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Dynamics of a rate equation describing cluster-size evolution

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Abstract

In this paper, we show dynamics of Smoluchowski's rate equation which has been widely applied to studies of aggregation processes (i.e., the evolution of cluster-size distribution) in physics. We introduce dissociation in the rate equation while dissociation is neglected in previous works. We prove the positiveness of solutions of the equation, which is a basic guarantee for the effectiveness of the model since the possibility that some solution may be negative is excluded. For the case of cluster coalesce without dissociation, we show both the equilibrium uniqueness and the equilibrium stability under the condition that the monomer deposition stops. For the case that clusters evolve with dissociation and there is no monomer deposition, we show the equilibrium uniqueness and prove the equilibrium stability if the maximum cluster size is not larger than three while we show the equilibrium stability by numerical simulations if the maximum size is larger than three.

Keywords: Equilibrium; Global stability; Cluster coalesce; Rate equation

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1. Introduction

In the growth processes of thin films by deposition techniques, clusters of different sizes evolve dynamically with the passage of time [1–3]. The classic model for characterizing cluster growth processes is the rate-equation approach developed by Smoluchowski [4]. In the rate equation, the evolution of the cluster-size distribution is described by the number and size of clusters where geometry is neglected. While Smoluchowski's rate-equation approach has provided powerful tools for studying aggregation processes [1–3], it is recognized to be nice in characterizing both agent-based coalition formation in electronic markets [5,6] and agent-based load balancing in grid computing [7–12]. While dissociation is neglected in the equation in previous works [1–4], it is considered in this work. In this paper, we focus on the rate equation describing the cluster evolution.

The mechanism in the evolution of cluster-size distribution is as follows [1,2]. On a two-dimensional smooth surface, monomers are deposited randomly. Each monomer on the smooth surface moves randomly. When a monomer meets another monomer, they may be "frozen" and form a cluster of size two. When a monomer meets an existing cluster of size s ($s > 1$), it may join the cluster and form a cluster of size $s + 1$. Inversely, a monomer in an existing cluster may also leave the cluster and join other clusters. Therefore, monomers' behavior can be focused on two elements: leaving and joining. Both the attachment rate and the detachment rate will determine the global dynamic behavior of the system. Based on the quantities of the number and size of clusters in the system, [1,2] described the evolution of the cluster-size distribution by the rate equation. In [1,2], a detailed study of the evolution, scaling and percolation of clusters were presented and compared with the results obtained using the rate equation.

Since solutions of the rate equation denote the quantities of clusters with different sizes, they should not be negative with the passage of time. In fact, suppose there is a counterexample where some component of a solution is negative at some time, then the model in [1,2] would be not effective since the number of clusters must not be negative. While experiments in [1,2] showed the convergence of cluster-size distributions, the stability of solutions of the rate equation is not studied. Hence, the primary problems we tackle in this paper are:

(i) showing the positiveness of solutions of the rate equation and
(ii) proving both the equilibrium uniqueness and the equilibrium stability.

In this paper, we show dynamics of the rate equation. We introduce dissociation in the rate equation while dissociation is neglected in previous works. We prove the positiveness of solutions of the equation, which is a basic guarantee for the effectiveness of the model since the possibility that some solution may be negative is excluded. For the cluster coalesce without dissociation, we show both the equilibrium uniqueness and the equilibrium stability under the condition that the monomer deposition stops. In the case that clusters evolve with dissociation and there is no monomer deposition, we show the equilibrium uniqueness and prove the equilibrium stability if the maximum cluster size is not larger than three while we show the equilibrium stability by numerical simulations if the maximum size is larger than three.

2. Description of the rate equation

We describe the rate equation characterizing the evolution of cluster-size distribution in this section and explain the terms in the model one by one. We focus on two quantities in the cluster evolution: the number and size of clusters.

Let $N_1(t)$ be the number of monomers at time t. Let n be the maximum size of clusters. For $2 \leqslant s \leqslant n$, let $N_s(t)$ be the number of clusters of size s at time t. Then the evolution of cluster-size distribution described in Section 1 can be characterized by the following rate equation:

$$\begin{aligned} N_1' &= F + 2D_2N_2 - 2K_1N_1^2 + \sum_{s=3}^{n} D_sN_s - N_1\sum_{s=2}^{n-1} K_sN_s, \\ N_s' &= K_{s-1}N_1N_{s-1} + D_{s+1}N_{s+1} - D_sN_s - K_sN_1N_s, \quad 2 \leqslant s \leqslant n-1, \\ N_n' &= K_{n-1}N_1N_{n-1} - D_nN_n, \end{aligned} \tag{1}$$

where $F > 0$, $D_s \geqslant 0$, $K_s > 0$ and $N_s \geqslant 0$ as $1 \leqslant s \leqslant n$. The term $N_s' = dN_s(t)/dt$ denotes the rate of change in the number of clusters of size s, $s \geqslant 1$. Parameter K_s denotes the rate at which monomers join clusters of size s. Parameter D_s denotes the rate at which monomers leave clusters of size s.

The first equation in (1) describes the change rate of monomers. Parameter F denotes the rate of deposition of monomers. The term "$+2D_2N_2$" shows that one cluster of size 2 becomes two monomers after a monomer's leaving. The term "$-2K_1N_1^2$" shows that two monomers become one cluster of size 2 after a monomer's joining. For $s \geqslant 3$, the term "$+D_sN_s$" shows that one cluster of size s becomes one monomer and one cluster of size $s-1$ after a monomer's leaving. For $s \geqslant 2$, the term "$-K_sN_1N_s$" shows that one monomer and one cluster of size s become one cluster of size $s+1$ after a monomer's joining.

The second equation of (1) describes the change rate of clusters of size s as $s \geqslant 2$. The term "$K_{s-1}N_1N_{s-1}$" denotes that one monomer and one cluster of size $s-1$ produce one cluster of size s after a monomer's joining. The term "$+D_{s+1}N_{s+1}$" denotes that one cluster of size $s+1$ produces one monomer and one cluster of size s after a monomer's leaving. The term "$-K_sN_1N_s$" denotes that one monomer and one cluster of size s produce one cluster of size $s+1$ after a monomer's joining. The term "$-D_sN_s$" denotes that one cluster of size s produces one monomer and one cluster of size $s-1$ after a monomer's leaving.

In the third equation of (1), the change rate of clusters of size n is described. Since the number n is the maximum cluster size, single monomers will not join a cluster of size n and there is no cluster of size $n+1$. Then the change rate of clusters of size n increases as a single monomer joins a cluster of size $n-1$, and decreases as a single monomer leaves a cluster of size n.

Hence, system (1) is in agreement with the mechanism of the evolution of cluster-size distribution described in Section 1. While dissociation is neglected in previous works [1–4], it is considered in Eq. (1) in this work. Our idea of introducing dissociation is motivated from [5,6]. In [5,6], Lerman and Shehory applied the rate equation in [4] in describing buyers' coalition formation in markets where they naturally considered buyers' dissocia-

tion. Introducing dissociation in modelling the submonolayer cluster growth will make the model more complete than the previous one.

