Journal of Statistical Physics, Vol. 96, Nos. 5/6, 1999

Spouge's Conjecture on Complete and Instantaneous Gelation

Intae Jeon[1]

Received November 24, 1998; final May 3, 1999

We investigate the stochastic counterpart of the Smoluchowski coagulation equation, namely the Marcus–Lushnikov coagulation model. It is believed that for a broad class of kernels, all particles are swept into one huge cluster in an arbitrarily small time, which is known as a complete and instantaneous gelation phenomenon. Indeed, Spouge (also Domilovskii *et al.* for a special case) conjectured that $K(i, j) = (ij)^\alpha$, $\alpha > 1$, are such kernels. In this paper, we extend the above conjecture and prove rigorously that if there is a function $\psi(i, j)$, increasing in both i and j such that $\sum_{j=1}^{\infty} 1/(j\psi(i, j)) < \infty$ for all i, and $K(i, j) \geqslant ij\psi(i, j)$ for all i, j, then complete and instantaneous gelation occurs. Evidently, this implies that any kernels $K(i, j) \geqslant ij(\log(i+1)\log(j+1))^\alpha$, $\alpha > 1$, exhibit complete instantaneous gelation. Also, we conjuncture the existence of a critical (or metastable) sol state: if $\lim_{i+j\to\infty} ij/K(i, j) = 0$ and $\sum_{i,j=1}^{\infty} 1/K(i, j) = \infty$, then gelation time T_g satisfies $0 < T_g < \infty$. Moreover, the gelation is complete after T_g.

KEY WORDS: Marcus–Lushnikov process; Smoluchowski coagulation equation; gelation; complete and instantaneous gelation; sol–gel interaction.

INTRODUCTION

Suppose clusters are moving around in a container with volume V. The Marcus–Lushnikov process (or ML process) is a stochastic model describing the coagulation dynamics: a given i-cluster and j-cluster stick together to form an $(i+j)$-cluster at rate $K(i, j)/V$, where $K(i, j)$ is a given non-negative kernel. Here, by i-cluster, we mean a cluster which consists of i particles. The kernel, $K(i, j)$, is divided by V due to the density dependent nature of this model, which is described nicely in Chap. 30 of Ethier and Kurtz's book [EK].

[1] Department of Mathematics, Ohio State University, Columbus, Ohio 43210; e-mail: jeon@math.ohio-state.edu.

Obviously, this ML process is a stochastic counterpart of the well known Smoluchowski coagulation equation, which consists of an infinite set of non-linear ordinary differential equations given by

$$\frac{d}{dt} C_t(j) = \frac{1}{2} \sum_{k=1}^{j-1} K(j-k, k)\, C_t(j-k)\, C_t(k) - \sum_{k=1}^{\infty} K(j, k)\, C_t(j)\, C_t(k) \qquad (1)$$

for $j = 1, 2, 3, \ldots$, where $C_t(j) \geqslant 0$ is the expected number of j-clusters per unit volume.

Various aspects of this model have been extensively studied by many authors in physical literature for a couple of decades, and a variety of applications in physical, chemical and biological science have been provided [BCP, BP, GL, HEZ, LT, Lu, Ma, Mc, N. vE1, vE2]. Recently, rigorous mathematics was brought to bear on this model making the cooperation between mathematics and physics more fruitful. For readers who are interested in the mathematical aspects of this model, we recommend the survey paper of Aldous [A1].

Perhaps one of the most interesting phenomena related to this model is gelation, the density dropping phenomenon. Indeed, the total density of particles $\rho_t = \sum_{i=1}^{\infty} iC_t(i)$ seems to be conserved since particles are neither created nor destroyed. However, for a broad class of kernels, it has been proved that ρ_t starts to decrease after a certain finite time. This is interpreted as an indication of the formation of gel, or an infinite size cluster [A1, A2, BH, HEZ, J1, J2, LT, vD, vZL, Zs, ZHE].

Gelation in the case $K(i, j) = ij$ is known to be equivalent to the emergence of a giant component in the random graph theory, a result which was initiated by Erdös–Rényi and extensively studied by many authors including Bollobás, Pittel, Janson *et al.* See [B, ER, P. JKLP].

We summarize some known results about gelation, referring to [A1] for a more detailed discussion.

(a) If $K(i, j) \leqslant (ij)^{1/2}$, there is no gelation [W, H].

(b) If $(ij)^{1/2+\varepsilon} \leqslant K(i, j)$ and $K(i, j) = o(ij)$, then gelation occurs [LT, J1, J2].

Also, it is believed that

(c) if there exist $\gamma > 2\alpha > 2$ such that $i^\alpha + j^\alpha \leqslant K(i, j) \leqslant (ij)^{\gamma/2}$, then instantaneous gelation occurs, i.e., ρ_t starts to decrease from the beginning [CC, vD, A1].

On the other hand, for the stochastic model, gelation does not seem to be defined clearly. However, for the case $\lim_{i+j \to \infty} K(i, j)/(ij) = 0$, a necessary and sufficient condition for gelation for the scaled ML process was given in the author's previous paper [J2]. It is natural to try to extend this condition to the general $K(i, j)$. In Section 1, we will define several different forms of gelation. In any event, it should be sufficient to say gelation occurs if all particles are swept into one huge cluster in a finite time.

The cluster sizes of the ML process were studied rigorously by Aldous and by the author. Aldous, in [A2], considered the kernel

$$K(i, j) = \frac{2(ij)^{\alpha}}{(i+j)^{\alpha} - i^{\alpha} - j^{\alpha}}, \qquad 1 < \alpha < 2$$

He showed that if $t < 1$, then the maximum cluster size is $o(n^{\varepsilon})$, for any $\varepsilon > 0$. His result seems to be related to a lower bound on the gelation time. In [J2], we proved implicitly that if $(ij)^{1/2+\varepsilon_1} \leqslant K(i, j)$ and $\lim_{i+j\to\infty} K(i, j)/(ij) = 0$, then, in a finite time, the maximum cluster size becomes larger than $O(n^{1-\varepsilon_2})$ for any $0 < \varepsilon_1 < \frac{1}{2}$, $0 < \varepsilon_2$. We also provided an upper bound of gelation time.

