline{\rho}$ for all t, then $\rho^\infty[\mathbf{f}(t)] = \rho^\infty[\mathbf{f}(0)]$.

(c) Choose $\kappa(n) = n$ and $\mathbf{b}(.) = \mathbf{f}(.)$. If $M[\mathbf{f}(0)] = M_0 < \infty$, and m is the solution of the IVP

$$m'(t) = m(t)^2, \quad m(0) = M_0, \tag{2.46}$$

then

$$M[\mathbf{f}(t)] = m(t) = \frac{M_0}{1 - M_0 \cdot t}. \tag{2.47}$$

Proof: (a) Like in the nonlinear case (compare (1.9)),

$$\partial_t \rho^N = \sum_{n=1}^{N} \kappa(n) f_n \left(\sum_{\ell=1}^{N-n} b_\ell - \overline{\rho} \right) \le 0. \tag{2.48}$$

(b) Define

$$M^N[\mathbf{f}] := \sum_{n=1}^{N} n f_n. \tag{2.49}$$

Then because of

$$\begin{aligned} &\sum_{n=1}^{N} n \sum_{i=1}^{n-1} \kappa(i) f_i b_{n-i} - \overline{\rho} \sum_{n=1}^{N} n\kappa(n) f_n \\ &= \sum_{i=1}^{N} \kappa(i) f_i \sum_{\ell=1}^{N-i} \ell b_\ell - \sum_{n=1}^{N} n\kappa(n) f_n \left(\overline{\rho} - \sum_{\ell_1}^{N-n} b_\ell \right) \le M[\mathbf{f}] \cdot M[\mathbf{b}], \end{aligned} \tag{2.50}$$

$M^N[\mathbf{f}(t)]$ and $M[\mathbf{f}(t)]$ are bounded by $m(t)$.

As long as $M[\mathbf{f}]$ is bounded, the condition $\|\mathbf{b}(t)\|_1 = \overline{\rho}$ and the theorem on dominated convergence yield $\partial_t \rho^\infty[\mathbf{f}] = 0$ when passing to the limit $N \to \infty$ in (2.48).

(c) From (b) follows $M[\mathbf{f}(t)] \leq m(t)$. For the inequality $M[\mathbf{f}(t)] \geq m(t)$, see Remark 3.5(a). $\square$

A central role in our investigations is played by a monotonicity property which is formulated in the following. The solution $\mathbf{f}$ of the above system depends on an initial condition $\mathbf{f}(0) = \gamma \in \mathcal{L}^1_+$, the function $\mathbf{b}(.)$ (serving as a "background medium"), and the function κ governing the interaction frequency. We indicate this dependence by writing $\mathbf{f}[\gamma, \mathbf{b}(.), \kappa]$.

2.4 Proposition: Suppose given $\gamma^{(1)}, \gamma^{(2)} \in \mathcal{L}^1_+$, two monotonous functions $\kappa^{(1)}, \kappa^{(2)} : \mathbb{N} \to [1, \infty)$ satisfying (2.40), and $\mathbf{b}^{(1)}, \mathbf{b}^{(2)} : [0, T] \to \mathcal{L}^1_+$ such that

$$\gamma^{(1)} \succeq \gamma^{(2)}, \tag{2.51}$$

$$\mathbf{b}^{(1)}(t') \succeq \mathbf{b}^{(2)}(t) \quad \text{for} \quad t', t \in [0, T], \quad t' \geq t, \tag{2.52}$$

$$\kappa^{(1)} \leq_c \kappa^{(2)}. \tag{2.53}$$

Then

$$\mathbf{f}[\gamma^{(1)}, \mathbf{b}^{(1)}(.), \kappa^{(1)}](t) \succeq \mathbf{f}[\gamma^{(2)}, \mathbf{b}^{(2)}(.), \kappa^{(2)}](t) \quad \text{for} \quad t \in [0, T]. \tag{2.54}$$

The proof will be given in the next section in terms of a Markov chain process producing a stochastic solution of the Smoluchowski system.

2.2 A Markov jump process

Under the condition (2.38), the system (2.39) may be interpreted as the generator of a certain non-homogeneous Markov jump process $X(t)$ on the countable state space $\overline{\mathbb{N}}$. (See, e.g. [8]; here we adopt the notation used there.) The realizations of $X(t)$ are piecewise constant, right continuous paths. The states $n \in \mathbb{N}$ are exponential holding states with waiting times distributed according to $\exp(-\kappa(n)\overline{\rho}t)$. The state ∞ is a trap.

For the description of $X(t)$ we use the following construction which is of particular use for the proof of the monotonicity properties of Proposition 2.4. Consider a pair $\omega = (\zeta, \xi)$, where ζ is a realization of a Poisson process with parameter $\overline{\rho}$, and $\xi = (\xi_n)_{n=0}^{\infty}$ is a sequence of i.i.d. random variables on $([0,1], d\lambda)$. ($d\lambda$ is the Lebesgue measure on $[0,1]$.) The path ζ is piecewise constant and increasing and can be described as

$$\zeta(t) = n \quad \text{for} \quad t \in [\tau_n, \tau_{n+1}), \tag{2.55}$$

where $\tau_0 = 0$ and the increments $\Delta\tau_n = \tau_{n+1} - \tau_n$ are independent and distributed according to $\exp(-\overline{\rho}t)$. Almost surely we have

$$\lim_{n\to\infty} \tau_n = \infty. \tag{2.56}$$

By recursion we now define the piecewise constant path $X(t, \omega)$ related to the Smoluchowski system with initial condition

$$\mathbf{f}(0) = \gamma \in \mathcal{L}^1_+, \quad \|\gamma\|_1 \le 1 \quad . \tag{2.57}$$

First of all, we define $T_0 := 0$ and $X_0 \in \overline{\mathbb{N}}$ distributed according to γ by putting

$$X_0 := \begin{cases} n & \text{if} \ \sum_{i=1}^{n-1} \gamma_i < \xi_0 \le \sum_{i=1}^{n} \gamma_i \\ \infty & \text{if} \ \sum_{i=1}^{\infty} \gamma_i < \xi_0 \quad . \end{cases} \tag{2.58}$$

