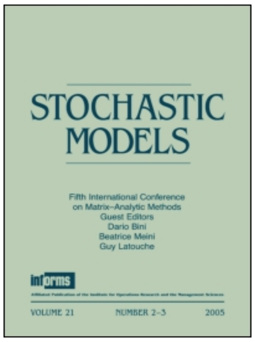

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Transient detailed balance and product form for reaction networks

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ABSTRACT
This paper explores the boundary of the set of reaction networks that have an exact transient (truncated) multidimensional Poisson or product-form distribution for the number of particles of different types. Motivated by the birth–death process, we introduce the notions of transient detailed balance and delay functions, and use these notions to obtain the novel transient product-form distribution in a coagulation-fragmentation process for polymers with a tree-like structure from that of the pure coagulation process.

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1. Introduction

1.1. Motivation

A *reaction network* is a Markov chain $\mathbf{X} = (X(t),\ t \geq 0)$ on a state-space $S \subseteq \mathbb{N}_0^N$, $N \in \mathbb{N}$, $\mathbb{N}_0 = \mathbb{N} \cup \{0\}$, with cardinality $|S| \geq 2$ and with transition rates $Q = (q(\mathbf{n}, \mathbf{n}'), \mathbf{n}, \mathbf{n}' \in S)$ given by

$$q(\mathbf{n}, \mathbf{n}') = \begin{cases} \displaystyle\sum_{\{\mathbf{g},\mathbf{g}' \in \mathbb{N}_0^N : \mathbf{n}-\mathbf{g}+\mathbf{g}'=\mathbf{n}'\}} q(\mathbf{g}, \mathbf{g}'; \mathbf{n}-\mathbf{g}), & \mathbf{n}' \neq \mathbf{n}, \\ \displaystyle -\sum_{\mathbf{n}' \neq \mathbf{n}} q(\mathbf{n}, \mathbf{n}'), & \mathbf{n}' = \mathbf{n}, \end{cases} \tag{1}$$

where

$$q(\mathbf{g}, \mathbf{g}'; \mathbf{m}) = \lambda(\mathbf{g}, \mathbf{g}') \prod_{k=1}^{N} \binom{m_k + g_k}{g_k}, \qquad \text{for all } \mathbf{g}, \mathbf{g}', \mathbf{m} \in \mathbb{N}_0^N, \tag{2}$$

and the *reaction rates* $\lambda : \mathbb{N}_0^N \times \mathbb{N}_0^N \to [0, \infty)$ are bounded. State $\mathbf{n} = (n_1, \ldots, n_N)$ represents the presence of n_k particles of type k, $k = 1, \ldots, N$, and $q(\mathbf{g}, \mathbf{g}'; \mathbf{m})$ represents the rate at which a batch of particles $\mathbf{g}$ reacts in state $\mathbf{m} + \mathbf{g}$ to form a batch of particles $\mathbf{g}'$. A reaction network is completely characterized by the reaction

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rates λ and the initial distribution $P(X(0) = \mathbf{n})$, $\mathbf{n} \in \mathbb{N}_0^N$, with induced state-space $S = \{\mathbf{n} \in \mathbb{N}_0^N \mid \exists t \geq 0 : P(X(t) = \mathbf{n}) > 0\}$.

The equilibrium distribution $\pi : S \to [0, 1]$ of reaction networks in physics often satisfies detailed balance:

$$\pi(\mathbf{n})q(\mathbf{n}, \mathbf{n}') = \pi(\mathbf{n}')q(\mathbf{n}', \mathbf{n}), \quad \text{for all } \mathbf{n}, \mathbf{n}' \in S.$$

A sufficient condition for detailed balance is the existence of non-negative coefficients $c_1, \ldots, c_N$ that satisfy the *equilibrium macroscopic reaction equations* (see Boucherie and van Dijk[5]):

$$\lambda(\mathbf{g}', \mathbf{g}) \prod_{k=1}^{N} \frac{c_k^{g_k'}}{g_k'!} = \lambda(\mathbf{g}, \mathbf{g}') \prod_{k=1}^{N} \frac{c_k^{g_k}}{g_k!}, \quad \text{for all } \mathbf{g}, \mathbf{g}' \in \mathbb{N}_0^N. \tag{3}$$

If these coefficients exist, then the equilibrium distribution is truncated multidimensional Poisson (see Boucherie and van Dijk[5]):

$$\pi(\mathbf{n}) = B \prod_{k=1}^{N} \frac{c_k^{n_k}}{n_k!}, \quad \mathbf{n} \in S, \tag{4}$$

where B is the normalizing constant, and c_i represents the mean number of particles of type i, $i = 1, \ldots, N$.