One interesting question is the relation between the microscopic picture and the macroscopic picture, or the relation between the ML process and the Smoluchowski coagulation equation. In [J2], we proved that if $\lim_{i+j\to\infty} K(i, j)/(ij) = 0$, then any weak limit of the scaled ML process solves the Smoluchowski coagulation equation even after gelation (for an extension to the continuous case, see Norris [N]). This is due to the relatively small size of the sol–gel interaction. However, if $\lim_{i+j\to\infty} ij/K(i, j) = 0$, then, since the sol–gel interaction is enormous, a lot of sol mass seems to be transfered directly to the gel in a short period of time. Therefore, in this case, it is natural to conjecture that any reasonable limit of the ML process will not solve the Smoluchowski coagulation equation after gelation. Also, since most kernels of this type seem to exhibit instantaneous gelation, it is believed that there is no time interval in which the macroscopic picture can be obtained from the microscopic picture.

Indeed, for the case $K(i, j) = (ij)^2$, it was conjectured by Domilovskii, Lushnikov and Piskunov in 1978 that instantaneous and complete gelation occurs, i.e., that all particles are swept into one huge cluster in an arbitrarily small time [DLP]. In 1985, this conjecture was extended by Spouge to the case $K(i, j) = (ij)^{\alpha}$, $\alpha > 1$ [Sp]. See also the supporting work of van Dongen [vD].

Our main result is the extension and resolution of the above conjecture. Indeed, we prove that if there is a function $\psi(i, j)$, increasing in both i and j such that $K(i, j) \geqslant ij\psi(i, j)$ for all i, j and if $\sum_{j=1}^{\infty} 1/(j\psi(j)) < \infty$, where $\psi(j) = \psi(1, j)$, then complete and instantaneous gelation occurs. (cf. Theorem 1 in Section 1.) As a consequence, we confirm that for such kernels, no non-zero solution of the Smoluchowski coagulation equation is a limit of the scaled ML process. This is one of the reasons our results are of interest. The main idea of the proof, so the main idea of this paper, is introduced in Section 1 after the statement of Conjecture 1.

One may argue that if the kernel $K(i, j)$ satisfies $\lim_{i+j \to \infty} ij/K(i, j) = 0$, then the model is unphysical; the reason being that the number of active sites on a cluster cannot increase faster than its size [vD]. Since the number of pairs of active sites between i and j-clusters is of order at most ij, we have $K(i, j) \leqslant kij$ for some $k < \infty$. However, we may consider higher order interactions, for example, the interaction between a pair of particles of an i-cluster and another pair of particles of a j-cluster. In this case, the number of active sites on an i-cluster is the number of pairs of particles in the cluster and, thus, is of order i^2 [M]. This argues for $K(i, j) \propto (ij)^2$. Furthermore kernels $K(i, j) = (i^2 j + ij^2)^{1/2} (i^{1/3} + j^{1/3})$ and $K(i, j) = (i^{1/3} + j^{1/3})^2 \, |i^{2/3} - j^{2/3}|$ are used as physical models; [A1, CC, DLP, vD, Z] the former describes a model for inertia and gravitational settling, while the latter describes a model for gravitationally distributed particles with a Maxwellian velocity distribution.

Besides, as van Dongen has indicated [vD], the above case has meaning and interest as a completion of the known cases $K(i, j) \leqslant Mij$. As we will see in the proof of Theorem 1, their mathematical structures are quite similar.

1. THE SCALED MARCUS–LUSHNIKOV PROCESS AND MAIN THEOREMS

In this section, we construct a sequence of finite state Markov chains (a scaled Marcus–Lushnikov process) associated with the rate constants $K(i, j)$, $i, j \geqslant 1$. In the nth Markov chain, there are n particles, each of size $1/n$, which coagulate to form clusters. These clusters coagulate among themselves at rates determined by $K(i, j)$. With this scaling, the Markov chains can be thought of as discrete, stochastic approximations to solutions of the Smoluchowski equations.

Remark. The reason for scaling the particle size to $1/n$ is to ensure that the total mass per unit volume is normalized to 1. This is consistent with the author's previous paper [J2], in which, by such scaling, we have the advantage of seeing that the configuration space is a compact subset of l_2 space.

Notation:

(a) Let $\mathbb{N} = \{0, 1, 2, ...\}$, $\mathbb{N}^+ = \{1, 2, 3, ...\}$.

(b) Let $E=\{\eta\in\mathbb{E}^{\mathbb{N}^+} : \eta(k)\geqslant 0$ for all k and $\sum_{k=1}^{\infty} k\eta(k)\leqslant 1\}$.

(c) Let $E_n=\{(1/n)\,\eta : \eta\in\mathbb{N}^{\mathbb{N}^+}, \sum_{k=1}^{\infty} k\eta(k)=n\}$.

(d) Let $\|\eta\|=\sum_{i=1}^{\infty} i\eta(i)$.

(e) Let $\|\eta\|_l=\sum_{i=l}^{\infty} i\eta(i)$.

(f) Let $l_\varepsilon(\eta)=\max\{l : \|\eta\|_l\geqslant\varepsilon\}$.

(g) $[\,\cdot\,]$ represents the largest integer function.

(h) Let $\{e_i\}_{i=1}^{\infty}$ be the basis of $\mathbb{R}^{\mathbb{N}^+}$, i.e., $e_i=(0, 0,..., 0, 1, 0, 0,...)$, where 1 is located in the ith coordinate.

Remark. Note that any $\eta\in E_n$ can be expressed by $\eta=\sum_{i=1}^{n}\eta(i)\,e_i$. Here, $(1/n)\,e_i$ can be interpreted as an i-cluster and the size of the particles which form the i-cluster is $1/n$.