Given T_k and $X_k = n$, we set

$$T_{k+1} := \begin{cases} T_k & \text{if} \quad X_k = \infty \\ T_k + \Delta\tau_k/\kappa(X_k) & \text{else} \end{cases} \tag{2.59}$$

and

$$X_{k+1} := \begin{cases} \infty & \text{if} \quad X_k = \infty \\ X_k + \Delta x_k & \text{else} \end{cases} \tag{2.60}$$

where

$$\Delta x_k := \begin{cases} n & \text{if} \quad \sum_{i=1}^{n-1} b_i(T_{k+1}) < \overline{\rho}\xi_{k+1} \leq \sum_{i=1}^{n} b_i(T_{k+1}) \\ \infty & \text{if} \quad \sum_{i=1}^{\infty} b_i(T_{k+1}) < \overline{\rho}\xi_{k+1} \quad . \end{cases} \tag{2.61}$$

Finally, we define the *explosion time*

$$T^* := \lim_{n\to\infty} T_n \tag{2.62}$$

and the path

$$X(t) = \begin{cases} X_n & \text{for} \quad t \in [T_n, T_{n+1}) \\ \infty & \text{if} \qquad t \geq T^* \quad . \end{cases} \tag{2.63}$$

For $n \in \overline{\mathbb{N}}$ define

$$f_n(t) := P(\{\omega : X(t) = n\}). \tag{2.64}$$

Then by construction f_n is continuous, $f_n(0) = \gamma_n$ for $n \in \mathbb{N}$, and by the formula on conditional probabilities follows

$$f_n(t + \Delta t) = f_n(t) \cdot (1 - \kappa(n)\overline{\rho}\Delta t) + \sum_{i=1}^{n-1} f_i(t)\kappa(i) \int_t^{t+\Delta t} b_{n-i}(s)ds + \mathcal{O}(\Delta t^2). \tag{2.65}$$

This proves

2.5 Lemma: $\mathbf{f} = (f_n)_{n\in\mathbb{N}}$ as defined by (2.64) is a solution of (2.39), (2.57).

Finally, we notice that

$$\rho^N[\mathbf{f}(t)] = P(\{\omega : X(t) \leq N\}). \tag{2.66}$$

To indicate the dependence of the paths on γ, $\mathbf{b}$ and κ, we write $T_n[\gamma, \mathbf{b}(.), \kappa]$, $X_n[\gamma, \mathbf{b}(.), \kappa]$, and $X[\gamma, \mathbf{b}(.), \kappa](t)$. The proof of Proposition 2.4 now follows from

2.6 Lemma: Let $\gamma^{(i)}, \mathbf{b}^{(i)}(.), \kappa^{(i)}$, $i = 1, 2$, be as in Proposition 2.4.
(a) For all $k \in \mathbb{N}$,

$$T_k[\gamma^{(1)}, \mathbf{b}^{(1)}(.), \kappa^{(1)}] \geq T_k[\gamma^{(2)}, \mathbf{b}^{(2)}(.), \kappa^{(2)}] \qquad a.s., \tag{2.67}$$

$$X_k[\gamma^{(1)}, \mathbf{b}^{(1)}(.), \kappa^{(1)}] \leq X_k[\gamma^{(2)}, \mathbf{b}^{(2)}(.), \kappa^{(2)}] \qquad a.s. \tag{2.68}$$

(b) For all $N \in \mathbb{N}$, $t \geq 0$

$$\{\omega : X[\gamma^{(2)}, \mathbf{b}^{(2)}(.), \kappa^{(2)}](t) \leq N\} \subseteq \{\omega : X[\gamma^{(1)}, \mathbf{b}^{(1)}(.), \kappa^{(1)}](t) \leq N\}. \tag{2.69}$$

Proof: For short we write $T_k^{(i)}$ instead of $T_k[\gamma^{(i)}, \mathbf{b}^{(i)}(.), \kappa^{(i)}]$ and similarly $X_k^{(i)}$ and $X^{(i)}(t)$.

(a) For $k = 0$, (2.67) and (2.68) hold by construction. Suppose (2.67) and (2.68) are valid for $k \in \mathbb{N}$. Then $T_{k+1}^{(1)} \geq T_{k+1}^{(2)}$ holds because of

$$\kappa^{(1)}(X_k^{(1)}) \leq \kappa^{(1)}(X_k^{(2)}) \leq \kappa^{(2)}(X_k^{(2)}). \tag{2.70}$$

Furthermore, from

$$\mathbf{b}^{(1)}(T_{k+1}^{(1)}) \succeq \mathbf{b}^{(2)}(T_{k+1}^{(2)}) \tag{2.71}$$

we deduce $\Delta x_{k+1}^{(1)} \leq \Delta x_{k+1}^{(2)}$ and $X_{k+1}^{(1)} \leq X_{k+1}^{(2)}$.

(b) Given $t > 0$, define $K^{(i)} \in \mathbb{N} \cup \{0\}$ such that

$$T_k^{(i)} \leq t < T_{k+1}^{(i)}. \tag{2.72}$$

Then because of $K^{(2)} \geq K^{(1)}$ and because the paths are monotonically increasing, we have

$$X^{(2)}(t) = X_{K^{(2)}}^{(2)} \geq X_{K^{(1)}}^{(2)} \geq X_{K^{(1)}}^{(1)} = X^{(1)}(t) \tag{2.73}$$

which proves (2.69). $\square$

3 Approximations to the Smoluchowski system

In the course of this section we will introduce two modifications of the Smoluchowski system. In the following we choose $\Delta t > 0$ and define $t_k := k\Delta t$. The first modification consists of a piecewise linearization and leads to the system in the time interval $[t_k, t_{k+1}]$,

$$\partial_t f_n(t) = \sum_{i=1}^{n-1} i f_i(t) f_{n-i}(t_k) - n\bar{\rho} f_n(t). \tag{3.1}$$

We will denote the solution $(f_n)_{n\in\mathbb{N}}$ of (3.1) (with given initial condition) as $\mathbf{f}^{(1)}$. The second modification concerns the stiffness problem caused by the factor $n\overline{\rho}$ in the loss term on the right hand side of (3.1). This leads to another approximation denoted as $\mathbf{f}^{(2)}$, which will be controlled by the results of section 2.