1.2. Contribution

This paper explores the boundary of the set of reaction networks that have a *transient* truncated multidimensional Poisson distribution:

$$P(X(t) = \mathbf{n}) = B(t) \prod_{k=1}^{N} \frac{c_k(t)^{n_k}}{n_k!}, \quad \mathbf{n} \in S, \quad t \geq 0, \tag{5}$$

for differentiable functions $B : [0, \infty) \to (0, \infty)$, $c_1, \ldots, c_N : [0, \infty) \to [0, \infty)$. This distribution is also referred to as *product-form* distribution, which is the term we will use in the sequel. The product-form distribution expresses independence of the particles.

Motivated by the birth–death process, we introduce the notions of *transient detailed balance*

$$\left(1 - \frac{dF(t)}{dt}\right) P(\mathbf{m} + \mathbf{g}, t)q(\mathbf{g}, \mathbf{g}', \mathbf{m}) = P(\mathbf{m} + \mathbf{g}', t)q(\mathbf{g}', \mathbf{g}, \mathbf{m}),$$

with *delay function* $F : [0, \infty) \to [0, \infty)$, from which we obtain the novel transient product-form distribution (5) for a coagulation-fragmentation process for polymers with a tree-like structure from that of the pure coagulation process. In addition, through several examples and counter-examples, we explore the boundary of the set of reaction networks that have an exact transient product-form distribution.

1.3. Reaction networks and product form in the literature

Reaction networks typically model chemical reactions and polymerization processes[10–12,20,24], physical systems[19], cloud formation[3], animal grouping[15,16,27], and networks of infinite server queues[13,18,21,26]. A reaction network may also be modeled as a stochastic Petri net[2,9,17,23,25]. For such networks, the transient distribution $P(X(t) = \mathbf{n}),\ \mathbf{n} \in S$, is of interest. For all except a few isolated cases for which the transient distribution is known in closed form, this distribution is (truncated) Poisson.

Widely applicable approximation methods for the transient distribution include the mean field approximation[22], the diffusion approximation[14], and the Ω-expansion[19]. Assuming the distribution is a product of the marginals, an approximation via linear differential equations for these marginals is obtained in Angius and Horváth[1]. To estimate their accuracy, these approximations also require exact results for the transient distribution.

There is ample literature on product form for the equilibrium or stationary distribution of Markov chains, see Boucherie and van Dijk[7] for an overview of results. Transient distributions for queueing networks of infinite server queues are studied in Refs.[13,18,21], and generalized in Ref.[26] to queueing networks consisting of infinite-server queues in the more general context of Poisson arrival location models. In Boucherie and Taylor[8], it is shown that the only single-routing queueing networks that have transient product-form distribution are networks of infinite server queues. For two particular clustering processes for polymers with a tree-like structure that have reaction rates proportional to the size of the polymers, the transient distribution is shown to be of product form[10,11]. These Markov chains seem to be the only cases for which product-form results for the transient distribution are available in the literature. This paper adds a few isolated cases of Markov chains with product-form transient distribution to the literature, and illustrates that additional cases that exhibit a truncated multidimensional Poisson distribution are unlikely.

1.4. Organization of the paper

In this paper, we investigate the transient distribution for reaction networks. We first state in Section 2 necessary and sufficient conditions for this to hold, and provide examples for these conditions from the literature: the single-routing network of infinite-server queues and coagulation processes with tree-like polymers. In Section 3, we define transient detailed balance and show how transient detailed balance can be used to find the transient product-form distribution in birth–death processes and in queueing networks consisting only of sources and sinks, and most notably in the coagulation-fragmentation process with tree-like polymers. Section 4 provides further examples and counter-examples to explore the boundary of the set of reaction networks that have transient product-form distribution.

2. Necessary and sufficient conditions for transient product form in reaction networks

This section provides our model and a technical lemma that is the starting point for our analysis as well as two examples from the literature that illustrate use of the technical lemma.

2.1. Model and technical lemma

Consider a reaction network, e.g., a Markov chain $\mathbf{X} = (X(t), t \geq 0)$ on a state-space $S \subseteq \mathbb{N}_0^N$, for some $N \in \mathbb{N}$, with transition rates given by (1) and (2) for some bounded function $\lambda : \mathbb{N}_0^N \times \mathbb{N}_0^N \to [0, \infty)$. Assume that $Q = (q(\mathbf{n}, \mathbf{n}'), \mathbf{n}, \mathbf{n}' \in S)$ is regular, and write $P(\mathbf{n}, t) = P(X(t) = \mathbf{n})$, for $\mathbf{n} \in S$ and $t \geq 0$. We write $\mathbf{e}_i$ for the ith unit vector in $\mathbb{R}^N$, and $\mathbf{e}_0 \in \mathbb{R}^N$ for the vector consisting only of zeros.

Suppose the initial distribution $P(X(0) = \mathbf{n}), \mathbf{n} \in S$, is a multidimensional truncated Poisson or product-form distribution (6). The following technical lemma provides a necessary and sufficient condition for $P(\mathbf{n}, t)$ to have product-form distribution *for all* $t \geq 0$.