Let $\{K(i, j)\}_{i,j=1}^{\infty}$ be a nonnegative sequence such that $K(i, j)=K(j, i)$. Let $\Delta_{ij}^n=(1/n)(e_{i+j}-e_i-e_j)$, and let $\delta_i^j=1$, if $i=j$, and 0, if $i\neq j$. Let X_t^n be the Markov chain on E_n with generator

$$Lf(\eta)=\frac{n}{2}\sum_{i+j\leqslant n}(f(\eta+\Delta_{i,j}^n)-f(\eta))\,K(i, j)\,\eta(i)\left(\eta(j)-\delta_i^j\,\frac{1}{n}\right) \tag{2}$$

We may describe the dynamics as follows: The process waits at state η for an exponentially distributed amount of time with parameter

$$\lambda^n(\eta)\doteq\frac{n}{2}\sum_{i+j\leqslant n}K(i, j)\,\eta(i)\left(\eta(j)-\delta_i^j\,\frac{1}{n}\right) \tag{3}$$

then jumps to state $\eta+\Delta_{ij}^n$ (or i and j cluster coagulate to form $i+j$ cluster) with probability

$$\frac{n}{2\lambda^n(\eta)}K(i, j)\,\eta(i)\left(\eta(j)-\delta_i^j\,\frac{1}{n}\right) \tag{4}$$

Since, for each n, the state space consists of finitely many points, i.e., $|E_n|<\infty$, there is a unique well defined pure jump process, say X_t^n on E_n for each n. We will call this sequence of processes $\{X_t^n\}_{n=1}^{\infty}$ the scaled Marcus–Lushnikov process, and we will denote it simply by X_t^n. In general, we assume that the initial configuration $X_0^n=\eta^n$, $\eta^n\in E_n$ satisfies $\eta^n(i)\to\eta_0(i)$ for all i and for some $\eta_0\in E$, where $\sum_{i=1}^{\infty} i\eta_0(i)=1$. In particular, unless stated otherwise, we assume a mono-disperse initial condition, i.e., $X_0^n=e_1$.

Before we state the main theorems, we will introduce several different definitions of gelation in the ML process. Note that, in a finite particle system, since the density is algebraically conserved, it does not make any sense to define gelation using density.

Suppose there is a subsequence $X_t^{n_k}$ of X_t^n which converges weakly to a stochastic process X_t. Then it has been proved in [J2] that for any fixed $t>0$, the following are equivalent.

I There exists a nondecreasing function $\phi: \mathbb{N}^+ \to \mathbb{N}^+$ such that $\lim_{n\to\infty} \phi(n)=\infty$, and there exists $\varepsilon>0$ such that

$$\limsup_{k\to\infty} P\{\|X_t^{n_k}\|_{\phi(n_k)} \geqslant \varepsilon\} > 0$$

II There exists $\varepsilon>0$ such that

$$P\{\|X_t\| \leqslant 1-\varepsilon\} > 0$$

With this equivalence in mind, regardless of the existence of a weak limit, let us define the stochastic gelation (sto-gel) time T_g of X^n as the infimum of all t satisfying the property **I**. If $T_g<\infty$, then we say that sto-gel occurs.

Another possible definition of gelation dealt with in [J2] is strong gelation, which indicates the formation of a cluster of order n size.

For $0<\alpha\leqslant 1$, let τ_n^α be the hitting time of a cluster size greater than or equal to αn, i.e.,

$$\tau_n^\alpha \doteq \inf\{t: X_t^n(k)>0 \text{ for some } k\geqslant \alpha n\}$$

(Note that if $\alpha<\beta$, then $\tau_n^\alpha \leqslant \tau_n^\beta$ a.s.)

Define the strong gelation time by

$$t_g^s = \inf\{t>0: \exists 0<\alpha\leqslant 1 \text{ such that } \limsup_{n\to\infty} P\{\tau_n^\alpha \leqslant t\}>0\}$$

We say that strong gelation occurs if $t_g^s<\infty$.

Finally, we say that complete gelation occurs, if

$$\lim_{n\to\infty} P\{\tau_n^1 \leqslant t\} = 1$$

for some $t<\infty$. It is clear that complete gelation implies strong gelation and strong gelation implies sto-gel.

Now, for any fixed $t>0$, define $G_n(t)$ by

$$G_n(t) = \left\{ X_t^n = \frac{1}{n} e_n \right\}$$

i.e., the event of hitting n-cluster by time t.

Theorem 1. Suppose there is a function $\psi(i, j)$, increasing in both i and j such that $K(i, j) \geqslant ij\psi(i, j)$ for all i, j. If $\sum_{j=1}^{\infty} 1/(j\psi(j)) < \infty$, where $\psi(j) = \psi(1, j)$, then for any $t > 0$.

$$\lim_{n \to \infty} P(G_n(t)) = 1$$

i.e., complete and instantaneous gelation occurs.

Remark. For example, if $K(i, j) \geqslant ij(\log(i+1) \log(j+1))^{\alpha}$, $\alpha > 1$, then the theorem implies that complete instantaneous gelation occurs. It also implies that, for any k and $t > 0$, $X_t^n(k) \to 0$ in probability.

Instead of the mono-disperse initial condition, we may assume that there is a gel mass initially. In this case, we can weaken the condition on $K(i, j)$ as follows.

Theorem 2. Suppose $\lim_{i+j \to \infty} ij/K(i, j) = 0$ and $X_0^n = \eta_0^n \in E_n$. If there is a function $\phi(n) \to \infty$, such that

$$\lim_{n \to \infty} \sum_{i \geqslant \phi(n)} i\eta_0^n(i) > \varepsilon$$

for some $\varepsilon > 0$, then complete and instantaneous gelation occurs.

One natural question arises, then, about the mono-disperse case in which $\lim_{i+j \to \infty} ij/K(i, j) = 0$ and the condition in Theorem 1 fails. In this case, we conjecture the followings and we will discuss the motivation after the proof of Theorem 2.

Conjecture 1. Assume the mono-disperse initial condition. If $\lim_{i+j \to \infty} ij/K(i, j) = 0$ and $\sum_{j=1}^{\infty} 1/jK(j) = \infty$, where $K(j) = K(1, j)$, then the stochastic gelation time T_g satisfies $0 < T_g < \infty$. Moreover, for any $t > T_g$,

$$\lim_{n \to \infty} P(G_n(t)) = 1$$

i.e., the gelation is not instantaneous but complete.

This conjecture indicates the existence of a critical sol state. If we add gel mass (for example, an n order cluster) at any time $t < T_g$, no matter how small the size of the cluster is, the whole system gels instantaneously and completely due to the infinite size of the sol-gel interaction (see Theorem 2). Perhaps this metastable state is reminiscent of the supercooled liquids [AD, D].

Our proof of Theorem 1 which is the main idea of this paper is based on showing that almost all sample paths exhibit high speed mass transfers. More precisely, we prove the following three steps.

(a) Step I (Proposition 1): For any fixed l, there is a high probability that it will take a short time for a small amount of mass to be transferred to clusters of size $\geqslant l$.