In this paper, parameters K_s are assumed to be constant as time t varies while they may be different for different indexes s. For example, parameter K_2 is different from K_3 in the case of $K_2 = 0.2$ and $K_3 = 0.3$. That is, for different sizes s of clusters, the attachment rates K_s may be different, which means that the size of clusters is considered in Eq. (1). In [2], parameters K_s are functions of size s: K_s are proportional to $D \cdot S^p$, where D is the diffusion rate of single adatoms and p is a constant, see [2, p. 8783]. If $p = 0$, then Eq. (1) describes point (zero size) islands. If $p \neq 0$, then Eq. (1) describes nonzero size islands. Since both parameters D and p are constants as time t varies, parameters K_s are constant as time t varies. Then parameters K_s are effective for both point islands and nonzero size islands. Similar discussions can be given for parameters D_s. Hence, the size of clusters is considered in Eq. (1) while the geometry of clusters is neglected.

Now we discuss a minor error in [2] while [1,4] corrected the error without a theoretical proof. Ignoring dissociation (i.e., letting $D_s = 0$ in system (1)), [2] described the evolution of cluster-size distribution as follows, see [2, p. 8783]:

$$\begin{aligned} N_1' &= F - K_1 N_1^2 - N_1 \sum_{s=2}^{n-1} K_s N_s, \\ N_s' &= K_{s-1} N_1 N_{s-1} - K_s N_1 N_s, \quad 2 \leqslant s \leqslant n-1, \\ N_n' &= K_{n-1} N_1 N_{n-1}, \end{aligned} \tag{2}$$

where the second term "$-K_1 N_1^2$" on the right-hand side of the first equation in (2) is not accurate. In fact, suppose the deposition of monomers stops after a time period of deposition, i.e., let $F = 0$ as time $t > t_1$, where $t_1 > 0$. Then the amount of monomers on the surface must remain constant. Let n be the maximum size of clusters, we have

$$\sum_{s=1}^{n} s N_s(t) = \sum_{s=1}^{n} s N_s(t_1) \quad \text{as } t > t_1,$$

that is,

$$\sum_{s=1}^{n} s N_s'(t) = 0 \quad \text{as } t > t_1.$$

However, it follows from (2) that $\sum_{s=1}^{n} s N_s'(t) = K_1 N_1^2(t) > 0$ as $t > t_1$. This is a contradiction. Hence, the second term "$-K_1 N_1^2$" on the right-hand side of the first equation in (2) should be replaced with "$-2K_1 N_1^2$", which means that two monomers become one cluster of size 2 after a monomer's joining. After the replacement, it can be verified that $\sum_{s=1}^{n} s N_s'(t) = 0$ as $t > t_1$.

Let

$$\mathbf{R}_+^n = \{N = (N_1, N_2, \ldots, N_n)\colon\ N_s \geqslant 0,\ 1 \leqslant s \leqslant n\},$$

$$\Sigma_n = \mathbf{R}_+^n \cap \left\{ N\colon \sum_{s=1}^{n} s N_s = M \right\},$$

where $n > 0$ and $M = \sum_{s=1}^{n} sN_s(0)$. Let $\operatorname{int}\mathbf{R}_+^n$ be the inner points of $\mathbf{R}_+^n$ and denote $\operatorname{int}\Sigma_n = \operatorname{int}\mathbf{R}_+^n \cap \Sigma_n$.

3. Positiveness of solutions

While system (1) has been widely used in describing dynamic scaling of cluster-size distribution and has been verified to be effective by physical experiments [1–3], it should be proven from a mathematical perspective. One of the problems we are concerned with is the positiveness of solutions of (1). Since solutions of the model denote the quantities of clusters with different sizes, they should not be negative with the passage of time.

In this section, we show that solutions of (1) initiated from $\mathbf{R}_+^n$ (i.e., as $t = t_0$) will remain in $\operatorname{int}\mathbf{R}_+^n$ as $t > t_0$. We consider two cases:

(i) the initial vector $N(t_0) = (N_1(t_0), N_2(t_0), \ldots, N_n(t_0))$ is on the bound of $\mathbf{R}_+^n$, i.e., $N(t_0) \in \partial\mathbf{R}_+^n$;
(ii) the initial vector $N(t_0) = (N_1(t_0), N_2(t_0), \ldots, N_n(t_0))$ is in the interior region of $\mathbf{R}_+^n$, i.e., $N(t_0) \in \operatorname{int}\mathbf{R}_+^n$.

In the case of $N(t_0) \in \partial\mathbf{R}_+^n$, some components of $N(t_0)$ are positive and others are zero. First we study two types of $N(t_0)$ in Lemmas 2–3:

(a) $N(t_0) = (\ldots, +, 0, \ldots, 0, +, \ldots)$;
(b) $N(t_0) = (\ldots, +, 0, \ldots, 0)$.

Then we show the positiveness of solutions for the case of $N(t_0) \in \partial\mathbf{R}_+^n$ in Lemma 4. In the case of $N(t_0) \in \operatorname{int}\mathbf{R}_+^n$, we prove the positiveness of solutions in Lemma 5. Without loss of generality, we assume $t_0 = 0$ in this work since system (1) is autonomous.

Lemma 1. *Let $N(t)$ be a solution of* (1) *with $N(0) \in \mathbf{R}_+^n$. Then there is $\delta_1 > 0$ such that*

$$N_1(t) > 0 \quad \text{as } t \in (0, \delta_1).$$

Proof. It follows from $F > 0$ that monomers are deposited to the smooth surface continuously, then without loss of generality, we can assume that $N_1(0) > 0$ and $N_s(0) \geqslant 0$ as $s > 1$. By the continuity of $N_1(t)$, there is $\delta_1 > 0$ such that $N_1(t) > 0$ as $t \in (0, \delta_1)$. □

Lemma 2. *Let $N(t)$ be a solution of* (1) *with $N(0) \in \partial\mathbf{R}_+^n$. If there are l and i, $1 \leqslant l < i$, such that $N_l(0) > 0$, $N_k(0) = 0$ as $l + 1 \leqslant k \leqslant i$ and $N_{i+1}(0) > 0$, then there is $\delta > 0$ such that*

$$N_s(t) > 0 \quad \text{as } t \in (0, \delta) \text{ and } l + 1 \leqslant s \leqslant i.$$

Proof. It follows from [13] that $N_s(t)$ is analytic as $s \geqslant 1$. Since $N_l(0) > 0$, then there is δ_l, $0 < \delta_l < \delta_1$, such that $N_l(t) > 0$ as $t \in (0, \delta_l)$. Since $N_i(0) = 0$ and $N_{i+1}(0) > 0$, it follows from the ith equation of (1) that

$$N_i'(0) = K_{i-1}N_1(0)N_{i-1}(0) + D_{i+1}N_{i+1}(0) > 0,$$

that is, there is η_i, $0 < \eta_i < \delta_l$, such that $N_i(t) > 0$ as $t \in (0, \eta_i)$.