Lemma 2.1. Assume that the distribution of $X(0)$ is given by

$$P_0(\mathbf{n}) = B_0 \prod_{k=1}^{N} \frac{\xi_k^{n_k}}{n_k!}, \tag{6}$$

for some $B_0 > 0,\ \xi_1, \ldots, \xi_N \geq 0$ and all $\mathbf{n} \in S$. If there are differentiable functions

$$B : [0, \infty) \to (0, \infty),\ c_1, \ldots, c_N : [0, \infty) \to [0, \infty)$$

such that, for all $\mathbf{n} \in S$,

$$\begin{aligned} &\frac{1}{B(t)} \frac{dB(t)}{dt} + \sum_{k=1}^{N} \frac{n_k}{c_k(t)} \frac{dc_k(t)}{dt} \\ &= \sum_{\mathbf{g}' \neq \mathbf{0}} \left\{ \lambda(\mathbf{g}', \mathbf{0}) \prod_{k=1}^{N} \frac{c_k(t)^{g'_k}}{g'_k!} - \lambda(\mathbf{0}, \mathbf{g}') \right\} + \sum_{\mathbf{g} \neq \mathbf{0}} \prod_{k=1}^{N} \frac{n_k!}{(n_k - g_k)!} \left(\frac{1}{c_k(t)} \right)^{g_k} \\ &\quad \times \sum_{\mathbf{g}'} \left\{ \lambda(\mathbf{g}', \mathbf{g}) \prod_{k=1}^{N} \frac{c_k(t)^{g'_k}}{g'_k!} - \lambda(\mathbf{g}, \mathbf{g}') \prod_{k=1}^{N} \frac{c_k(t)^{g_k}}{g_k!} \right\}, \end{aligned} \tag{7}$$

with initial conditions

$$c_k(0) = \xi_k, \quad k = 1, \ldots, N, \tag{8}$$

$$B(0) = B_0, \tag{9}$$

then

$$P(\mathbf{n}, t) = B(t) \prod_{k=1}^{N} \frac{c_k(t)^{n_k}}{n_k!}, \qquad \text{for all } \mathbf{n} \in S \text{ and all } t \geq 0. \tag{10}$$

Conversely, if (6) is the distribution of $X(0)$ and $P(\mathbf{n}, t)$ is of the form (10), then $B(t)$, $\{c_k(t)\}_{k=1}^N$ satisfy (7) for all $t \geq 0$, with initial conditions (8) and (9).

Proof. Because Q is regular, the transient probabilities $P(\mathbf{n}, t)$ of $\mathbf{X}$ are differentiable in t, for all $t \geq 0$ and $\mathbf{n} \in S$, right-handedly at 0, and are the minimal non-negative solution to the Kolmogorov *forward*, or *master* equations

$$\frac{dP(\mathbf{n}, t)}{dt} = \sum_{\mathbf{n}' \in S} \left\{ P(\mathbf{n}', t) q(\mathbf{n}', \mathbf{n}) - P(\mathbf{n}, t) q(\mathbf{n}, \mathbf{n}') \right\}, \quad \mathbf{n} \in S, \quad t \geq 0, \tag{11}$$

with initial distribution

$$P(\mathbf{n}, 0) = P_0(\mathbf{n}), \qquad \mathbf{n} \in S.$$

Replacing $q(\mathbf{n}, \mathbf{n}')$ and $q(\mathbf{n}', \mathbf{n})$ in (11) by (1) and (2) and inserting (10) in (11) implies that (10) solves (11) if $B, c_1, \ldots, c_N$ satisfy (7), (8), and (9). Conversely, if $P(\mathbf{n}, t)$ is of the form (10), then insertion in the forward equations (11) implies (7), (8), and (9). □

Condition (7) in Lemma 2.1 neither has a clear interpretation, nor can it be directly used to characterize the transient product-form distribution. The importance of this technical lemma lies in the observation that (7) contains a macroscopic equation for $B(t)$, $\{c_k(t)\}_{k=1}^N$, and the implications of this observation on the reaction rates that allow a transient product-form distribution (10). Condition (7) will be the basis for our analysis.

The right-hand side of (7) contains the form of the equilibrium macroscopic reaction equations (3). Moreover, in equilibrium the left-hand side of (7) equals zero, so that (7) shows that existence of a non-negative solution $\{c_k\}_{k=1}^N$ of the generalized version

$$\sum_{\mathbf{g}'} \left\{ \lambda(\mathbf{g}', \mathbf{g}) \prod_{k=1}^{N} \frac{c_k^{g_k'}}{g_k'!} - \lambda(\mathbf{g}, \mathbf{g}') \prod_{k=1}^{N} \frac{c_k^{g_k}}{g_k!} \right\} = 0, \quad \text{for all } \mathbf{g} \in \mathbb{N}_0^N, \tag{12}$$

of the equilibrium macroscopic reaction equations is sufficient for the equilibrium distribution to be of product form.