3.1 Piecewise linear systems

For a given continuous $\mathcal{L}^1_+$-valued function $\mathbf{b}(t)$ with $\|\mathbf{b}(t)\|_1 \leq \overline{\rho}$ consider the linear initial value problem (IVP) for $\mathbf{g}$

$$\partial_t g_n = \sum_{i=1}^{n-1} i g_i b_{n-i} - n\overline{\rho} g_n, \quad \mathbf{g}(T_0) = \mathbf{h}(T_0). \tag{3.2}$$

In the case $\mathbf{b} \equiv \mathbf{g}$, (3.2) is the original Smoluchowski system. Due to the linearity of the system, $\mathbf{g}$ can be written as

$$\mathbf{g}(t) = \sum_{r=1}^{\infty} g_r(T_0) \cdot \mathbf{s}^{[0,r]}(t) \tag{3.3}$$

where the *fundamental solutions* $\mathbf{s}^{[0,r]} = \mathbf{s}^{[0,r]}[\mathbf{b}]$ are defined as the unique solutions of the IVP's

$$\partial_t s_n = \sum_{i=1}^{n-1} i s_i b_{n-i} - n\overline{\rho} s_n, \quad s_n(T_0) = \delta_{n,r} \quad . \tag{3.4}$$

We introduce the approximation $\mathbf{g}^{(1)}$ of $\mathbf{g}$ given by

$$\mathbf{g}^{(1)}(t) = \sum_{r=1}^{\infty} g_r(T_0) \cdot \mathbf{s}^{[1,r]}(t) \tag{3.5}$$

where $\mathbf{s}^{[1,r]} = \mathbf{s}^{[0,r]}[\mathbf{b}(T_0)]$ is the fundamental solution obtained by replacing $\mathbf{b}(.)$ with its initial value $\mathbf{b}(T_0)$. The following results are easily derived.

3.1 Lemma: (a) Suppose $\mathbf{b}(.)$ satisfies $\mathbf{b}(t) \succeq \mathbf{b}(t')$ for all $T_0 \leq t \leq t'$. Then $\mathbf{g}$ and $\mathbf{g}^{(1)}$ are monotone in the sense that for all $t' \geq t$

$$\mathbf{g}(t) \succeq \mathbf{g}(t'), \quad \mathbf{g}^{(1)}(t) \succeq \mathbf{g}^{(1)}(t'). \tag{3.6}$$

Furthermore, for all $t \geq T_0$

$$\mathbf{g}(t) \succeq \mathbf{g}^{(1)}(t). \tag{3.7}$$

(b) If $\mathbf{b}$ satisfies in addition the inequalities (1.20),

$$b_n(t_0) \cdot \exp(-\overline{\rho} nt) \leq b_n(t_0 + t) \leq [b_n(t_0) + \phi_n(t) \cdot t] \cdot \exp(-\overline{\rho} nt), \tag{3.8}$$

then there exist constants γ_n (not depending on $\mathbf{g}(T_0)$ or $\mathbf{b}$) such that for all $t \in [T_0, T_0 + 1]$

$$|g_n(t) - g_n^{(1)}(t)| \leq \gamma_n (t - T_0)^2. \tag{3.9}$$

Proof: (a) is a special case of Proposition 2.4.

(b) follows from the Lipschitz continuity of b_n with Lipschitz constants not depending on $\mathbf{g}(T_0)$ or $\mathbf{b}$. We want to mention that a more detailed analysis using the function $\mathbf{a}$ of formulas (1.17), (1.18) yield

$$|s_n^{[0,r]}(t) - s_n^{[1,r]}(t)| \leq t^2 \Phi_n (t - T_0) \cdot \exp\left(-n\overline{\rho}(t - T_0)\right) \tag{3.10}$$

from which we may derive a more detailed estimate for $|g_n(t) - g_n^{(1)}(t)|$. $\square$

Now let $\mathbf{f}$ be a solution of the Smoluchowski system, and define the continuous function $\mathbf{f}^{(1)}$ by $\mathbf{f}^{(1)}(t_0) = \mathbf{f}(t_0)$, and in $[t_k, t_{k+1}]$ as the solution of

$$\partial_t f_n = \sum_{i=1}^{n-1} i f_i f_{n-i}(t_k) - n\overline{\rho} f_n. \tag{3.11}$$

Then a consequence of the preceding lemma is

3.2 Lemma: (a) $\mathbf{f}^{(1)}$ is monotonically decreasing, and for all $t \geq t_0$

$$\mathbf{f}(t) \preceq \mathbf{f}^{(1)}(t). \tag{3.12}$$

(b) Fix $T_1 > T_0$. Then there exist constants c_n and M_n such that for all $\Delta t \leq 1$ and $t \in [T_0, T_1]$

$$|f_n(t) - f_n^{(1)}(t)| \leq \Delta t c_n \exp(M_n (t - T_0)) \quad . \tag{3.13}$$

Proof: (a) For $t \in [t_0, t_1]$, the inequalities follow from Lemma 3.1(a). The proof for $t \in [t_k, t_{k+1}]$, $k > 0$, follows by induction from Propositions 2.3(a) and 2.4.
(b) Lemma 3.1(b) proves that the approximation $\mathbf{f}^{(1)}$ for $\mathbf{f}$ is consistent with order 1. Thus (b) follows from standard arguments of numerics for ODE's. (Keep in mind that for the estimate (3.13) we only need to consider the *finite-dimensional* systems $(f_i)_{i=1}^n$ resp. $(f_i^{(1)})_{i=1}^n$.) $\square$

3.2 Truncation of interaction frequency

One major problem in establishing a numerical scheme is the stiffness of the system $\mathbf{f}$ caused by the loss term $-n\overline{\rho} f_n$. Using the description as a stochastic process of section 2.2, a particle starting from state r at time t_i, stays in this state for some time distributed according to $r\exp(-rt)$ and then jumps into some state $r + \ell > r$. The new jump time is shorter (in the mean), since it is distributed as $(r+\ell)\exp(-(r+\ell)t)$. The next modification we introduce is a slight truncation in the sense that in the whole time interval $[t_k, t_{k+1})$ the distribution of the jump times is fixed at the initial distribution $r\exp(-rt)$. As we find below, we are able to control this modification in terms of the moment $M[\mathbf{f}]$.