Suppose there is c, $0 < c < \eta_i$, such that $N_{i-1}(t) \leqslant 0$ as $t \in (0, c)$. It follows from the analyticity of $N_{i-1}(t)$ that there is δ_{i-1}, $0 < \delta_{i-1} < c$, such that $N_{i-1}'(t) \leqslant 0$ as $t \in (0, \delta_{i-1})$. It follows from the $(i-1)$th equation of (1) that

$$N_{i-1}' = N_1(K_{i-2}N_{i-2} - K_{i-1}N_{i-1}) - D_{i-1}N_{i-1} + D_iN_i,$$

then $N_{i-2}(t) < 0$ as $t \in (0, \delta_{i-1})$.

Let $z = N_{i-1} + N_{i-2}$, then $z(0) = 0$ and $z(t) < 0$ as $t \in (0, \delta_{i-1})$. It follows from the analyticity of $z(t)$ that there is δ_{i-2}, $0 < \delta_{i-2} < \delta_{i-1}$, such that $z'(t) < 0$ as $t \in (0, \delta_{i-2})$. Since

$$N_{i-2}' = N_1(K_{i-3}N_{i-3} - K_{i-2}N_{i-2}) - D_{i-2}N_{i-2} + D_{i-1}N_{i-1},$$

then

$$z' = N_1(K_{i-3}N_{i-3} - K_{i-1}N_{i-1}) - D_{i-2}N_{i-2} + D_iN_i < 0 \quad \text{as } t \in (0, \delta_{i-2}),$$

that is, $N_{i-3}(t) < 0$ as $t \in (0, \delta_{i-2})$.

We can use this method inductively, then there is δ_k, $0 < \delta_k < \delta_{k+1}$, such that $N_{k-1}(t) < 0$ as $t \in (0, \delta_k)$ and $l \leqslant k \leqslant i-2$. Let $k = l$, then there is δ_l, $0 < \delta_l < \delta_{l+1}$, such that $N_l(t) < 0$ as $t \in (0, \delta_l)$. This contradicts that $N_l(t) > 0$ as $t \in (0, \eta_l)$.

Hence, there is η_{i-1}, $0 < \eta_{i-1} < \eta_i$, such that $N_{i-1}(t) > 0$ as $t \in (0, \eta_{i-1})$. We can use this method inductively, then there is η_k, $0 < \eta_k < \eta_{k+1}$, such that $N_k(t) > 0$ as $t \in (0, \eta_k)$ and $l \leqslant k \leqslant i-1$. Then there is η_l, $0 < \eta_l < \eta_{l+1}$, such that $N_l(t) > 0$ as $t \in (0, \eta_l)$. Let $\delta = \eta_l$, then $N_s(t) > 0$ as $t \in (0, \delta)$ and $l + 1 \leqslant s \leqslant i$. □

Lemma 2 covers all initial conditions as follows:

$$(\ldots, +, 0, \ldots, 0, +, \ldots, +, 0, \ldots, 0, +, \ldots, +, 0, \ldots, 0, +, \ldots).$$

For example, consider the case of two isolated (interval of) values of s:

$$(\ldots, +, 0, \ldots, 0, +, \ldots, +, 0, \ldots, 0, +, \ldots),$$

that is, there are $1 \leqslant l < i \leqslant j < k \leqslant n$ such that

$$N_s(0) > 0 \quad \text{as } 1 \leqslant s \leqslant l, \qquad N_s(0) = 0 \quad \text{as } l < s < i,$$
$$N_s(0) > 0 \quad \text{as } i \leqslant s \leqslant j, \qquad N_s(0) = 0 \quad \text{as } j < s < k,$$

and $N_s(0) > 0$ as $k \leqslant s \leqslant n$.

For the interval of values $l < s < i$, it follows from Lemma 2 that there is $\delta_{li} > 0$ such that

$$N_s(t) > 0 \quad \text{as } l < s < i \text{ and } t \in (0, \delta_{li}).$$

For the interval of values $j < s < k$, it follows from Lemma 2 that there is $\delta_{jk} > 0$ such that

$$N_s(t) > 0 \quad \text{as } j < s < k \text{ and } t \in (0, \delta_{jk}).$$

Let $\delta_{lk} = \min\{\delta_{li}, \delta_{jk}\}$, then $\delta_{lk} > 0$ and

$$N_s(t) > 0 \quad \text{as } l < s < i,\ j < s < k \text{ and } t \in (0, \delta_{lk}).$$

Since initial values $N_s(0)$ are positive elsewhere ($1 \leqslant s \leqslant l$, $i \leqslant s \leqslant j$, $k \leqslant s \leqslant n$), by the continuity of $N_s(t)$, then there is $\delta_{1n} > 0$ such that

$$N_s(t) > 0 \quad \text{as } 1 \leqslant s \leqslant l,\ i \leqslant s \leqslant j,\ k \leqslant s \leqslant n \text{ and } t \in (0, \delta_{1n}).$$

Let $\delta = \min\{\delta_{lk}, \delta_{1n}\}$, then $\delta > 0$ and

$$N_s(t) > 0 \quad \text{as } 1 \leqslant s \leqslant n \text{ and } t \in (0, \delta).$$

Lemma 3. *Let $N(t)$ be a solution of* (1) *with $N(0) \in \partial\mathbf{R}^n_+$. If there is i, $1 \leqslant i \leqslant n-1$, such that $N_i(0) > 0$ and $N_s(0) = 0$ as $i+1 \leqslant s \leqslant n$. Then there is $\delta > 0$ such that*

$$N_s(t) > 0 \quad \text{as } t \in (0, \delta) \text{ and } i+1 \leqslant s \leqslant n.$$

Proof. It follows from [13] that $N_s(t)$ is analytic as $1 \leqslant s \leqslant n$. Since $N_i(0) > 0$, then there is δ_i, $0 < \delta_i < \delta_1$, such that $N_i(t) > 0$ as $t \in (0, \delta_i)$.

Suppose there is c, $0 < c < \delta_i$, such that $N_{i+1}(t) \leqslant 0$ as $t \in (0, c)$. It follows from the analyticity of $N_{i+1}(t)$ that there is δ_{i+1}, $0 < \delta_{i+1} < c$, such that $N'_{i+1}(t) \leqslant 0$ as $t \in (0, \delta_{i+1})$. It follows from the $(i+1)$th equation of (1) that

$$N'_{i+1} = N_1(K_i N_i - K_{i+1}N_{i+1}) - D_{i+1}N_{i+1} + D_{i+2}N_{i+2} \leqslant 0,$$

then $N_{i+2}(t) < 0$ as $t \in (0, \delta_{i+1})$.

Let $z = N_{i+1} + N_{i+2}$, then $z(0) = 0$ and $z(t) < 0$ as $t \in (0, \delta_{i+1})$. It follows from the analyticity of $z(t)$ that there is δ_{i+2}, $0 < \delta_{i+2} < \delta_{i+1}$, such that $z'(t) < 0$ as $t \in (0, \delta_{i+2})$. Since

$$N'_{i+2} = N_1(K_{i+1}N_{i+1} - K_{i+2}N_{i+2}) - D_{i+2}N_{i+2} + D_{i+3}N_{i+3},$$

then

$$z' = N_1(K_i N_i - K_{i+2}N_{i+2}) - D_{i+1}N_{i+1} + D_{i+3}N_{i+3} < 0 \quad \text{as } t \in (0, \delta_{i+2}),$$

that is, $N_{i+3}(t) < 0$ as $t \in (0, \delta_{i+2})$.