For systems that were shown in the literature to have a transient product-form distribution, the coefficients $\{c_k(t)\}_{k=1}^N$ satisfy a transient macroscopic equation, that, clearly, cannot contain information on the states $\mathbf{n} \in S$. To obtain a transient macroscopic equation from (7), observe that the second term in the left-hand side of (7) is linear in n_k, $k = 1, \ldots, N$, whereas the second term in the right-hand side is proportional to $n_k(n_k - 1) \cdots (n_k - g_k + 1)$ for all $\mathbf{g}$ that may be involved in a transition. As a consequence, in general, a solution $B(t)$, $\{c_k(t)\}_{k=1}^N$ of (7) that is independent of $\mathbf{n}$ will not exist. Lemma 2.1 is the starting point for the characterization of reaction rates λ that allow for a transient product-form distribution.

We now present two examples from the literature of systems that have a transient product-form distribution: the network of infinite-server queues and the pure coagulation process with tree-like polymers. These examples illustrate the technical condition (7) and provide the basis for the novel examples in Sections 3 and 4.

2.2. Number of particles in a transition conserved: Queueing networks

A network of single-routing infinite-server queues is an example of a reaction network in which the number of particles in each transition is conserved, i.e., in each transition one particle transforms into one particle of another type. Here, n_k represents the number of customers present at queue k, $k = 1, \dots, N$.

The reaction rate $\lambda(\mathbf{e}_0, \mathbf{e}_j)$ denotes the arrival rate to queue j, $\lambda(\mathbf{e}_i, \mathbf{e}_j)/\sum_{k=0}^{N}\lambda(\mathbf{e}_i, \mathbf{e}_k)$ is the probability that a customer departing from queue i joins queue j, and $\lambda(\mathbf{e}_i, \mathbf{e}_0)/\sum_{k=0}^{N}\lambda(\mathbf{e}_i, \mathbf{e}_k)$ is the probability that a customer departing from queue i leaves the system, $i, j = 1, \dots, N$. If $\mathbf{g}$ or $\mathbf{g}'$ is not a unit vector, or if $\mathbf{g} = \mathbf{g}' = \mathbf{e}_0$, then $\lambda(\mathbf{g}, \mathbf{g}') = 0$.

This transition structure implies that the second term in the right-hand side of (7) is proportional to n_k for all $\mathbf{g}$ that may be involved in a transition, and therefore both the second term in left-hand side and the right-hand side of (7) are now linear in each n_k. Thus, (7) can be factorized into $N + 1$ differential equations that are all independent of the state:

$$\frac{1}{B(t)}\frac{dB(t)}{dt} = \sum_{i=1}^{N}\{\lambda(\mathbf{e}_i, \mathbf{e}_0)c_i(t) - \lambda(\mathbf{e}_0, \mathbf{e}_i)\}, \tag{13}$$

$$\frac{dc_k(t)}{dt} = \sum_{j=0}^{N}\left\{\lambda(\mathbf{e}_j, \mathbf{e}_k)c_j(t) - \lambda(\mathbf{e}_k, \mathbf{e}_j)c_k(t)\right\}, \quad k = 1, \dots, N, \tag{14}$$

where $c_0(t) = 1$ for all $t \geq 0$, with initial conditions

$$B(0) = B_0,$$
$$c_k(0) = \xi_k, \quad k = 1, \dots, N.$$

Note that (14) can be interpreted as the Kolmogorov forward equations (11) for the Markov chain recording the position of a single customer at state-space $\{\mathbf{e}_i,\ i = 0, \dots, N\}$ with transition rates λ and transient distribution $c(\mathbf{e}_i)$, $i = 1, \dots, N$, implying existence of a non-negative solution of the differential equations (14).

Lemma 2.1 now implies that the solution $B, c_1, \dots, c_N$ of these differential equations gives a transient product-form distribution (10), provided the initial distribution is of the form (6). For the open queueing network at state-space $S = \mathbb{N}_0^N$, the normalizing constant is $B(t) = \exp[-\sum_{k=1}^{N} c_k(t)]$, so that particles are indeed independent. This result is established in Refs.[13,18,21], and is generalized in Ref.[26]

to queueing networks consisting of infinite-server queues in the more general context of Poisson arrival location models.