Choose $\mathbf{b} \in \mathcal{L}_+^1$ with $\|\mathbf{b}\|_1 = \overline{\rho}$ and define $\mathbf{s}^{[2,r]}$ as the solution of

$$\partial_t s_n^{[2,r]} = r \sum_{i=1}^{n-1} s_i^{[2,r]} b_{n-i} - r\overline{\rho} s_n^{[2,r]}, \quad s_n(T_0) = \delta_{r,n}. \tag{3.14}$$

3.3 Lemma: (a) $\mathbf{s}^{[2,r]}$ satisfies the *mass conservation property*

$$\|\mathbf{s}^{[2,r]}(t)\|_1 = 1 \quad \text{for} \quad t \geq T_0 \tag{3.15}$$

(b) For all $n \leq r$, $s_n^{[2,r]} - s_n^{[1,r]} \equiv 0$.
(c) For $q > 0$

$$|s_{r+q}^{[2,r]}(t) - s_{r+q}^{[1,r]}(t)| \leq 2\,(r+q)^2\,(t - T_0)^2. \tag{3.16}$$

(d) For $t' \geq t \geq 0$,

$$\mathbf{s}^{[2,r]}(t) \succeq \mathbf{2}^{[2,r]}(t') \succeq \mathbf{s}^{[1,r]}(t') \tag{3.17}$$

(e) For $t \geq 0$,

$$M[\mathbf{s}^{[2,r]}(t)] = r\left(1 + M[\mathbf{b}] \cdot (t - T_0)\right). \tag{3.18}$$

Proof: (a) Because of $\|\mathbf{b}\|_1 = \overline{\rho}$, $\|\mathbf{s}^{[2,r]}(t)\|$ satisfies the differential equation

$$\partial_t \|\mathbf{s}^{[2,r]}\| = 0 \tag{3.19}$$

(b) From the definition follows

$$s_n^{[1,r]} \equiv s_n^{[2,r]} \equiv 0 \qquad \text{for} \quad n < r \tag{3.20}$$

$$s_r^{[1,r]}(t) = s_r^{[2,r]}(t) \;=\; \exp\left(-r\overline{\rho}(t - T_0)\right) \geq 1 - r\overline{\rho}(t - T_0). \tag{3.21}$$

(c) From (a) and (b) follows

$$s_{r+q}(t) \;\leq\; r\overline{\rho}(t - T_0) \quad \text{for} \quad q \geq 1. \tag{3.22}$$

Thus for $q > 0$

$$s_{r+q}^{[1,r]}(t) = \sum_{i=r}^{r+q-1} i b_{r+q-i} \int_{T_0}^{t} s_i(\tau) \exp[-(r+q)\overline{\rho}(t-\tau)] d\tau \tag{3.23}$$

which leads to

$$\begin{aligned} 0 \;&\leq\; s_{r+q}^{[1,r]}(t) - r b_q \int_{T_0}^{t} s_r^{[1,r]}(\tau) \exp[-(r+q)\overline{\rho}(t-\tau)] d\tau \\ &\leq\; (r+q) \underbrace{\left(\sum_{i=r+1}^{r+q-1} b_{r+q-i}\right)}_{\leq \|b\|_1 = \overline{\rho}} \cdot r\overline{\rho} \int_{T_0}^{t} (\tau - T_0) \exp[-(r+q)\overline{\rho}(t-\tau)] d\tau \\ &\leq\; \frac{1}{2}[(r+q)\overline{\rho}]^2 \cdot (t - T_0)^2 \end{aligned} \tag{3.24}$$

and

$$-\frac{1}{2}[(r+q)\overline{\rho}]^2 \cdot (t-T_0)^2 \leq s_{r+q}^{[1,r]}(t) - rb_q \int_{T_0}^{t} s_r^{[1,r]}(\tau)d\tau \leq [(r+q)\overline{\rho}]^2 \cdot (t-T_0)^2 \quad (3.25)$$

Similarly,

$$-\frac{1}{2}[r\overline{\rho}]^2 \cdot (t-T_0)^2 \quad \leq \quad s_{r+q}^{[2,r]}(t) - rb_q \int_{T_0}^{t} s_r^{[1,r]}(\tau)d\tau \leq [r\overline{\rho}]^2 \cdot (t-T_0)^2 \qquad (3.26)$$

which proves statement (c).

(d) follows from Propositions 2.3(a) and 2.4, and (e) from

$$\begin{aligned} \partial_t M[\mathbf{s}^{[2,r]}] &= r \cdot \left(\sum_{n=1}^{\infty} n \sum_{i=1}^{n-1} s_i^{[2,r]} b_{n-i} - \overline{\rho} \sum_{n=1}^{\infty} n s_n^{[2,r]} \right) && (3.27) \\ &= r \cdot \|\mathbf{s}^{[2,r]}\|_1 M[\mathbf{b}] = r \cdot M[\mathbf{b}] \qquad \square && (3.28) \end{aligned}$$

We now arrive at the approximation system which will serve us as the basis of a numerical scheme. Given a time step $\Delta t > 0$, choose $t_k = k \cdot \Delta t$, and define $\mathbf{f}^{\Delta t}$ as the continuous function given by $\mathbf{f}^{\Delta t}(0) = \mathbf{f}(0)$, and in $[t_k, t_{k+1}]$ by

$$\mathbf{f}^{\Delta t}(t) = \sum_{r=1}^{\infty} f_r^{\Delta t}(t_k) \mathbf{s}^{[2,r,k]}(t) \qquad (3.29)$$

where $\mathbf{s}^{[2,r,k]}$ is given by the differential equation (3.14) and the initial condition $s_n^{[2,r,k]}(t_k) = \delta_{r,n}$. The main results concerning $\mathbf{f}^{\Delta t}$ are collected in the following theorem. Let $\mathbf{f}$ denote solution of the original Smoluchowski IVP. Furthermore, let t_{gel} be the gelation time as defined in section 1.1.