We can use this method inductively, then there is δ_k, $0 < \delta_k < \delta_{k-1}$, such that $N_{k+1}(t) < 0$ as $t \in (0, \delta_k)$ and $i+3 \leqslant k \leqslant n-1$. Here, n is assumed to be the maximum size of clusters during the time period $(0, \delta_1)$. Let $k = n-1$, then there is δ_{n-1}, $0 < \delta_{n-1} < \delta_{n-2}$, such that $N_l(t) < 0$ as $t \in (0, \delta_{n-1})$ and $i+2 \leqslant l \leqslant n$.

Let $w = \sum_{l=i+1}^{n} N_l$, then $w(0) = 0$ and $w(t) < 0$ as $t \in (0, \delta_{n-1})$. Since

$$w' = K_i N_1 N_i - D_{i+1}N_{i+1} > 0 \quad \text{as } t \in (0, \delta_{n-1}),$$

this is a contradiction. Hence, there is η_{i+1}, $0 < \eta_{i+1} < \delta_i$, such that $N_{i+1}(t) > 0$ as $t \in (0, \eta_{i+1})$. We can use this method inductively, then there is η_k, $0 < \eta_k < \eta_{k-1}$, such that $N_k(t) > 0$ as $t \in (0, \eta_k)$ and $i+1 < k \leqslant n$.

Let $\delta = \eta_n$, then $N_s(t) > 0$ as $t \in (0, \delta)$ and $i+1 \leqslant s \leqslant n$. □

By Lemmas 1–3, all initial conditions on ∂R^n_+ are covered. The initial conditions can be divided into two types, one is

$$(+,\dots,+,0,\dots,0,+,\dots,+,0,\dots,0,+,\dots,+),$$

and the other is

$$(+,\dots,+,0,\dots,0,+,\dots,+,0,\dots,0,+,\dots,+,0,\dots,0).$$

While the former type is covered by Lemma 2, the latter type is covered by Lemmas 2–3. For example, consider the case

$$(+,\dots,+,0,\dots,0,+,\dots,+,0,\dots,0),$$

that is, there are $1 \leqslant i < j \leqslant k < n$ such that

$$N_s(0) > 0 \quad \text{as } 1 \leqslant s \leqslant i, \qquad N_s(0) = 0 \quad \text{as } i < s < j,$$
$$N_s(0) > 0 \quad \text{as } j \leqslant s \leqslant k, \qquad N_s(0) = 0 \quad \text{as } k < s \leqslant n.$$

For the interval of values $i < s < j$, it follows from Lemma 2 that there is $\delta_{ij} > 0$ such that

$$N_s(t) > 0 \quad \text{as } i < s < j \text{ and } t \in (0, \delta_{ij}).$$

For the interval of values $k < s \leqslant n$, it follows from Lemma 3 that there is $\delta_{kn} > 0$ such that

$$N_s(t) > 0 \quad \text{as } k < s \leqslant n \text{ and } t \in (0, \delta_{kn}).$$

Let $\delta_{in} = \min\{\delta_{ij}, \delta_{kn}\}$, then $\delta_{in} > 0$ and

$$N_s(t) > 0 \quad \text{as } i < s < j,\ k < s \leqslant n \text{ and } t \in (0, \delta_{in}).$$

Since initial values $N_s(0)$ are positive elsewhere ($1 \leqslant s \leqslant i,\ j \leqslant s \leqslant k$), similar to the discussion behind Lemma 2, there is $\delta > 0$ such that

$$N_s(t) > 0 \quad \text{as } 1 \leqslant s \leqslant n \text{ and } t \in (0, \delta).$$

It follows from Lemmas 1–3 that

Lemma 4. *Let $N(t)$ be a solution of* (1) *with $N(0) \in \partial \mathbf{R}^n_+$, then there is $\delta > 0$ such that $N(t) \in \operatorname{int} \mathbf{R}^n_+$ as $t \in (0, \delta)$.*

The following lemma considers the case of $N(0) \in \operatorname{int} \mathbf{R}^n_+$.

Lemma 5. *Let $N(t)$ be a solution of* (1) *with $N(0) \in \operatorname{int} \mathbf{R}^n_+$, then $N(t) \in \operatorname{int} \mathbf{R}^n_+$ as $t > 0$.*

Proof. Suppose there are $t_1 > 0$, $m > 0$, $k > 0$ and $m + k < n$ such that

$$N_i(t) > 0 \quad \text{as } t \in [0, t_1) \text{ and } m \leqslant i \leqslant m + k + 1,$$
$$N_m(t_1) > 0, \qquad N_{m+k+1}(t_1) > 0,$$

and

$$N_s(t_1) = 0 \quad \text{as } m + 1 \leqslant s \leqslant m + k,$$

where n is assumed to be the maximum size of clusters during the time period $(0, t_1)$. Let $z = \sum_{j=m+1}^{m+k} N_j$, then $z(t_1) = 0$ and $z(t) > 0$ as $t \in [0, t_1)$. Hence, $z'(t_1) \leqslant 0$, which contradicts that

$$z'(t_1) = K_m N_1 N_m - D_{m+1} N_{m+1} - K_{m+k} N_1 N_{m+k} + D_{m+k+1} N_{m+k+1} > 0.$$

Suppose there are $t_1, m > 0$ and $m < n$ such that

$$N_i(t) > 0 \quad \text{as } t \in [0, t_1) \text{ and } m \leqslant i \leqslant n,$$

and

$$N_m(t_1) > 0 \quad \text{and} \quad N_s(t_1) = 0 \quad \text{as } m + 1 \leqslant s \leqslant n.$$

Let $w = \sum_{j=m+1}^{n} N_j$, then $w(t_1) = 0$ and $w(t) > 0$ as $t \in [0, t_1)$. Hence, $w'(t_1) \leqslant 0$, which contradicts that

$$w'(t_1) = K_m N_1 N_m - D_{m+1} N_{m+1} > 0.$$

Therefore, we have $N_s(t) > 0$ as $t > 0$ and $1 \leqslant s \leqslant n$, i.e., $N(t) \in \operatorname{int} \Sigma_n$ as $t > 0$. □

While Lemma 4 shows that solutions of (1) initiated from $\partial \mathbf{R}_+^n$ go into $\operatorname{int} \mathbf{R}_+^n$ with the passage of time, Lemma 5 shows that solutions of (1) will remain in $\operatorname{int} \mathbf{R}_+^n$ after they go into $\operatorname{int} \mathbf{R}_+^n$ at some time. Hence, we have

Theorem 1. *Each solution $N(t)$ of* (1) *with $N(0) \in \mathbf{R}_+^n$ satisfies that $N(t) \in \operatorname{int} \mathbf{R}_+^n$ as $t > 0$.*