In contrast to these positive results, Boucherie and Taylor[8] consider both the closed and open general Kelly-Whittle network with transition rates

$$q(\mathbf{n}, \mathbf{n} - \mathbf{e}_i + \mathbf{e}_j) = \frac{\psi(\mathbf{n} - \mathbf{e}_i)}{\phi(\mathbf{n})}\lambda(\mathbf{e}_i, \mathbf{e}_j), \quad \mathbf{n} \in S,$$

where $S \subseteq \mathbb{N}_0^N$, $\psi : \mathbb{N}_0^N \to [0, \infty)$, $\phi : S \to (0, \infty)$, and show that for the network to have a transient product-form distribution it must be that $\psi = \phi$, and

$$\frac{\psi(\mathbf{n} - \mathbf{e}_i)}{\phi(\mathbf{n})} = n_i, \quad i = 1, \ldots, N,$$

i.e., the only single-routing queueing networks with transient product-form distribution are networks of infinite-server queues: *Reaction networks are the only networks in which each transition transforms one particle into one other particle and have transient product-form distribution.*

2.3. Number of particles in a transition not conserved: Clustering processes

A clustering process is a reaction network in which molecules consisting of atoms can cluster together to larger molecules (coagulation), or break into smaller molecules (fragmentation). Here, n_k represents the number of molecules in the system that consists of precisely k atoms, $k = 1, \ldots, N$. A well-studied clustering process is the case where at most two molecules can react simultaneously. The reaction rates $\lambda(\mathbf{g}, \mathbf{g}')$ have the form[10,11]

$$\lambda(\mathbf{e}_i + \mathbf{e}_j, \mathbf{e}_{i+j}) = K_{ij}, \quad 1 \leq i, j \leq i + j \leq N, \tag{15}$$

$$\lambda(\mathbf{e}_{i+j}, \mathbf{e}_i + \mathbf{e}_j) = F_{ij}, \quad 1 \leq i, j \leq i + j \leq N, \tag{16}$$

for given non-negative numbers $K_{ij}, F_{ij}, i, j \in \{1, \ldots, N\}$. Typical choices for K_{ij} are $K_{ij} = 1$ corresponding to linear polymers, and $K_{ij} = i + j$ and $K_{ij} = ij$ corresponding to polymers with a tree-like structure[12].

Transient product-form results (10) are derived in Refs.[10,11] for the pure coagulation process with the fragmentation coefficients F_{ij} equal zero for tree-like polymers, $K_{ij} = i + j$ or $K_{ij} = ij$ for all i, j, and mono-dispersed initial conditions, that is the system starts in state $N\mathbf{e}_1$, i.e., with only mono-mers present.

The number of atoms N does not change during transitions, which implies that the system admits a conservation-of-mass property:

$$\sum_{k=1}^{N} k n_k = N \text{ for all } \mathbf{n} \in S, \tag{17}$$

which immediately implies that the normalization constant is

$$B(t) = N! \quad \text{for all } t \geq 0,$$

and that (7) reduces to

$$\sum_{k=1}^{N} \frac{n_k}{c_k(t)} \frac{dc_k(t)}{dt} = \sum_{1\le i\le j\le N} \left\{ \frac{n_{i+j}}{c_{i+j}(t)} K_{ij} \frac{c_i(t)c_j(t)}{1+\delta_{ij}} - n_i(n_j - \delta_{ij}) K_{ij} \frac{1}{1+\delta_{ij}} \right\}, \tag{18}$$

where δ_{ij} is the Kronecker-delta that equals 1 if $i = j$ and 0 otherwise. Invoking conservation-of-mass (17), for $K_{ij} = i + j$ we find $\sum_{i,j} K_{ij} n_i n_j = \sum_i 2Nn_i$, and for $K_{ij} = ij$ we find $\sum_{i,j} K_{ij} n_i n_j = \sum_i iNn_i$. Then, both left-hand side and right-hand side of (18) are linear in n_k, so that we may reduce (18) into a set of N differential equations for $c_1(t), \ldots, c_N(t)$, that are all independent of the state $\mathbf{n}$. This is the key observation to derive the transient product-form distribution for this type of clustering processes. The macroscopic equations for $\{c_k(t)\}_{k=1}^N$ are, for $k = 1, \ldots, N$,

$$K_{ij} = ij : \frac{dc_k(t)}{dt} = \frac{1}{2} \sum_{i+j=k} ijc_i(t)c_j(t) - \frac{1}{2} k(N-k)c_k(t), \tag{19}$$

$$K_{ij} = i + j : \frac{dc_k(t)}{dt} = \frac{1}{2} \sum_{i+j=k} (i+j)c_i(t)c_j(t) - (N-k)c_k(t), \tag{20}$$

with initial conditions $c_k(0) = \delta_{k1}$, and have a unique non-negative solution, see Refs.[10,11], which implies that the transient distribution for these clustering processes has product form (10).

3. Transient detailed balance

Motivated by the birth–death process, this section introduces transient detailed balance, and novel transient product-form distributions. Most notably, we obtain the transient distribution for the coagulation-fragmentation process with tree-like polymers for the case $K_{ij} = i + j$.