(b) Step II (Proposition 2): There exists a function ϕ on $\mathbb{N}^+$ such that $\phi(n) \to \infty$ and such that there is a high probability that, for large l, it will take a short time for the mass in clusters of size $\geqslant l$ to be transferred to clusters of size $\geqslant \phi(n)$.

(c) Step III (Proposition 3): If (b) occurs, since the sol-gel interaction is enormous there is a high probability that in a short time all mass will be swept into the gel.

To prove the above steps, we depend heavily on the stochastic dominance of jump processes similar to the proofs in [J2]. These arguments can be justified by the coupling method and we refer to, for example, Chap. 5 of Lindvall [Lin].

The remainder of this section is devoted to evoking some conceptual aspects of gel formation of the ML process and to summarizing related works done mostly in [J2].

To classify gelation kernels, it seems useful to consider the rate of interaction per unit mass between i and j clusters,

$$\frac{K(i, j)\, \eta(i)\, \eta(j)}{i\eta(i)\; j\eta(j)} = \frac{K(i, j)}{ij}$$

Letting $j \to \infty$, we have the rate of interaction per unit mass between sol and gel as follows.

Definition 1. For the kernel $K(i, j)$, define

$$\text{(a)}\quad I^+(i) = \limsup_{j \to \infty} \frac{K(i, j)}{ij}, \qquad i = 1, 2,...,$$

$$\text{(b)}\quad I^-(i) = \liminf_{j \to \infty} \frac{k(i, j)}{ij}, \qquad i = 1, 2,...$$

To summarize the known results in terms of the above definition:

(a) If $I^-(i) = 0$ for all i, then any weak limit of the scaled Marcus–Lushnikov process solves the integral version of the Smoluchowski coagulation equation. In addition, if $\lim_{i+j \to \infty} (ij)^\alpha / K(i, j) = 0$, for some $\alpha > \frac{1}{2}$, then stochastic gelation occurs [J2].

(b) If $0 < \varepsilon \leqslant I^-(i) \leqslant I^+(i) \leqslant M < \infty$ for all i, then strong gelation occurs [J2].

Finally, from Theorem 1 of this paper,

(c) if $I^-(i) = \infty$ for all i, then under the same conditions as those established in Theorem 1, complete and instantaneous gelation occurs. Also, no nonzero solution of the Smoluchowski coagulation equation is a limit of the ML process.

2. PROOF OF THEOREM 1

In this section, we prove Theorem 1 by showing three propositions. For any $k \leqslant n$ and $\delta > 0$, let

$$A_\delta^k \doteq \left\{ \eta \in E : \sum_{i=k}^{\infty} i\eta(i) \geqslant \delta \right\}$$

Proposition 1. For any fixed l and $\varepsilon_1, \varepsilon_2 > 0$, under the same hypothesis of Theorem 1, there exists $\delta > 0$, such that

$$P\{X_t^n \in A_\delta^l\} > 1 - \varepsilon_1$$

for all $t < \varepsilon_2$ and for all n large.

Proof. Proof will be done by an induction on l. First, the case $l = 1$ is clear. Assume the induction hypothesis for l. That is, for every $\varepsilon_1, \varepsilon_2 > 0$ there exists δ_1 such that

$$P\{X_t^n \in A_{\delta_1}^l\} > 1 - \frac{\varepsilon_1}{4}$$

for all $t \geqslant \varepsilon_2/2$. Note that $K(i, j) \geqslant cij$ for $c = \psi(1, 1)$. Without loss of generality, assume $\delta_1 < \frac{1}{2}$ and choose $\delta < (c\varepsilon_2\delta_1)/16 \wedge \delta_1/2$. Let

$$J_1 = \{(i, j) : i \leqslant l \leqslant j \text{ or } j \leqslant l \leqslant i\}$$

$$J_2 = \{(i, j) \in J_1 : i \leqslant j\}$$

For any $\eta \in A_{\delta_1}^l \backslash A_\delta^{l+1}$, let $\lambda_l(\eta)$ be the rate of jumps including only the clusters of size $\leqslant l$ and those of size $\geqslant l$, i.e.,

$$\lambda_l(\eta) = \frac{n}{2} \sum_{(i, j) \in J_1} K(i, j)\, \eta(i) \left(\eta(j) - \frac{1}{n} \delta_i^j \right)$$

Then

$$\lambda_l(\eta) \geqslant \frac{n}{2} \sum_{(i,\,j) \in J_2} K(i,\,j)\,\eta(i) \left(\eta(j) - \frac{1}{n}\,\delta_i^j\right) \tag{5}$$

Since $\eta \in A^l_{\delta_1} \backslash A^{l+1}_{\delta}$ implies $\sum_{i \leqslant l} i\eta(i) \geqslant 1-\delta$ and $\sum_{i \geqslant l} i\eta(i) \geqslant \delta_1$, and since $\eta(l) \leqslant \sum_{i=1}^n i\eta(i) = 1$, from (5),

$$\begin{aligned}
\lambda_l(\eta) &\geqslant \frac{n}{2} \sum_{(i,\,j) \in J_2} K(i,\,j)\,\eta(i)\,\eta(j) - \frac{1}{2}\,K(l,\,l)\,\eta(l)\\
&\geqslant \frac{cn}{2} \sum_{(i,\,j) \in J_2} ij\eta(i)\,\eta(j) - K(l,\,l)\,\eta(l)\\
&\geqslant \frac{cn}{2}\,\delta_1(1-\delta) - K(l,\,l) \geqslant \frac{c\delta_1 n}{4}
\end{aligned} \tag{6}$$

for large n.

Let

$$\mu = \frac{c\delta_1 n}{4}$$

$$\lambda_{ij}(\eta) = \frac{n}{2}\,K(i,\,j)\,\eta(i) \left(\eta(j) - \frac{1}{n}\,\delta_i^j\right)$$

Let Y^n_t be a Poisson process with intensity μ, defined on a common probability space with X^n_t. Note that for any $\eta \in A^l_{\delta_1} \backslash A^{l+1}_{\delta}$, since $\sum_{(i,\,j) \in J_1} \lambda_{ij}(\eta) \geqslant \mu$ from (5) and (6), we can find $\{\mu_{ij}\}_{(i,\,j) \in J_1}$ such that $\lambda_{ij}(\eta) \geqslant \mu_{ij}$ for all $(i,\,j) \in J_1$ and $\sum_{(i,\,j) \in J_1} \mu_{ij} = \mu$. Let

$$T^{l+1}_{\delta} = \inf\{t > 0 : X^n_t \in A^{l+1}_{\delta}\}$$

Consider the coupled process

$$(X^n,\,Y^n)_{t \wedge T^{l+1}_{\delta}}$$

whose jump rates are given by

$$(\eta,\,k) \to \begin{cases} (\eta + \Delta^n_{ij},\,k) \text{ with intensity } \lambda_{ij} & \text{if} \quad (i,\,j) \in J_1^c\\ (\eta + \Delta^n_{ij},\,k) \text{ with intensity } \lambda_{ij} - \mu_{ij} & \text{if} \quad (i,\,j) \in J_1\\ (\eta + \Delta^n_{ij},\,k+1) \text{ with intensity } \mu_{ij} & \text{if} \quad (i,\,j) \in J_1 \end{cases}$$

This coupling shows that at any jump of Y_t^n, X_t^n makes a jump including i and j clusters such that $i \leqslant l \leqslant j$.