4. Stability without dissociation

In this section, we show the evolution of cluster-size distribution on a surface without dissociation, i.e., $D_s = 0$ as $s > 1$. In order to focus on the dynamic behavior of clusters, we assume that after a time period of monomer deposition, the deposition of monomers stops from time t_1, where $t_1 > 0$, i.e., $F > 0$ as $t \in (0, t_1)$ and $F = 0$ as $t \geqslant t_1$. We focus on the dynamics of (1) as $t \geqslant t_1$. Then $\sum_{s=1}^{n} s N_s(t_1) = M > 0$ and system (1) becomes:

$$\begin{aligned} N_1' &= -2K_1 N_1^2 - N_1 \sum_{s=2}^{n-1} K_s N_s, \\ N_s' &= K_{s-1} N_1 N_{s-1} - K_s N_1 N_s, \quad 2 \leqslant s \leqslant n-1, \\ N_n' &= K_{n-1} N_1 N_{n-1}. \end{aligned} \tag{3}$$

Since dissociation is ignored in (3), there would be no evolution of cluster-size distribution if $N_1(t_1) = 0$. Hence, we assume $N_1(t_1) > 0$. Similar to the proof of Theorem 1, we have

Lemma 6. *Each solution $N(t)$ of* (3) *with $N(t_1) \in \Sigma_n$ satisfies that $N(t) \in \operatorname{int} \Sigma_n$ as $t > t_1$, where $\Sigma_n = \mathbf{R}_+^n \cap \{N\colon \sum_{s=1}^{n} s N_s = M\}$.*

Since $\sum_{s=1}^{n} s N_s' = 0$, we have

Theorem 2. *Let* $N(t) = (N_1(t), N_2(t), \ldots, N_n(t))$ *be a solution of* (3) *with* $N(t_1) \in \Sigma_n$. *Then*

$$\sum_{s=1}^{n} sN_s(t) = M \quad \text{as } t > t_1.$$

Theorem 2 shows that the amount of monomers remains constant during the evolution of cluster-size distribution as $t > t_1$, which is in agreement with experiments in [1,2] when $F = 0$.

It can be verified that $N^* = (0, \ldots, 0, M/n)$ is an equilibrium of (3), then we have

Theorem 3. *Equilibrium* N^* *of* (3) *is globally asymptotically stable in* $\{N\colon\ N \in \Sigma_n$ *and* $N_1 > 0\}$.

Proof. By the replacement of $d\tau = N_1(t)\,dt$, the first $n-1$ equations of (3) become:

$$\begin{aligned} N_1' &= -2K_1N_1 - \sum_{s=2}^{n-1} K_sN_s, \\ N_s' &= K_{s-1}N_{s-1} - K_sN_s, \quad 2 \leqslant s \leqslant n-1. \end{aligned} \tag{4}$$

System (4) is a linear model with coefficient matrix

$$A = \begin{pmatrix} -2K_1 & -K_2 & \ldots & -K_{n-1} \\ K_1 & -K_2 & 0 & 0 \\ 0 & \ldots & \ldots & 0 \\ 0 & \ldots & K_{n-2} & -K_{n-1} \end{pmatrix}.$$

It follows from Hurwitz's criteria [14] that the real parts of eigenvalues of matrix A are negative since

$$\begin{aligned} &2K_1 > 0, \\ &\det\begin{pmatrix} 2K_1 & K_2 \\ -K_1 & K_2 \end{pmatrix} = 3K_1K_2 > 0, \\ &\quad\vdots \\ &\det(-A) = nK_1K_2\ldots K_{n-1} > 0. \end{aligned}$$

Hence, the original point of (4) is globally asymptotically stable, i.e., all solutions of (4) converge to the original point. It follows from Theorem 2 that equilibrium N^* of (3) is globally asymptotically stable in $\{N\colon\ N \in \Sigma_n$ and $N_1 > 0\}$. □

5. Stability with dissociation

In this section, we show the evolution of cluster-size distribution on a surface with dissociation, i.e., $D_s > 0$ as $s > 1$. In order to focus on the dynamic behavior of clusters, we assume that after a time period of monomer deposition, the deposition of monomers

stops from time t_1 where $t_1 > 0$, i.e., $F > 0$ as $t \in (0, t_1)$ and $F = 0$ as $t \geqslant t_1$ in system (1). We focus on the dynamics of (1) as $t \geqslant t_1$. Then $\sum_{s=1}^{n} sN_s(t_1) = M > 0$ and system (1) becomes:

$$\begin{aligned} N_1' &= 2D_2N_2 - 2K_1N_1^2 + \sum_{s=3}^{n} D_sN_s - N_1\sum_{s=2}^{n-1} K_sN_s, \\ N_s' &= K_{s-1}N_1N_{s-1} + D_{s+1}N_{s+1} - D_sN_s - K_sN_1N_s, \quad 2 \leqslant s \leqslant n-1, \\ N_n' &= K_{n-1}N_1N_{n-1} - D_nN_n. \end{aligned} \tag{5}$$

Since $\sum_{s=1}^{n} sN_s(t_1) = M > 0$, then there is $\eta_1 > 0$ such that $N_1(t) > 0$ as $t \in (t_1, t_1 + \eta_1)$. In fact, if $N_1(t_1) = 0$, then it follows from the first equation of (5) that $N_1'(t_1) > 0$. Hence, similar to the proof of Theorem 1, we have

Lemma 7. *Each solution $N(t)$ of* (5) *with $N(t_1) \in \Sigma_n$ satisfies that $N(t) \in \operatorname{int}\Sigma_n$ as $t > t_1$, where $\Sigma_n = \mathbf{R}_+^n \cap \{N\colon \sum_{s=1}^{n} sN_s = M\}$.*

Since $\sum_{s=1}^{n} sN_s' = 0$, we have

Theorem 4. *Let $N(t) = (N_1(t), N_2(t), \ldots, N_n(t))$ be a solution of* (5) *with $N(t_1) \in \Sigma_n$. Then*

$$\sum_{s=1}^{n} sN_s(t) = M \quad \text{as } t > t_1.$$

Theorem 4 shows that the amount of monomers remains constant during the evolution of cluster-size distribution with dissociation.

Theorem 5. *There is a unique equilibrium N^* of* (5)*, where $N^* = (N_1^*, N_2^*, \ldots, N_n^*)$, N_1^* satisfies*

$$\sum_{s=1}^{n} sh_sN_1^{*s} - M = 0,$$

$$h_1 = 1, \qquad h_s = \frac{K_1K_2\ldots K_{s-1}}{D_2D_3\ldots D_s} \quad \text{as } 2 \leqslant s \leqslant n,$$

and

$$N_s^* = h_sN_1^{*s} \quad \text{as } 1 \leqslant s \leqslant n.$$

Proof. By the definition of equilibrium, the right-hand sides of equations in (5) should be zero at the equilibrium. Let $N^* = (N_1^*, N_2^*, \ldots, N_n^*)$ be the equilibrium of (5). Then by the right-hand side of the nth equation of (5), we have

$$N_n^* = \frac{K_{n-1}}{D_n}N_1^*N_{n-1}^*.$$

It follows from the right-hand side of the $(n-1)$th equation in (5) that

$$N_{n-1}^* = \frac{K_{n-2}}{D_{n-1}} N_1^* N_{n-2}^*$$

by replacing N_n^* with $\frac{K_{n-1}}{D_n} N_1^* N_{n-1}^*$. Inductively, it follows from the right-hand side of the ith $(2 \leqslant i \leqslant n-2)$ equation in (5) that

$$N_s^* = \frac{K_{s-1}}{D_s} N_1^* N_{s-1}^* \quad \text{as } 2 \leqslant s \leqslant n.$$

Hence,

$$N_2^* = \frac{K_1}{D_2} N_1^{*2},$$
$$N_3^* = \frac{K_2}{D_3} N_1^* N_2^* = \frac{K_1 K_2}{D_2 D_3} N_1^{*3}.$$

Inductively, we have

$$N_s^* = \frac{K_1 K_2 \dots K_{s-1}}{D_2 D_3 \dots D_s} N_1^{*s} \quad \text{as } 2 \leqslant s \leqslant n.$$

Then we have $N_s^* = h_s N_1^{*s}$ as $1 \leqslant s \leqslant n$.