3.1. Motivation: The birth–death process

Consider the birth–death process $\mathbf{X}_\mu$ with birth rates λ, death rates $n\mu$, and state-space $S = \mathbb{N}_0$ starting at time $t = 0$ in state $n = 0$. The transient distribution $P_\mu(n; t) = P(\mathbf{X}_\mu(t) = n)$, $n \in S$, is Poisson[28]:

$$P_\mu(n,t) = \frac{c_\mu(t)^n}{n!} \mathrm{e}^{-c_\mu(t)}, \qquad n = 0, 1, 2, \ldots,$$

with

$$c_\mu(t) = \frac{\lambda}{\mu}(1 - \mathrm{e}^{-\mu t}), \qquad t \ge 0.$$

For $\mu = 0$, $\mathbf{X}_0$ is the pure-birth process with birth rate λ, and $c_0(t) = \lambda t$.

The transient distributions for the birth–death and pure-birth process are related as

$$P_\mu(n, t) = P_0(n, F_\mu(t)),$$

with

$$F_\mu(t) = \frac{1}{\mu}(1 - e^{-\mu t}), \quad t \geq 0.$$

The function F_μ seems to express a *delay function*: the death rates $n\mu$ delay the pure-birth process.

Further observe that the birth–death process satisfies

$$\left(1 - \frac{dF_\mu(t)}{dt}\right) \lambda P_\mu(n, t) = (n+1)\mu P_\mu(n+1, t)$$

and that $\lim_{t\to\infty} 1 - \frac{dF_\mu(t)}{dt} = 1$: the detailed balance property carries over to the transient distribution, and reduces to detailed balance in equilibrium.

This section explores these observations in general reaction networks.

3.2. Transient detailed balance

Definition 3.1. *A reaction network* $\mathbf{X}$ *with reaction rates* $\lambda : \mathbb{N}_0^N \times \mathbb{N}_0^N \to [0, \infty)$ *is called one sided if, for all* $\mathbf{g}, \mathbf{g}' \in \mathbb{N}_0^N$, $\lambda(\mathbf{g}, \mathbf{g}') > 0$ *implies* $\lambda(\mathbf{g}', \mathbf{g}) = 0$.

Definition 3.2. *Let* $\mathbf{X}$ *be a one-sided reaction network with transient product-form distribution*

$$P(\mathbf{n}, t) = B(t) \prod_{k=1}^{N} \frac{c_k(t)^{n_k}}{n_k!}, \quad \mathbf{n} \in S, \quad t \geq 0. \tag{21}$$

Suppose that there are functions $G : [0, \infty) \to [0, \infty)$ *and* $\mu : \mathbb{N}_0^N \times \mathbb{N}_0^N \to [0, \infty)$ *such that, for all* $\mathbf{g}, \mathbf{g}' \in \mathbb{N}_0^N$ *and all* $t \geq 0$,

$$G(t) \prod_{k=1}^{N} \frac{c_k(t)^{g_k}}{g_k!} \lambda(\mathbf{g}, \mathbf{g}') = \prod_{k=1}^{N} \frac{c_k(t)^{g'_k}}{g'_k!} \mu(\mathbf{g}', \mathbf{g}). \tag{22}$$

The reaction network $\mathbf{X}_r$ *with reaction rates*

$$\lambda_r(\mathbf{g}, \mathbf{g}') = \begin{cases} \lambda(\mathbf{g}, \mathbf{g}') & \text{if} \quad \lambda(\mathbf{g}, \mathbf{g}') > 0, \\ \mu(\mathbf{g}, \mathbf{g}') & \text{if} \quad \lambda(\mathbf{g}', \mathbf{g}) > 0, \end{cases} \tag{23}$$

is then called the two-sided version of $\mathbf{X}$ *corresponding to* G *and* μ.

The functions G and μ may be obtained by substitution of λ and $\{c_k(t)\}_{k=1}^N$ for the one-sided network into (22) similar to the steps used for networks in equilibrium, see Refs.[12,20].

The following theorem gives conditions for the two-sided version $\mathbf{X}_r$ of $\mathbf{X}$ to be a "delayed version" of $\mathbf{X}$, in particular $P_r(\mathbf{n}, t) = P(\mathbf{n}, F(t))$ for all $\mathbf{n}$ and all $t \geq 0$, provided that (24) has a solution. We will call function F the *delay function*.