3.4 Theorem: (a) For all $t \geq 0$,

$$\|\mathbf{f}^{\Delta t}(t)\|_1 = \|\mathbf{f}(0)\|_1 = 1 \qquad (3.30)$$

(b) For all $t \geq 0$,

$$\mathbf{f}^{\Delta t}(t) \succeq \mathbf{f}(t) \qquad (3.31)$$

(c) For all $t \geq 0$,

$$M[\mathbf{f}^{\Delta t}(t)] \nearrow M[\mathbf{f}(t)] \quad \text{as} \quad \Delta t \searrow 0 \tag{3.32}$$

(d) Choose $T > 0$ fixed. There exist constants $c_n > 0$ such that

$$\sup_{t \in [0,T]} |f_n^{\Delta t}(t) - f_n(t)| \leq c_n \Delta t \tag{3.33}$$

(e) Choose $T < T_{gel}$ and $\Delta t_k := T/k$; denote $\mathbf{f}^{(k)} := \mathbf{f}^{\Delta t_k}$. Then for all $\epsilon > 0$ there exists $K(\epsilon)$ such that

$$\sup_{k \geq K(\epsilon)} \sup_{t \in [0,T]} \sup_{N \in \mathbb{N}} \left| \rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}^{(k)}(t)] \right| \leq \epsilon \tag{3.34}$$

(f) Define T and $\mathbf{f}^{(k)}$ as in (e). Then for all $\epsilon > 0$ there exists $K(\epsilon)$ such that

$$\sup_{k \geq K(\epsilon)} \sup_{t \in [0,T]} \sup_{N \in \mathbb{N}} \left| f_n^{(k)}(t)] - f_n^{(k)}(t)] \right| \leq \epsilon \tag{3.35}$$

(g) For $t \leq t_{gel}$

$$\lim_{N \to \infty} \lim_{\Delta t \searrow 0} \rho^N[\mathbf{f}^{\Delta t}(t)] = \lim_{\Delta t \searrow 0} \lim_{N \to \infty} \rho^N[\mathbf{f}^{\Delta t}(t)] = \|\mathbf{f}(0)\|_1, \tag{3.36}$$

and for $t > t_{gel}$

$$\lim_{N \to \infty} \lim_{\Delta t \searrow 0} \rho^N[\mathbf{f}^{\Delta t}(t)] < \lim_{\Delta t \searrow 0} \lim_{N \to \infty} \rho^N[\mathbf{f}^{\Delta t}(t)] = \rho(0). \tag{3.37}$$

Proof: (a) follows from the mass conservation property of Lemma 3.3(a).
(b) is a consequence of

$$\mathbf{s}^{[2,r]}(t) \succeq \mathbf{s}^{[1,r]}(t) \succeq \mathbf{s}(t) \tag{3.38}$$

(compare Lemmas 3.1(a) and 3.3(d)) and Proposition 2.3(a).
(c) From Lemma 3.3(e) follows that $M[\mathbf{f}^{\Delta t}]$ is linear in $[t_i, t_{i+1}]$, and

$$M[\mathbf{f}^{\Delta t}(t_{i+1})] = M[\mathbf{f}^{\Delta t}(t_i)] \left(1 + \Delta t \cdot M[\mathbf{f}^{\Delta t}(t_i)]\right) \tag{3.39}$$

Therefore $M[\mathbf{f}^{\Delta t}(t)]$ is the Euler approximation of the convex functional $M[\mathbf{f}(t)]$ and thus a lower approximation.
(d) The results of Lemmas 3.2(b) and 3.3(c) show that for $N \in \mathbb{N}$, the finite system $(f_n^{\Delta t})_{n=1}^N$ is an approximation of $(f_n)_{n=1}^N$ with consistency order Δt. Thus for finite time intervals $[0, T]$, the global approximation error is of order Δt.
(e) $\mathbf{f}^{\Delta t}$ satisfies the assumptions of Lemma 2.2. Thus from Lemma 2.2(a) we conclude that there exist $K, N_0 \in \mathbb{N}$ such that

$$\sup_{k \geq K(\epsilon)} \sup_{t \in [0,T]} \sup_{N > N_0} \left| \rho^N[\mathbf{f}^{(k)}(t)] - \rho^N[\mathbf{f}^{(k)}(t)] \right| \leq \epsilon \tag{3.40}$$

The remaining part follows from the uniform convergence of $f_n^{\Delta t}(t)$ given in (d).
To prove (f), we use the uniform convergence (e) and the arguments of the proof of Lemma 2.2(b).
(g) is a consequence of

$$\rho^N[\mathbf{f}^{\Delta t}(t)] \nearrow \|\mathbf{f}^{\Delta t}(t)\|_1 = \|\mathbf{f}^{\Delta t}(0)\|_1 \quad \text{as} \quad N \to \infty \tag{3.41}$$

and

$$\lim_{N \to \infty} \lim_{\Delta t \searrow 0} \rho^N[\mathbf{f}^{\Delta t}(t)] = \lim_{N \to \infty} \rho^N[\mathbf{f}(t)] = \rho[\mathbf{f}(t)]. \qquad \square \tag{3.42}$$

3.5 Remarks: (a) Formula (3.39) proves that $M[\mathbf{f}^{\Delta t}(t)]$ converges for $\Delta t \searrow 0$ to $m(t)$ given by (2.47). Since the correct value $M[\mathbf{f}(t)]$ is an upper bound of $M[\mathbf{f}^{\Delta t}(t)]$, this proves that $M[\mathbf{f}(t)] \geq m(t)$ which completes the proof of Proposition 2.3(c).
(b) Theorem 3.4 shows different types of convergence $\mathbf{f}^{\Delta t}(t) \to \mathbf{f}(t)$. For $T < t_{gel}$ we have uniform convergence in $[0, T]$ which breaks down at the gelation time. At the same moment, the interchange of limits $\Delta t \searrow 0$ and $N \to \infty$ is no longer allowed.

4 Numerical investigations

4.1 A numerical scheme

We are going to establish an easy to handle and efficient scheme for the simulation of $\mathbf{f}^{\Delta t}$. Since $\mathbf{f}^{\Delta t}$ is an approximation to $\mathbf{f}$ of order Δt, it is sufficient to construct a numerical scheme of the same order. Here, we restrict to the simplest explicit version, and we will prove its applicability for the transition to the gelation phase.