Let $G(z) = \sum_{s=1}^{n} s h_s z^s - M$, then $G(0) = -M < 0$, $G(M) > 0$ and

$$G'(z) = \sum_{s=1}^{n} s^2 h_s z^{s-1} > 0 \quad \text{as } z > 0.$$

Hence, function $G(z)$ is monotonous and has a unique solution of $G(z) = 0$ in $(0, M)$. Therefore, there is a unique equilibrium of (5) in int Σ_n and N_1^* is the root of $G(z) = 0$. □

Theorem 6. *For the cases of $n = 2$ and $n = 3$, the unique equilibrium of* (5) *is globally asymptotically stable in Σ_n.*

Proof. (i) In the case of $n = 2$, system (5) becomes

$$N_1' = 2D_2 N_2 - 2K_1 N_1^2,$$
$$N_2' = -D_2 N_2 + K_1 N_1^2, \tag{6}$$

where $N_1 + 2N_2 = M$. Since $N_2 = \frac{1}{2}(M - N_1)$, then the first equation of (6) becomes

$$N_1' = H(N_1), \tag{7}$$

where $H(N_1) = D_2 M - D_2 N_1 - 2K_1 N_1^2$ and $0 \leqslant N_1 \leqslant M$. Then $H(N_1)' = -D_2 - 4K_1 N_1 < 0$ as $N_1 > 0$. Let (N_1^*, N_2^*) be the unique equilibrium of (6). Then $H(N_1^*) = 0$, $N_1' = H(N_1) < 0$ as $N_1 > N_1^*$, and $N_1' = H(N_1) > 0$ as $N_1 < N_1^*$. Hence, the unique equilibrium of (7) is globally asymptotically stable on $[0, M]$, i.e., the equilibrium of (6) is globally asymptotically stable in Σ_2.

(ii) In the case of $n = 3$, system (5) becomes

$$N_1' = 2D_2N_2 - 2K_1N_1^2 + D_3N_3 - K_2N_1N_2 - K_3N_1N_3,$$
$$N_2' = D_3N_3 + K_1N_1^2 - D_2N_2 - K_2N_1N_2,$$
$$N_3' = K_2N_1N_2 - D_3N_3,$$

where $N_1 + 2N_2 + 3N_3 = M$. Since $N_3 = (M - N_1 - 2N_2)/3$, then we have

$$N_1' = f_1(N_1, N_2),$$
$$N_2' = f_2(N_1, N_2), \tag{8}$$

where $0 \leqslant N_1 + 2N_2 \leqslant M$, $N_1 \geqslant 0$, $N_2 \geqslant 0$ and

$$f_1(N_1, N_2) = 2D_2N_2 + D_3(M - N_1 - 2N_2)/3 - 2K_1N_1^2 - K_2N_1N_2,$$
$$f_2(N_1, N_2) = -D_2N_2 + D_3(M - N_1 - 2N_2)/3 + K_1N_1^2 - K_2N_1N_2.$$

Since

$$\frac{\partial f_1}{\partial N_1} = -4K_1N_1 - K_2N_2 - D_3/3,$$
$$\frac{\partial f_2}{\partial N_2} = -K_2N_1 - D_2 - 2D_3/3,$$

then

$$\frac{\partial f_1}{\partial N_1} + \frac{\partial f_2}{\partial N_2} = -D_2 - D_3 - (4K_1 + K_2)N_1 - K_2N_2 < 0.$$

It follows from Dulac's criteria [14] that there is no periodic orbit of (8).

Let $Q(N_1^*, N_2^*, N_3^*)$ be the unique equilibrium of (5) as $n = 3$, then $q(N_1^*, N_2^*)$ is an equilibrium of (8). The Jacobian matrix of (8) at the equilibrium is:

$$A = \begin{pmatrix} \frac{\partial f_1}{\partial N_1} & \frac{\partial f_1}{\partial N_2} \\ \frac{\partial f_2}{\partial N_1} & \frac{\partial f_2}{\partial N_2} \end{pmatrix}.$$

Since

$$\left.\frac{\partial f_1}{\partial N_1} + \frac{\partial f_2}{\partial N_2}\right|_q = -D_2 - D_3 - (4K_1 + K_2)N_1^* - K_2N_2^* < 0,$$
$$\det A|_q = D_2\big(D_3 + 3K_2N_2^*\big) + K_1N_1^*\big(6K_2N_1^* + 4D_3\big) > 0,$$

the eigenvalues of A have negative real parts, i.e., equilibrium q of (8) is asymptotically stable.

It follows from Poincare–Bendixon theorem [14] that all solutions of (8) converge to q as $t \to \infty$, i.e., the equilibrium q of (8) is globally asymptotically stable, then the equilibrium Q of (5) is globally asymptotically stable in Σ_3. □

In the cases of $n > 3$, the present work does not prove the global stability of the equilibrium theoretically. The reason is that it involves $n-1$ ($n-1 \geqslant 3$) differential equations and analyzing a system of $n-1$ ($n-1 \geqslant 3$) differential equations is difficult and to the best of our knowledge there is no general method for such analysis unlike the case when $n-1=2$