Theorem 3.1. *Let* $\mathbf{X}$ *be a one-sided reaction network with transient product-form distribution (21), and let* $\mathbf{X}_r$ *be the two-sided version of* $\mathbf{X}$ *corresponding to* G *and* μ*. If there exists a function* $F : [0, \infty) \to [0, \infty)$ *that satisfies*

$$\frac{dF(t)}{dt} = 1 - G(F(t)), \qquad t \geq 0, \tag{24}$$

with initial condition

$$F(0) = 0,$$

then the transient distribution P_r *of* $\mathbf{X}_r$ *is*

$$P_r(\mathbf{n}, t) = P(\mathbf{n}, F(t)), \qquad \mathbf{n} \in S. \tag{25}$$

Proof. Write $\mathcal{T}_{\mathbf{X}} = \{(\mathbf{g}, \mathbf{g}') \in \mathbb{N}_0^N \times \mathbb{N}_0^N : \lambda(\mathbf{g}, \mathbf{g}') > 0\}$. For the two-sided version $\mathbf{X}_r$ corresponding to G and μ, we may now insert $P_r(\mathbf{n}, t) = P(\mathbf{n}, F(t))$ into (7) for the two-sided process:

$$\begin{aligned}
&\frac{1}{B(F(t))}\frac{dB(F(t))}{dF(t)}\frac{dF(t)}{dt} + \sum_{k=1}^{N} \frac{n_k}{c_k(F(t))}\frac{dc_k(F(t))}{dF(t)}\frac{dF(t)}{dt} \\
&= \sum_{(\mathbf{g}',\mathbf{g})\in\mathcal{T}_{\mathbf{X}}} \prod_{k=1}^{N} \frac{n_k!}{(n_k-g_k)!}\left(\frac{1}{c_k(F(t))}\right)^{g_k} \\
&\quad\times \left\{\prod_{k=1}^{N}\frac{c_k(F(t))^{g_k'}}{g_k'!}\lambda(\mathbf{g}',\mathbf{g}) - \prod_{k=1}^{N}\frac{c_k(F(t))^{g_k}}{g_k!}\mu(\mathbf{g},\mathbf{g}')\right\} \\
&\quad - \sum_{(\mathbf{g},\mathbf{g}')\in\mathcal{T}_{\mathbf{X}}} \prod_{k=1}^{N} \frac{n_k!}{(n_k-g_k)!}\left(\frac{1}{c_k(F(t))}\right)^{g_k} \\
&\quad\times \left\{\prod_{k=1}^{N}\frac{c_k(F(t))^{g_k}}{g_k!}\lambda(\mathbf{g},\mathbf{g}') - \prod_{k=1}^{N}\frac{c_k(F(t))^{g_k'}}{g_k'!}\mu(\mathbf{g}',\mathbf{g})\right\}. \\
&= \sum_{(\mathbf{g}',\mathbf{g})\in\mathcal{T}_{\mathbf{X}}} \prod_{k=1}^{N} \frac{n_k!}{(n_k-g_k)!}\left(\frac{1}{c_k(F(t))}\right)^{g_k}\{1-G(F(t))\}\prod_{k=1}^{N}\frac{c_k(F(t))^{g_k'}}{g_k'!}\lambda(\mathbf{g}',\mathbf{g}) \\
&\quad - \sum_{(\mathbf{g},\mathbf{g}')\in\mathcal{T}_{\mathbf{X}}} \prod_{k=1}^{N} \frac{n_k!}{(n_k-g_k)!}\left(\frac{1}{c_k(F(t))}\right)^{g_k}\{1-G(F(t))\}\prod_{k=1}^{N}\frac{c_k(F(t))^{g_k}}{g_k!}\lambda(\mathbf{g},\mathbf{g}').
\end{aligned}$$

Condition (24) makes sure that the terms $\frac{dF(t)}{dt}$ cancel out, and the above expression reduces to (7) for the one-sided process $\mathbf{X}$ evaluated in $F(t)$. The initial condition $F(0) = 0$ implies $P_r(\mathbf{n}, 0) = P(\mathbf{n}, 0)$. This completes the proof. □

Observe that (22) carries over to an expression for the transient distribution that we call the *transient detailed balance* property when we evaluate this equation at *delayed time* $F(t)$ and applying (24):

$$\left(1 - \frac{dF(t)}{dt}\right) P_r(\mathbf{m}+\mathbf{g}, t) q_r(\mathbf{g}, \mathbf{g}', \mathbf{m}) = P_r(\mathbf{m}+\mathbf{g}', t) q_r(\mathbf{g}', \mathbf{g}, \mathbf{m}), \tag{26}$$

for all $\mathbf{g}$, $\mathbf{g}'$, $\mathbf{m}$, and $t \geq 0$, where q_r is defined in (2), which, in turn, implies

$$\left(1 - \frac{dF(t)}{dt}\right) P_r(\mathbf{n}, t) q_r(\mathbf{n}, \mathbf{n}') = P_r(\mathbf{n}', t) q_r(\mathbf{n}', \mathbf{n}), \quad \text{for all } \mathbf{n} \in S. \tag{27}$$

We now present three examples of processes for which the transient product-form distribution may be obtained from Theorem 3.1: the birth–death process, and the generalized star network are examples of single-routing networks as presented in Example 2.2, and the coagulation-fragmentation process with tree-like polymers and $K_{ij} = i + j$ of the clustering process of Example 2.3. The latter two examples are novel cases that have a transient product-form distribution.