Given $\mathbf{b} \in \mathcal{L}^1_+$ with

$$b_n \geq 0, \quad \|\mathbf{b}\|_1 = \overline{\rho} \tag{4.1}$$

we define the mapping $\Theta = \Theta[\mathbf{b}] = (\theta_{i,j}) : \mathbb{R}^{\mathbb{N}} \to \mathbb{R}^{\mathbb{N}}$ by

$$\theta_{i,j} = \begin{cases} 0 & \text{if} \quad i < j \\ -\overline{\rho} & \text{if} \quad i = j \\ b_{i-j} & \text{if} \quad i > j \end{cases} . \tag{4.2}$$

For numerical reasons we also introduce a truncated version. For $N \in \mathbb{N}$ define the $N \times N$-matrix $\Theta^N = (\theta^N_{i,j})$ by

$$\theta^N_{i,j} = \begin{cases} \theta_{i,j} & \text{if} \quad N > i \geq j \\ 0 & \text{if} \quad j = N \\ \overline{\rho} - \sum_{k=1}^{N-1} b_{k-j} & \text{if} \quad N = i > j \end{cases} . \tag{4.3}$$

For $0 \leq \lambda \leq 1/\overline{\rho}$ define

$$P_\lambda = (p^\lambda_{i,j}) := I + \lambda\Theta, \quad P^N_\lambda = (p^{\lambda,N}_{i,j}) := I + \lambda\Theta^N. \tag{4.4}$$

It is evident that the system

$$\eta_0 = \mathbf{f}(0), \quad \eta_{i+1} = P_{\Delta t}\eta_i, \quad i = 0, 1, 2, \ldots \tag{4.5}$$

describes the first order Euler scheme for the ODE system

$$\partial_t f_n = \sum_{i=1}^{n-1} f_i b_{n-i} - \overline{\rho} f_n. \tag{4.6}$$

The following statements are obvious resp. straightforward.

4.1 Remarks: (a) For all $\lambda \in [0, 1/\overline{\rho}]$, P_λ and P_λ^N are stochastic matrices in the sense that their coefficients satisfy

$$p_{i,j}^\lambda, p_{i,j}^{\lambda,N} \geq 0, \quad \text{and} \quad \sum_{j=1}^{\infty} p_{i,j}^\lambda = \sum_{j=1}^{N} p_{i,j}^{\lambda,N} = 1. \tag{4.7}$$

(b) From (a) follows that P_λ and P_λ^N are mass conserving in the sense that for $\mathbf{f} \in \mathcal{L}_+^1$

$$\|P_\lambda \mathbf{f}\|_1 = \|\mathbf{f}\|_1, \quad \rho^N[P_\lambda^N \mathbf{f}] = \rho^N[\mathbf{f}]. \tag{4.8}$$

(c) P_λ is transient, satisfying

$$(P_\lambda)^k \rightarrow_c 0 \quad \text{for} \quad k \rightarrow \infty. \tag{4.9}$$

P_λ^N is obtained from P_λ by redirecting all paths which leave the lower state space $\{1, \ldots, N\}$ under P_λ into state N. The state N is a trap under P_λ^N, and

$$\lim_{k\rightarrow\infty} (P_\lambda^N)^k = \begin{pmatrix} 0 & \cdots & \cdots & 0 \\ \vdots & & & \vdots \\ 0 & \cdots & \cdots & 0 \\ 1 & \cdots & \cdots & 1 \end{pmatrix} \tag{4.10}$$

(d) If $\|\mathbf{b}\|_1 < \overline{\rho}$, then P_λ^N is still a stochastic matrix. We may turn P_λ into a stochastic matrix by adding an additional state ∞ to the state space $\mathbb{N}$ and by defining

$$\theta_{\infty,j} := \begin{cases} \overline{\rho} - \sum_{i=1}^{\infty} b_i & \text{for} \quad j \in \mathbb{N} \\ 0 & \text{for} \quad j = \infty \end{cases} . \tag{4.11}$$

To avoid stiffness problems, we have to numerically approximate $\mathbf{s}^{[2,r]}(\Delta t)$ as given in section 3.2 by $(P_{\Delta t/r})^r \mathbf{s}^{[2,r]}(0)$ which yields an approximation of the order 1, i.e.

$$\|\mathbf{s}^{[2,r]}(\Delta t) - (P_{\Delta t/r})^r \mathbf{s}^{[2,r]}(0)\|_1 = \mathcal{O}(\Delta t^2). \tag{4.12}$$

Finally we need the matrices $E_r \in \mathbb{R}^{\mathbb{N}\times\mathbb{N}}$ and $E_r^N \in \mathbb{R}^{N\times N}$ given by

$$e_{i,j} = \begin{cases} 1 & \text{if} \quad i = j = r \\ 0 & \text{else} \end{cases} \tag{4.13}$$

to define the matrices

$$Q_{\Delta t} := \sum_{r=1}^{\infty} (P_{\Delta t/r})^r E_r, \quad Q_{\Delta t}^N := \sum_{r=1}^{\infty} (P_{\Delta t/r}^N)^r E_r^N \quad . \tag{4.14}$$

For $\Delta t \leq \overline{\rho}$, these are again stochastic matrices which follows immediately from the Remarks 4.1. Furthermore, for $r, n < N$ and δ_r being the r-th canonical unit vector,

$$\left((P_{\Delta t/r})^r \mathbf{s}^{[2,r]}(0)\right)_n = (Q_{\Delta t}\delta_r)_n = \left(Q_{\Delta t}^N \delta_r\right)_n \quad . \tag{4.15}$$

We end up with the following first order scheme $\mathbf{f}^{\mathrm{num}}$ for $\mathbf{f}^{\Delta t}$ (and with this for our main object, the solution $\mathbf{f}$ of the Smoluchowski system) which is recursively defined by

$$\mathbf{f}^{\mathrm{num}}(0) = \mathbf{f}(0), \quad \mathbf{f}^{\mathrm{num}}((i+1)\Delta t) = Q_{\Delta t}\mathbf{f}^{\mathrm{num}}(i\Delta t) \quad . \tag{4.16}$$

Its finite dimensional version, which coincides with $\mathbf{f}^{\mathrm{num}}$ in all components $n < N$, reads

$$\mathbf{f}^{\mathrm{num},N}(0) = \mathbf{f}(0), \quad \mathbf{f}^{\mathrm{num},N}((i+1)\Delta t) = Q_{\Delta t}^N \mathbf{f}^{\mathrm{num},N}(i\Delta t) \quad . \tag{4.17}$$

4.2 Numerical transition to the gelation time

The numerical system derived in the preceding section is readily transformed into a computer program. There are several possibilities of its realization. First, we may take the scheme $\mathbf{f}^{\mathrm{num},N}$ as it is for the simulation of *finite* systems $\mathbf{f}^N(t) = (f_n(t)_{n=1}^N$. Since the numerical effort is at least of order $\mathcal{O}(N^2)$, this quantity should be only moderate for the sake of efficiency. Second, since Q is a stochastic matrix, we may implement a Monte Carlo scheme, thus being able to use much larger state spaces. Such a scheme has been proposed and proven useful for the study of gelation in a space dependent environment in [3, 4], and it can certainly compete with alternative stochastic schemes (see, e.g. [5, 7]) in particular concerning numerical efficiency. However, such systems are affected with unavoidable fluctuations which are hard to control quantitatively. Furthermore, answers to questions concerning a choice of threshold levels for the simulation of the gelation phase are not yet convincing. For this reason, we present a third realization, which is a hybrid code combining a deterministic and a stochastic part.