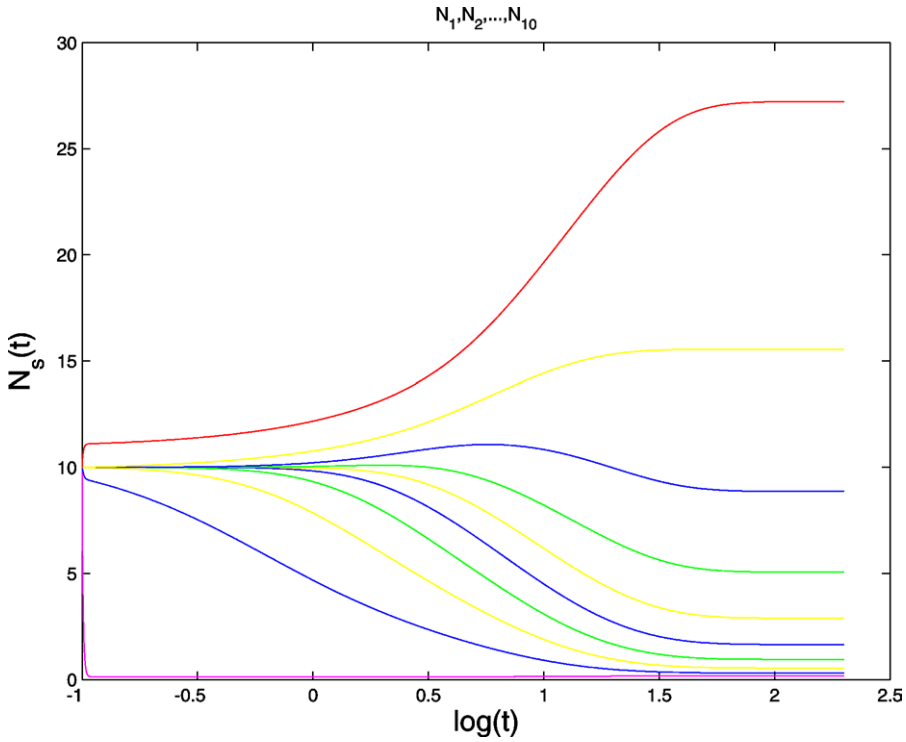

Fig. 1. Let $n = 10$, $M = 550$, $K_s = 0.1$ and $D_s = 0.01$ as $1 \leqslant s \leqslant 10$. Let $N(0) = (10, 10, \ldots, 10)$. Then up to time $t = 200$, the solution $N(t)$ of (5) converges to the stationary values (0.1778, 0.3083, 0.5381, 0.9442, 1.6538, 2.8903, 5.0637, 8.8752, 15.5516, 27.2089), which are consistent with the equilibrium obtained by the expression in Theorem 5: $N^* = (0.1752, 0.3070, 0.5378, 0.9422, 1.6507, 2.8920, 5.0669, 8.8771, 15.5527, 27.2484)$.

where the powerful Poincare–Bendixson theorem can be used. A series of numerical simulations that we have done show that in the cases of $n > 3$, the unique equilibrium of (5) is globally asymptotically stable without more complex behavior such as limit cycles and chaos. Numerical simulations are given below to show the global stability of the equilibrium. In Fig. 1, we show that a specific solution of (5) converges to the equilibrium in Theorem 5 while in Fig. 2, we show that the tenth components of five different solutions converge to the tenth component N_{10}^* of the equilibrium in Theorem 5.

Let

$$n = 10, \quad M = 550, \quad K_s = 0.1 \quad \text{and} \quad D_s = 0.01 \quad \text{as } 1 \leqslant s \leqslant 10.$$

We compare the equilibrium obtained by the expression in Theorem 5 and the stationary values obtained by numerical simulations as follows. To use the expression in Theorem 5, let $z = N_1^*$, then we have

$$G(z) = \sum_{i=1}^{10} i * (10 * z)^i / 10 - 550.$$

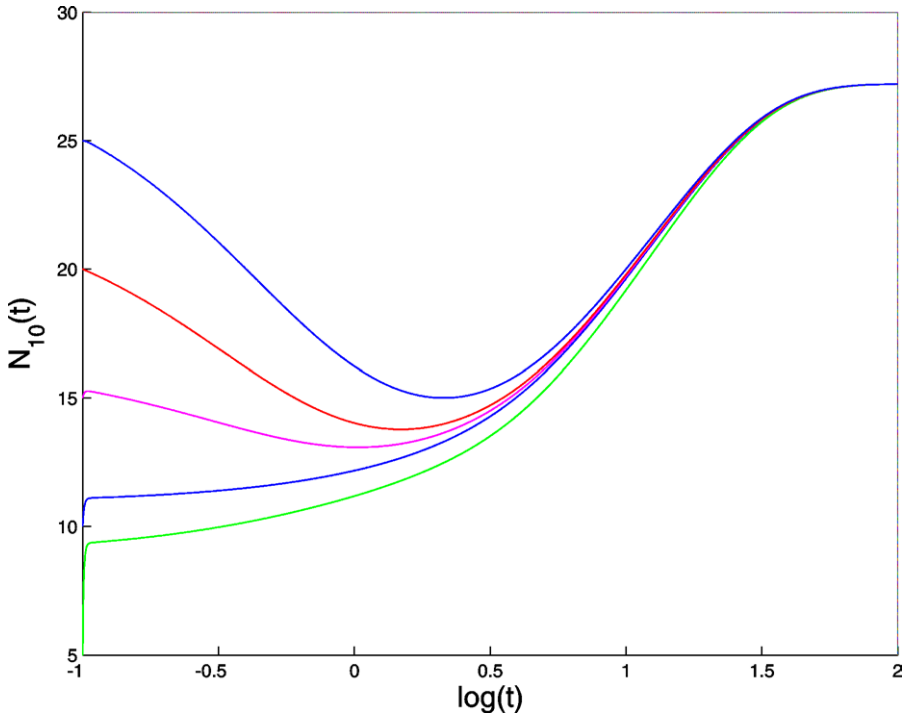

Fig. 2. Let $n = 10$, $M = 550$, $K_s = 0.1$ and $D_s = 0.01$ as $1 \leqslant s \leqslant 10$. The initial values of five solutions are as follows: $N^1(0) = (10, 10, 10, 10, 10, 10, 10, 10, 10, 10)$, $N^2(0) = (60, 10, 10, 10, 10, 10, 10, 10, 10, 5)$, $N^3(0) = (5, 10, 10, 10, 10, 10, 10, 10, 5, 15)$, $N^4(0) = (0, 10, 10, 10, 10, 10, 10, 10, 0, 20)$ and $N^5(0) = (6, 10, 10, 10, 10, 10, 3, 0, 25)$. The tenth components of the five different solutions of (5) converge to 27.2041, 27.2047, 27.2052, 27.2061, 27.2073, respectively, which are consistent with the value derived in Theorem 5: $N_{10}^* = 27.2484$.

By software MatLab, we obtain the root of $G(z) = 0$: $z = 0.1752$. Then we obtain the equilibrium

$$\begin{aligned} N^* &= \left(N_1^*, N_2^*, N_3^*, N_4^*, N_5^*, N_6^*, N_7^*, N_8^*, N_9^*, N_{10}^*\right) \\ &= (0.1752, 0.3070, 0.5378, 0.9422, 1.6507, 2.8920, 5.0669, 8.8771, \\ &\quad 15.5527, 27.2484). \end{aligned}$$

In Fig. 1, let $N(t_1) = (10, 10, 10, 10, 10, 10, 10, 10, 10, 10)$, simulations show that up to time $t = 200$, the solution $N(t)$ of (5) converges to the stationary values

$$\begin{aligned} N^* &= (0.1778, 0.3083, 0.5381, 0.9442, 1.6538, 2.8903, 5.0637, 8.8752, \\ &\quad 15.5516, 27.2089), \end{aligned}$$

which are consistent with the equilibrium obtained by the expression in Theorem 5.

In Fig. 2, we show that the tenth components of five different solutions of (5) converge to the same value N_{10}^*. The initial values of the five solutions are as follows:

$$N^1(0) = (10, 10, 10, 10, 10, 10, 10, 10, 10, 10),$$
$$N^2(0) = (60, 10, 10, 10, 10, 10, 10, 10, 10, 5),$$
$$N^3(0) = (5, 10, 10, 10, 10, 10, 10, 10, 5, 15),$$
$$N^4(0) = (0, 10, 10, 10, 10, 10, 10, 10, 0, 20),$$
$$N^5(0) = (6, 10, 10, 10, 10, 10, 10, 3, 0, 25).$$

Simulations show that up to time $t = 100$, the tenth components of the above five different solutions converge to $27.2041, 27.2047, 27.2052, 27.2061, 27.2073$, respectively, which are consistent with the value derived in Theorem 5: $N_{10}^* = 27.2484$.