Let

$$l_0 = \left[\frac{c\varepsilon_2 \delta_1 n}{16} \right]$$

To prove that it takes less than $\varepsilon_2/2$ time for Y_t^n to make l_0 jumps, notice that $\{ Y^n_{\varepsilon_2/2} < l_0 \}$ is a tail of the Poisson process, since

$$\frac{\varepsilon_2}{2} > \frac{\varepsilon_2}{4} \geqslant l_0 \mu$$

and

$$E Y^n_{l_0 \mu} = l_0$$

Therefore,

$$P\{ Y^n_{\varepsilon_2/2} < l_0 \} \leqslant \frac{\varepsilon_1}{4} \tag{7}$$

for large n.

Now, if $X^n_{\varepsilon_2/2}$ contains at least l_0 jumps including clusters of size $\leqslant l$ and those of size $\geqslant l$, then $X^n_{\varepsilon_2/2} \in A_\delta^{l+1}$. To see this, for any $\eta \in A^l_{\delta_1} \backslash A^{l+1}_\delta$, $\eta' \in E_n$, suppose there exist $\eta_1, \eta_2,..., \eta_M$ such that $\eta_1 = \eta$, $\eta_M = \eta'$ and $\eta_{m+1} = \eta_m + \Delta^n_{i_m, j_m}$.

Let

$$B = \{ m \colon i_m \leqslant l \leqslant j_m \}$$

If $|B| \geqslant l_0$, then

$$\begin{aligned} \|\eta'\|_{l+1} &\geqslant \left\| \eta + \sum_{m \in B} \Delta^n_{i_m, j_m} \right\|_{l+1} \\ &\geqslant |B| \; \|\Delta^n_{i_m, j_m}\|_{l+1} \\ &\geqslant \frac{l_0}{n} \\ &= \left[\frac{c\varepsilon_2 \delta_1 n}{16} \right] \frac{1}{n} \\ &\geqslant \delta \end{aligned}$$

for large n, which implies $\eta' \in A_\delta^{l+1}$. Therefore, for any $\eta_0 \in A_{\delta_1}^l \backslash A_\delta^{l+1}$, from (7),

$$\begin{aligned} P\{X_{\varepsilon_2/2}^n \in A_\delta^{l+1} \mid X_0^n = \eta_0\} &\geqslant P\{Y_{\varepsilon_2/2}^n \geqslant l_0 \mid X_0^n = \eta_0\} \\ &\geqslant 1 - P\{Y_{\varepsilon_2/2}^n < l_0 \mid X_0^n = \eta_0\} \\ &\geqslant 1 - \frac{\varepsilon_1}{4} \end{aligned} \tag{8}$$

Since $\{X_t^n \in A_\delta^{l+1}\}$ is an increasing event in t, and from (8), for all $t \geqslant \varepsilon_2$,

$$\begin{aligned} P\{X_t^n \in A_\delta^{l+1}\} &\geqslant P\{X_{\varepsilon_2}^n \in A_\delta^{l+1}\} \\ &\geqslant P\{X_{\varepsilon_2}^n \in A_\delta^{l+1}, X_{\varepsilon_2/2}^n \in A_{\delta_1}^l\} \\ &= \sum_{\eta \in A_{\delta_1}^l} P\{X_{\varepsilon_2/2}^n = \eta\} \ P\{X_{\varepsilon_2}^n \in A_\delta^{l+1} \mid X_{\varepsilon_2/2}^n = \eta\} \\ &= \sum_{\eta \in A_{\delta_1}^l} P\{X_{\varepsilon_2}^n = \eta\} \ P\{X_{\varepsilon_2/2}^n \in A_\delta^{l+1} \mid X_0^n = \eta\} \\ &\geqslant \sum_{\eta \in A_{\delta_1}^l} P\{X_{\varepsilon_2/2}^n = \eta\} \left(1 - \frac{\varepsilon_1}{4}\right) \\ &= P\{X_{\varepsilon_2/2}^n \in A_{\delta_1}^l\} \left(1 - \frac{\varepsilon_1}{4}\right) \\ &= \left(1 - \frac{\varepsilon_1}{4}\right)^2 \geqslant 1 - \varepsilon_1 \quad \blacksquare \end{aligned} \tag{9}$$

Now, recall that

$$l_\delta(\eta) = \max\{l : \|\eta\|_l \geqslant \delta\}$$

We will prove Lemma 1, which gives a sufficient condition, in terms of jumps of certain types, for $\eta \in A_\delta^l$ to move to A_δ^{2l}.

Lemma 1. Let $l < \delta n$, $0 < \delta < 1$, and $\eta_0 \in A_\delta^l$. Suppose there exist $\eta_1, \eta_2, ..., \eta_M$ such that

$$\eta_{m+1} = \eta_m + \Delta_{i_m, j_m}^n$$

for some i_m, j_m, and for $m = 0, 1, ..., M-1$. Let

$$G = \{m : i_m \leqslant l_\delta(\eta_m) \leqslant j_m\}$$

If $|G| \geqslant 3\delta n$ then

$$\eta_M \in A_\delta^{2l}$$

Proof. Let

$$G_1 = \{m \in G : j_m \geqslant 2l\}, \qquad G_2 = G \backslash G_1$$

First, to show $|G_2| \leqslant 2\delta n$, let

$$H_i = \{m \in G_2 : j_m = l + i\}, \qquad i = 0, 1, ..., l-1$$

Then $H_i \cap H_j = \phi$, for $i \neq j$ and $G_2 = \bigcup_{i=0}^{l-1} H_i$.