In the following we denote again as $\mathbf{f}^{\text{num}}$ the numerical simulation of the deterministic approach. We compare the simulated solutions with the exact solution

$$f_n(t) = \frac{1}{n}\frac{(nt)^{n-1}}{n!}\exp(-nt) \tag{4.18}$$

given in [9]. (Notice that the exact solutions e.g. mentioned in [1, 6], concern the *passive* gel and may be derived from (4.18) via the transformation (1.19).)

Before running the numerical scheme we have to determine a time step Δt. A criterium for a proper choice is found in the fact that the numerical value $M[\mathbf{f}^{\text{num}}]$ of the first moment is a lower bound for the correct value $M[\mathbf{f}]$ and that it is given as the Euler discretization of the correct function (2.47) (see Theorem 3.4 and Remark 3.5(a)). So it is easy to check this error depending on Δt in a simple scalar simulation. For $t = 0.9$ (e.g. close to t_{gel}) and for time steps Δt between 10^{-4} and 10^{-2} the calculations show relative errors of

$$\frac{M[\mathbf{f}(0.9)] - M[\mathbf{f}^{\text{num}}(0.9)]}{M[\mathbf{f}(0.9)]} \approx 20 \cdot \Delta t. \tag{4.19}$$

In Fig. 1 we compare for $\Delta t = 0.01$ the exact functions $\rho^N[\mathbf{f}(t)]$ (solid lines) with the numerical ones (dotted lines) for $N = 3, 9, 27, 81$ (curves from left to right). As was to be expected, the numerical values lie above the theoretical ones. However, they are reasonable approximations. When increasing N even more one observes that the functions approach a sharp edge at $t = 1$, the initialization of gelation. The simulation of this point will be the subject of the following investigations.

The next problem concerns the question of how to simulate the transition to the gel phase. The only possible way seems to be the truncation of the state space at some threshold level N_{tr} and to consider the whole mass above this level as gelated mass. E.g., truncating at $N_{tr} = 100$ leads to a picture presented in Fig. 2, where the thick solid line presents $\rho^{N_{tr}}[\mathbf{f}^{\mathrm{num}}]$ in comparison to the non-gelled mass of the exact solution (dashed line), the latter exhibiting a discontinuity in the first derivative at t_{gel}. (The time step is $\Delta t = 0.001$.) We observe that truncation at $N_{tr} = 100$ does not describe the approach to the gelation phase very well. (The deviations at times $t \gg t_{gel}$ result from the discretization error; the system is only of first order.) Let us have a course look at the mass distribution of the exact solution around the gelation time. Fig. 3 exhibits the "sparsely populated" areas of the state space depending on time. (The abszissa shows $100t$; the value 100 corresponds to the gelation time. The ordinate shows the state number N.) Here, a state k is marked white at time t, if

$$\sum_{\ell=k}^{\infty} f_\ell(t) < \Delta m \tag{4.20}$$

with a threshold level chosen as $\Delta m = 0.01$. The picture demonstrates the region between the dark domain and ∞ which is almost unpopulated. This means that mass leaving the dark domain very quickly passes over to ∞. A similar picture may be obtained for the *numerical* solution, if one replaces the sum in (4.20) with $\sum_{\ell=k}^{K} f_\ell^{\mathrm{num}}(t)$ and K reasonably large. In this case, mass is again moving away very fast from the dark region but it disappears in the upper state space and fails to reach ∞.

Taking these figures into consideration, choosing a threshold level in the white domain should give a good description of the gel transition. E.g., taking $N_{tr} = 100$ should be adequate for all times $t \geq 0$ except for $t \in [0.72, 1.30]$. In this situation it seems near at hand to supplement the deterministic truncated system by a stochastic part simulating the transition in the critical phase. To this aim we implement a feature letting particles emerge randomly into states $n > N_{tr}$ at rates $\Delta t \cdot \sum_{i=1}^{N_{tr}} i f_i f_{n-i}$, and we let these random particles evolve stochastically as in the MC scheme described in [4, Section 2.2.2]. As a new threshold N_{gel} for gelation we choose now a level $N_{gel} \gg N_{tr}$. As it turns out, the results are quite robust with respect to the choice of N_{gel}.

The results of a run are shown in Fig. 4. Here, we have chosen a time step $\Delta t = 0.001$, a gelation threshold $N_{gel} = 15000$ and the weight $1/N_{gel}$ for the random particles. The solid line shows the non-gelled mass consisting of the deterministic part up to state $n = N_{tr}$ and the stochastic particles within states $n < N_{gel}$. The dashed line shows the first moment M of the combined system. The thick dots represent the number of random particles. As it turns out, particles exist at times $0.721 < t < 1.270$ which lie completely in the time interval which we have identified above as the critical one for the simulation of gelation. The maximal number of 1164 particles showed up at time $t = 1.009$, i.e. very close to gelation time. The thin dots exhibit the whole gelated mass. As the thin solid line in Fig. 2 proves, the portion of non-gelled mass is in the neighborhood of t_{gel} quite close to the theoretical values (dashed line). These calculations have been performed on a usual PC (Intel, 3GHz, 512MB, MS-Windows) under Delphi. The pure calculation time (without IO operations) was 170 sec. which means an average of 0.11 sec. per time step. Thus the scheme is well-suited for simulations of space dependent problems.