Consider the case of uniform attachment-uniform detachment, i.e., $K_i = K$, $D_i = D$ as $1 \leqslant i \leqslant n$. Let

$$\tau = Kt, B = D/K,$$

then system (5) becomes (we still denote τ by t):

$$\begin{aligned} N_1' &= 2BN_2 + \sum_{k=3}^{n} BN_k - 2N_1^2 - N_1 \sum_{k=2}^{n-1} N_k, \\ N_s' &= -BN_s + BN_{s+1} + N_1N_{s-1} - N_1N_s, \quad 2 \leqslant s \leqslant n-1, \\ N_n' &= -BN_n + N_1N_{n-1}. \end{aligned} \tag{9}$$

Let $N^* = (N_1^*, N_2^*, \ldots, N_n^*)$ be the unique equilibrium of (9) and let $z = N_1^*/B$, then we have $N_s^* = Bz^s$, $s = 1, \ldots, n$, and

$$\sum_{s=1}^{n} sz^s = MB^{-1}. \tag{10}$$

It follows from the implicit function theorem [15] that Eq. (10) defines a smooth function $z = z(B)$ and we have

$$\frac{dz}{dB} = -\frac{M}{B^2 \sum_{s=1}^{n} s^2 z^{s-1}} < 0.$$

It follows from $N_s^* = Bz^s$ that $N_1^* = N_2^* = \cdots = N_n^*$ if and only if $z = 1$. By (10), $z = 1$ means that $B = 2M/(n + n^2)$. Since $N_s^* = Bz^s$ and $dz/dB < 0$, we have

(1) If $B = 2M/(n + n^2)$, then $z = 1$ and $N_1^* = N_2^* = \cdots = N_n^* = B$;
(2) If $B < 2M/(n + n^2)$, then $z > 1$ and $N_1^* < N_2^* < \cdots < N_n^*$;
(3) If $B > 2M/(n + n^2)$, then $z < 1$ and $N_1^* > N_2^* > \cdots > N_n^*$.

Hence, the distribution of clusters at the equilibrium is monotonous:

Theorem 7. *The equilibrium* $(N_1^*, N_2^*, \ldots, N_n^*)$ *of* (9) *satisfies*:

(i) *If* $B < \frac{2M}{n(n+1)}$, *then* $N_1^* < N_2^* < \cdots < N_n^*$;
(ii) *If* $B = \frac{2M}{n(n+1)}$, *then* $N_1^* = N_2^* = \cdots = N_n^*$;
(iii) *If* $B > \frac{2M}{n(n+1)}$, *then* $N_1^* > N_2^* > \cdots > N_n^*$.

It follows from Theorem 7 that the monotonicity of the equilibrium varies as B moves through the critical value $2M/(n+n^2)$: as B is less than the value, the larger the size of clusters, the larger the number of the clusters; as B is larger than the value, the larger the size of clusters, the smaller the number of the clusters. The monotonicity of the equilibrium shows a natural phenomenon in the cluster evolution: if the ratio of attachment rate to the detachment rate is larger than a criteria value, then the larger the size of clusters, the larger the number of the clusters on the surface; if the ratio of attachment rate to the detachment rate is less than the criteria value, then the larger the size of clusters, the smaller the number of the clusters on the surface.

Figure 3 shows the monotonicity of the equilibrium where components N_i^* $(1 \leqslant i \leqslant 5)$ of equilibrium N^* are plotted as functions of B and $B = 10^{-6}$, 10^{-4}, 10^{-2}, $1, 10$, 10^2, 10^3, respectively. Let $M = 20$, $n = 5$. As $B < 4/3$, we have $N_5^* > N_4^* > N_3^* > N_2^* > N_1^*$; as $B = 4/3$, we have $N_5^* = N_4^* = N_3^* = N_2^* = N_1^* = 4/3$; as $B > 4/3$, we have $N_1^* > N_2^* > N_3^* > N_4^* > N_5^*$. The monotonicity of the equilibrium changes as parameter B moves through the critical value $4/3$.

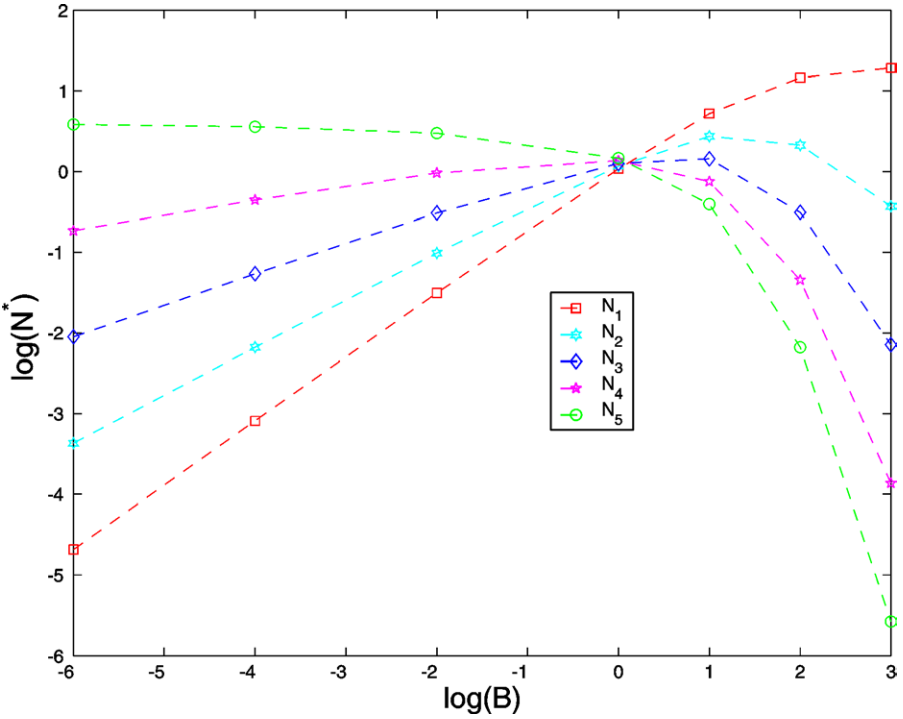

Fig. 3. Let $M = 20, n = 5$. Components $N_i^*(1 \leqslant i \leqslant 5)$ of N^* are plotted as functions of B and $B = 10^{-6}$, 10^{-4}, 10^{-2}, $1, 10$, 10^2, 10^3, respectively. As $B < 4/3$, we have $N_5^* > N_4^* > N_3^* > N_2^* > N_1^*$; as $B = 4/3$, we have $N_5^* = N_4^* = N_3^* = N_2^* = N_1^* = 4/3$; as $B > 4/3$, we have $N_1^* > N_2^* > N_3^* > N_4^* > N_5^*$. The monotonicity of the equilibrium changes as parameter B moves through the critical value $4/3$.

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