Claim:

$$|H_i| \leqslant \frac{\delta n}{l+i} + 1$$

Proof. Suppose $|H_i| > \delta n/(l+i) + 1$ and rewrite the elements of H_i in an increasing order, say $m_0, m_1, ..., m_{(r_0-1)}, m_{r_0}$. Then $r_0 > \delta n/(l+i) + 1$ and for any m_r, $0 \leqslant r < r_0$, since $i_{m_r} + j_{m_r} = i_{m_r} + l + i \geqslant l + i + 1$,

$$\begin{aligned}
\|\eta_{m_{(r+1)}}\|_{l+i+1} &= \|\eta_{m_r}\|_{l+i+1} + \|\Delta^n_{i_{m_r}, j_{m_r}}\|_{l+i+1} \\
&= \|\eta_{m_r}\|_{l+i+1} + \frac{1}{n}(i_{m_r} + j_{m_r}) \\
&\geqslant \|\eta_{m_r}\|_{l+i+1} + \frac{j_{m_r}}{n} \\
&= \|\eta_{m_r}\|_{l+i+1} + \frac{l+i}{n}
\end{aligned}$$

Therefore,

$$\begin{aligned}
\|\eta_{m_{(r_0-1)}}\|_{l+i+1} &\geqslant \|\eta_{m_0}\|_{l+i+1} + \frac{l+i}{n}(r_0 - 1) \\
&> \frac{l+i}{n} \frac{\delta n}{l+i} = \delta
\end{aligned}$$

Hence, $\eta_{m_{(r_0-1)}} \in A_\delta^{l+i+1}$ and for all $m > m_{(r_0-1)}$, $\eta_m \in A_\delta^{l+i+1}$.

Since $m_{r_0} - 1 \geqslant m_{(r_0-1)}$, $\eta_{(m_{r_0}-1)} \in A_\delta^{l+i+1}$, which implies

$$l_\delta(\eta_{(m_{r_0}-1)}) \geqslant l+i+1$$

But this is a contradiction to the fact that $j_{m_{r_0}} = l+i$, since

$$j_{m_{r_0}} \geqslant l_\delta(\eta_{(m_{r_0}-1)}) \geqslant l+i+1$$

Therefore, since $l < \delta n$,

$$|G_2| = \sum_{i=0}^{l-1} |H_i| \leqslant \sum_{i=0}^{l-1} \left(\frac{\delta n}{l+i} + 1 \right) \leqslant \delta n + l \leqslant 2\delta n$$

for large n, and the claim is proved.

Now, $|G_1| \geqslant \delta n$, since $|G_2| \leqslant 2\delta n$.

For any $m \in G_1$, since $j_{m-1} \geqslant 2l$, $i_{m-1} \leqslant l$,

$$\begin{aligned} \|\eta_m\|_{2l} &= \|\eta_{m-1} + \Delta^n_{i_{m-1}, j_{m-1}}\|_{2l} \\ &= \|\eta_{m-1}\|_{2l} + \frac{i_{m-1}}{n} \\ &\geqslant \|\eta_{m-1}\|_{2l} + \frac{1}{n} \end{aligned}$$

Moreover, since $\|\eta_m\|_{2l}$ is nondecreasing in m,

$$\|\eta_M\|_{2l} \geqslant \|\eta_0\|_{2l} + |G_1| \, \frac{1}{n} \geqslant \delta$$

Therefore,

$$\eta_M \in A_\delta^{2l} \quad ∎$$

Proposition 2. Let X_t^n be the ML process with $X_0^n = \eta_0 \in E_n$. Consider a function $\phi\colon \mathbb{N}^+ \to \mathbb{N}^+$ such that $\phi(n) \nearrow \infty$ and $\phi(n)\,\psi(4\phi(n), 4\phi(n)) = o(n)$. (Note that such function exists.) Then for any $\varepsilon_1, \varepsilon_2 > 0$ and $\delta < \frac{1}{2}$, under the same hypothesis of Theorem 1, there exists l_0 independent of n such that $\eta_0 \in A_\delta^{l_0}$ implies

$$P\{X_t^n \in A_\delta^{\phi(n)}\} > 1 - \varepsilon_2$$

for all $t \geqslant \varepsilon_1$.

Proof. First suppose $\eta_0 \in A_\delta^l \backslash A_\delta^{2l}$, for some $l < 2\phi(n)$, and let us prove that if

$$T^l = \inf\{t > 0 : X_t^n \in A_\delta^{2l}\}$$

then

$$P\left\{T^l \geqslant \frac{25}{\psi(l)}\right\} \leqslant \frac{C}{n}$$

for some constant $C = C(\delta)$ independent of l and n.

For any $\eta \in A_\delta^l \backslash A_\delta^{2l}$, let

$$J_1 = \{(i, j)\colon i \leqslant l_\delta(\eta) \leqslant j \text{ or } j \leqslant l_\delta(\eta) \leqslant i\}$$

$$J_2 = \{(i, j) \in J_1 : i \leqslant j\}$$

and let $\lambda_\delta(\eta)$ be the rate of jumps between clusters of size $\leqslant l_\delta(\eta)$ and those of size $\geqslant l_\delta(\eta)$, i.e.,

$$\begin{aligned} \lambda_\delta(\eta) &= \frac{n}{2} \sum_{(i,\, j) \in J_1} K(i, j)\, \eta(i) \left(\eta(j) - \frac{\delta_i^j}{n}\right) \\ &\geqslant \frac{n}{2} \sum_{(i,\, j) \in J_2} K(i, j)\, \eta(i) \left(\eta(j) - \frac{\delta_i^j}{n}\right) \end{aligned} \tag{10}$$

Note that for any $\eta \in A_\delta^l \backslash A_\delta^{2l}$, $l \leqslant l_\delta(\eta) < 2l$ and

$$\begin{aligned} l_\delta(\eta)\, \psi(l_\delta(\eta), l_\delta(\eta)) &\leqslant 2l\psi(2l, 2l) \\ &\leqslant 4\phi(n)\, \psi(4\phi(n), 4\phi(n)) \\ &= o(n) \end{aligned}$$

Therefore, from (10), since $k\eta(k) \leqslant \sum_i i\eta(i) = 1$, for any k,

$$\begin{aligned} \lambda_\delta(\eta) &\geqslant \frac{n}{2} \sum_{(i,\, j) \in J_2} i\eta(i)\, j\eta(j)\, \psi(i, j) - \frac{1}{2} (l_\delta(\eta))^2\, \psi(l_\delta(\eta), l_\delta(\eta))\, \eta(l_\delta(\eta)) \\ &\geqslant \frac{n}{2}\, \delta(1-\delta)\, \psi(l_\delta(\eta)) - l_\delta(\eta)\, \psi(l_\delta(\eta), l_\delta(\eta)) \\ &\geqslant \frac{\delta n \psi(l)}{4} \end{aligned} \tag{11}$$

for large n.