5 Figures

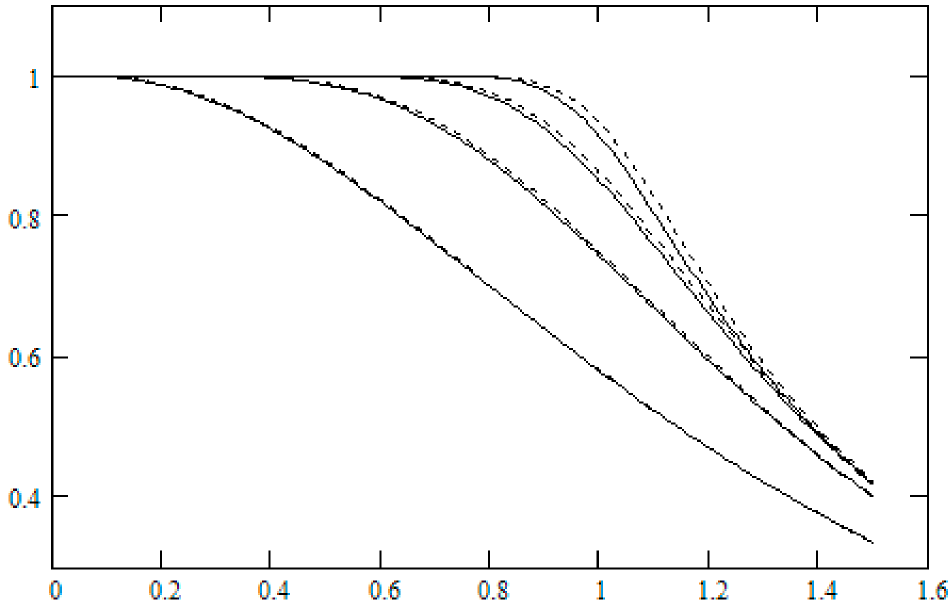

Fig. 1. Exact and numerical values of $\rho^N(t)$, $N = 3, 9, 27, 81$.

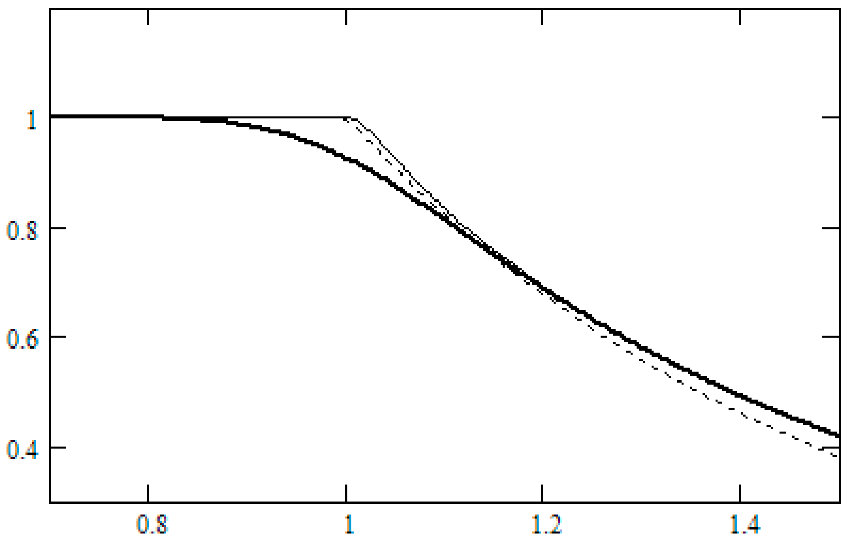

Fig. 2. Non-gelled mass: exact (dotted), truncated (thick solid), hybrid code (thin solid).

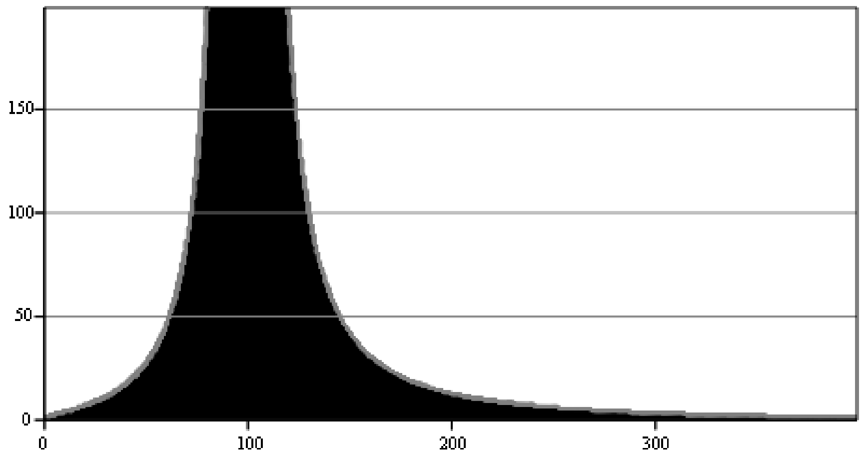

Fig. 3. Sparsely populated areas.

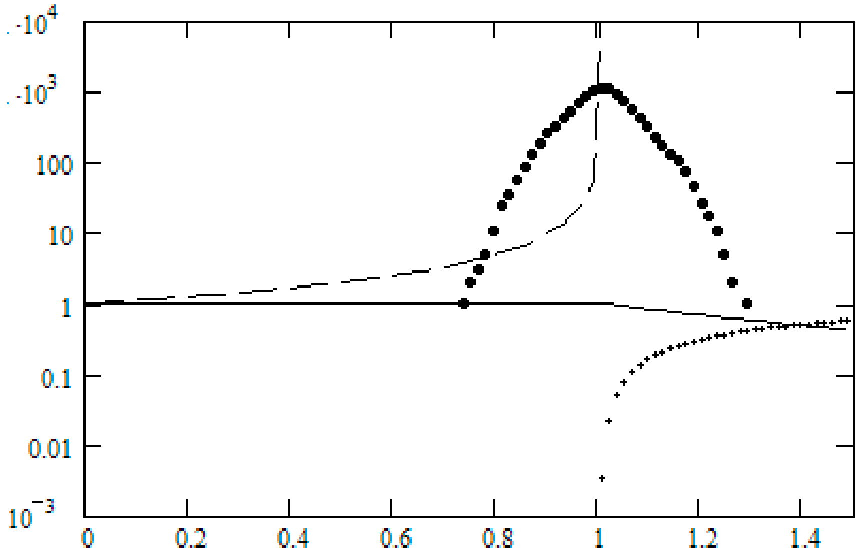

Fig. 4. Passing the gelation time with the hybrid code.

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