Let

$$\mu = \frac{\delta n \psi(l)}{4}$$

$$\lambda_{ij}(\eta) = \frac{n}{2} K(i, j)\, \eta(i) \left(\eta(j) - \frac{1}{n} \delta_i^j \right)$$

Similarly to Proposition 1, let Y_t^n be a Poisson process with intensity μ, defined on a common probability space with X_t^n. Note that for any $\eta \in A_\delta^l \setminus A_\delta^{2l}$, since $\sum_{(i, j) \in J_1} \lambda_{ij}(\eta) \geq \mu$, there exists $\{\mu_{ij}\}_{(i, j) \in J_1}$ such that $\lambda_{ij}(\eta) \geq \mu_{ij}$ for all $(i, j) \in J_1$ and $\sum_{(i, j) \in J_1} \mu_{ij} = \mu$. Then the coupled process $(X^n, Y^n)_{t \wedge T^l}$ defined as for Proposition 1 shows that at any jump time of Y_t^n, X_t^n makes jumps including i and j clusters such that $i \leqslant l_\delta(\eta) \leqslant j$.

Let

$$l_0 = [3\delta n] + 1$$

Define $\{T_k\}$, the jump time of Y^n, successively by

$$T_k = \inf\{t > 0 : Y^n_{(T_{k-1}+t)-} \neq Y^n_{(T_{k-1}+t)}\}$$

then each T_k is an exponential random variable with parameter μ.

Let

$$T = T_1 + T_2 + \cdots T_{l_0}$$

then

$$E(T) = \frac{4 l_0}{\delta n \psi(l)}$$

$$Var(T) = \frac{16 l_0}{(\delta n \psi(l))^2}$$

Therefore, by the Chebyshev inequality,

$$P\left\{T \geqslant \frac{8 l_0}{\delta n \psi(l)}\right\} \leqslant \left(\frac{\delta n \psi(l)}{4 l_0}\right)^2 \frac{16 l_0}{(\delta n \psi(l))^2}$$

$$= \frac{1}{l_0} \leqslant \frac{C}{n} \tag{12}$$

for some constant C independent of l and n.

From Lemma 1, we see that if X_t^n contains at least l_0 jumps between clusters of size less than or equal to and greater than or equal to $l_\delta(X_{\tau-}^n)$, where τ is the jump time, then $X_t^n \in A_\delta^{2l}$.

Therefore, since

$$\frac{8l_0}{\delta n\psi(l)} \leqslant \frac{25}{\psi(l)}$$

we have, from (12),

$$P\left\{T^l \geqslant \frac{25}{\psi(l)}\right\} \leqslant P\left\{T \geqslant \frac{8l_0}{\delta n\psi(l)}\right\} \leqslant \frac{C}{n} \tag{13}$$

for large n.

Next, for any $l \leqslant 2\phi(n)$ of the type $l = 2^r l_0$ for some l_0, let $a_r = 25/\psi(l)$, $r = 0, 1, \ldots$. Then from the above argument, for any $\eta_0 \in A_\delta^l$,

$$P\{X_{a_r}^n \in A_\delta^{2^{r+1}l_0} \mid X_0^n = \eta_0\} \geqslant P\{T^l < a_r\} \geqslant 1 - \frac{C}{n} \tag{14}$$

Now, consider l_0 large enough so that

$$\sum_{k=0}^{\infty} \frac{25}{\psi(2^k l_0)} < \varepsilon_1$$

Indeed, there is such an l_0 since

$$\begin{aligned}\sum_{k=0}^{\infty} \frac{1}{\psi(2^k l_0)} &= \sum_{k=0}^{\infty} \frac{2^k}{2^k\psi(2^k l_0)} \\ &\leqslant 2 \sum_{k=1}^{\infty} \sum_{i=2^{k-1}l_0+1}^{2^k l_0} \frac{1}{i\psi(i)} + \frac{1}{\psi(l_0)} \\ &\leqslant 2 \sum_{i=1}^{\infty} \frac{1}{i\psi(i)} + \frac{1}{\psi(l_0)} < \infty\end{aligned}$$

Let $b_r = \sum_{k=0}^{r-1} a_r$, and let $s = s(n)$ be the unique integer satisfying

$$2^{s-1} l_0 < \phi(n) \leqslant 2^s l_0$$

Suppose $\eta_0 \in A_\delta^{l_0}$, then, from (14),

$$\begin{aligned} P\{X_{b_s}^n \in A_\delta^{\phi(n)}\} &\geqslant P\{X_{b_s}^n \in A_\delta^{\phi(n)}, X_{b_r}^n \in A_\delta^{2^r l_0}, r = 1,..., s-1\} \\ &= \sum_{\eta_1 \in A_\delta^{2l_0}} \sum_{\eta_2 \in A_\delta^{4l_0}} \cdots \sum_{\eta_{s-1} \in A_\delta^{2^{s-1}l_0}} P\{X_{b_1}^n = \eta_1\} \\ &\quad \cdot P\{X_{b_2} = \eta_2 \mid X_{b_1}^n = \eta_1\} \cdots P\{X_{b_s} \in A_\delta^{\phi(n)} \mid X_{b_{s-1}}^n = \eta_{s-1}\} \\ &= \sum_{\eta_1 \in A_\delta^{2l_0}} \sum_{\eta_2 \in A_\delta^{4l_0}} \cdots \sum_{\eta_{s-1} \in A_\delta^{2^{s-1}l_0}} P\{X_{a_0}^n = \eta_1\} \\ &\quad \cdot P\{X_{a_1} = \eta_2 \mid X_0^n = \eta_1\} \cdots P\{X_{a_{s-1}} \in A_\delta^{\phi(n)} \mid X_0^n = \eta_{s-1}\} \\ &\geqslant \left(1 - \frac{C}{n}\right)^s \\ &= 1 - o(1) \end{aligned} \tag{15}$$

since $s = o(n)$.

Since $b_s < \varepsilon_1$,

$$P\{X_t \in A_\delta^{\phi(n)}\} > 1 - \varepsilon_2$$

for all $t \geqslant \varepsilon_1$ and for all large n. ∎

For any cadlag sample path ω of X_t^n, let $D_n(t)$ be the set of times in which coagulation occurs, i.e., let

$$D_n(t, \omega) \doteq \{s \leqslant t : \omega(s) = \omega(s-) + \Delta_{ij}^n \text{ for some } i, j\}$$

then the following lemma shows that before n jumps occur, X_t^n forms an n cluster.

Lemma 2. Suppose $\omega(0) = e_1$, then $|D_n(t, \omega)| = n - 1$ iff $\omega(t) = (1/n)\, e_n$.

Proof. See Lemma 5.3 in [J2].

Proposition 3. Suppose $\phi(n) \nearrow \infty$ such that $\phi(n) \leqslant n$. Let X_t^n be the scaled ML Process with $X_0^n = \eta_0 \in A_\delta^{\phi(n)}$, $\delta > 0$ for all n then for any $\varepsilon_1, \varepsilon_2 > 0$, under the same hypothesis of Theorem 1, there exists $N < \infty$ such that

$$P\left\{X_t = \frac{1}{n} e_n\right\} > 1 - \varepsilon_2$$

for all $t \geqslant \varepsilon_1$ and for all $n > N$.

Proof. for $\frac{1}{2} < \alpha < 1$ let

$$T = \inf\{t > 0 : X_t^n(i) > 0, \text{ for some } i \geqslant \alpha n\}$$

For any $\eta \in A_\delta^{\phi(n)} \backslash B_\alpha$, where

$$B_\alpha = \{\eta \in E_n : \eta(i) > 0 \text{ for some } i \geqslant \alpha n\}$$

consider the rate of total jumps $\lambda^n(\eta)$. From (3)

$$\begin{aligned}
\lambda^n(\eta) &= \frac{n}{2} \sum_{i,\, j} K(i, j)\, \eta(i) \left(\eta(j) - \frac{\delta_i^j}{n}\right) \\
&\geqslant \frac{n}{2} \sum_{i,\, \phi(n) \leqslant j} ij\eta(i) \left(\eta(j) - \frac{\delta_i^j}{n}\right) \psi(i, j) \\
&\geqslant \frac{n\psi(\phi(n))}{2} \left(\sum_{i,\, \phi(n) \leqslant j} ij\eta(i)\, \eta(j) - \sum_{\phi(n) \leqslant i \leqslant \alpha n} \frac{i}{n}\, i\eta(i) \right) \\
&\geqslant \frac{n\psi(\phi(n))}{2} \left(\sum_{\phi(n) \leqslant j \leqslant \alpha n} j\eta(j) - \alpha \sum_{\phi(n) \leqslant i \leqslant \alpha n} i\eta(i) \right) \\
&\geqslant \frac{n\psi(\phi(n))}{2} (1 - \alpha)\, \delta
\end{aligned} \tag{16}$$

for large n.

Let

$$\mu = \frac{n\psi(\phi(n))}{2} (1 - \alpha)\, \delta$$

Consider the Poisson process Y_t^n with parameter μ. By a comparison using a coupling method similar to those in Propositions 1 and 2, we see that X_t^n has more jumps than Y_t^n for any $t > 0$. Therefore, by Lemma 2, it is easy to see that

$$P\left\{T \geqslant \frac{2}{\psi(\phi(n))(1 - \alpha)\, \delta}\right\} \leqslant \frac{C}{n}$$

for some constant C.

Now, for any $\eta \in B_\alpha$, the total jump rate

$$\begin{aligned}\lambda^n(\eta) &= \frac{n}{2} \sum_{i,j} K(i,j)\, \eta(i) \left(\eta(j) - \frac{\delta_i^j}{n}\right) \\ &\geqslant \frac{n}{2} \sum_{i,\, \alpha n \leqslant j} ij\eta(i) \left(\eta(j) - \frac{\delta_i^j}{n}\right) \psi(i,j) \\ &\geqslant \frac{n\psi(\phi(n))}{2} \sum_{i,\, \alpha n \leqslant j} ij\eta(i)\, \eta(j) \\ &\geqslant \frac{n\alpha\psi(\phi(n))}{2} \end{aligned} \tag{17}$$

for large n.

Therefore, again by a similar argument, if we let

$$T = \inf\left\{t > 0 : X_t^n = \frac{1}{n} e_n\right\}$$

then from the fact that $\psi(l) = \psi(1, l) \to \infty$, as $l \to \infty$, we get Proposition 3. ∎

Proof of Theorem 1. Proof of the theorem is now a simple combination of Propositions 1, 2, and 3 with a conditional probability setup. ∎

Proof of Theorem 2. It is easy to see that for any $\eta \in E_n$ the total jump rate $\lambda^n(\eta)$ satisfies that $n/\lambda^n(\eta) = o(1)$. Therefore, similarly to Proposition 3, we have Theorem 2. ∎

About the Conjecture 1. Similarly to Proposition 3, consider two different cases such that $\eta \in B_\alpha$ and $\eta \in B_\alpha^c$. In both cases it is easy to see that the total jump rate

$$\lambda^n(\eta) \geqslant cn$$

for some constant $c > 0$. Therefore, from Lemma 2 and since it takes, with high probability, uniformly finite time to make $n-1$ jumps, we have

$$P\{T_g < \infty\} = 1 - o(1)$$

On the other hand, the condition $\sum_{i,j} 1/K(i,j) < \infty$ seems to be crucial to create a small amount of gel in an arbitrarily small time (Proposition 2). In other words, if $\sum_{i,j} 1/K(i,j) = \infty$, then it seems to take strictly positive amount of time to form a gel.

However, from Theorem 2, we see that, as soon as it forms a gel, no matter how small it is, all clusters are swept into the gel due to the infinite size of the sol-gel interaction.

ACKNOWLEDGMENTS

I would like to thank my thesis advisor, P. March, for showing me mathematical ideas and insights, and for his encouragement. A lot of ideas in this paper are extensions of those in my thesis. I thank D. Aldous for his consistent interest. I also thank unknown referees for their valuable suggestions and comments.

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