3.3. Example: Birth–death process

The birth–death process $\mathbf{X}_\mu$ from Section 3.1 is the two-sided version of the pure-birth process $\mathbf{X}_0$, with $G(t) = \mu t$ and $\mu(1,0) = \mu$. The delay function $F(t)$ in (25) is equal to $F(t) = (1 - e^{-\mu t})/\mu$. Observe that $\lim_{\mu \downarrow 0} F(t) = t$ yielding the pure-birth process.

3.4. Example: Generalized star network

Consider a reaction network on $\mathbb{N}_0^N$ characterized by

$$\lambda(\mathbf{e}_i, \mathbf{e}_j) = p_{ij}, \quad i \in R, \; j \notin R, \tag{28}$$

where R is a proper non-empty subset of $\{1, \ldots, N\}$ and p_{ij}, $i \in R$, $j \notin R$, satisfies $\sum_{j \notin R} p_{ij} = 1$ and $p_{ij} \geq 0$ for all $i \in R$, $j \notin R$. This models a communication network in which some nodes are strictly sending (the set R) and other nodes are strictly receiving (the complement of R). This reaction network has transient product-form distribution (21), where

$$\frac{dc_k(t)}{dt} = -c_k(t), \quad k \in R,$$
$$\frac{dc_k(t)}{dt} = \sum_{i \in R} c_i(t) p_{ik}, \quad k \notin R,$$

with solution

$$c_k(t) = c_k(0) \mathrm{e}^{-t}, \quad k \in R,$$
$$c_k(t) = \sum_{i \in R} c_i(0) p_{ik} (1 - \mathrm{e}^{-t}), \quad k \notin R,$$

under the natural assumption that $c_k(0) > 0$ iff $k \in R$.

From (23), we find that

$$G(t) = \mathrm{e}^t - 1$$

and

$$\mu(\mathbf{e}_j, \mathbf{e}_i) = \frac{c_i(0)p_{ij}}{\sum_{l\in R} c_l(0)p_{lj}}, \quad i \in R,\ j \notin R,$$

constitutes a two-sided version of $\mathbf{X}$. The delay function satisfies

$$\frac{dF(t)}{dt} = 2 - \mathrm{e}^{F(t)}, \quad t \geq 0,$$

and $F(0) = 0$.

3.5. Example: Coagulation fragmentation with $K_{ij} = i + j$

For coagulation-fragmentation processes corresponding to the pure coagulation process of Example 2.3, the fragmentation coefficients F_{ij} are commonly defined via the equilibrium macroscopic reaction equations (3), see, e.g., Ernst[12]. The fragmentation coefficients for the tree-like structures obtained by $K_{ij} = ij$ or $K_{ij} = i + j$ are

$$F_{ij} = \tau \frac{1}{1+\delta_{ij}} \frac{(i+j)!}{i!\,j!} \left(\frac{i}{i+j}\right)^{i-1} \left(\frac{j}{i+j}\right)^{j-1}, \tag{29}$$

where $\tau > 0$ is the ratio of the rate at which bonds between molecules are broken and the rate at which bonds are formed. Recall from Example 2.3, also see Van Dongen[11], that the clustering process $\mathbf{X}$ with $K_{ij} = i + j$ and with mono-dispersed initial conditions $\mathbf{X}(0) = N\mathbf{e}_1$ has transient product form (21) with $B(t) = N!$. The solution of the macroscopic equations (20) is[11]

$$c_k(t) = \frac{k^{k-1}}{k!} \left(\frac{1 - \exp[-t]}{N}\right)^{k-1} \exp\left[-\frac{(N-k)t}{N}\right], \quad t \geq 0, \quad k = 1, \ldots, N.$$

The pure coagulation process is a one-sided process. We may construct the two-sided version corresponding to G and μ invoking (22) of Definition 3.2. Since $\lambda(\mathbf{g}, \mathbf{g}') = i + j$ if $\mathbf{g} = \mathbf{e}_i + \mathbf{e}_j$, $\mathbf{g}' = \mathbf{e}_{i+j}$, $1 \leq i, j \leq i + j \leq N$, and $\lambda(\mathbf{g}, \mathbf{g}')$ is zero otherwise, G and μ should satisfy

$$G(t)c_i(t)c_j(t)(i+j) = c_{i+j}(t)\mu(\mathbf{e}_{i+j}, \mathbf{e}_i + \mathbf{e}_j), \quad 1 \leq i, j \leq i + j \leq N. \tag{30}$$

This is established precisely if

$$\mu(\mathbf{e}_{i+j}, \mathbf{e}_i + \mathbf{e}_j) = F_{ij}$$

and

$$G(t) = \tau(\exp(t) - 1)/N.$$

The delay function $F(t)$ solves

$$\frac{dF(t)}{dt} = 1 - \frac{\tau}{N}(\exp(F(t)) - 1)$$

with initial condition $F(0) = 0$. The solution is

$$F(t) = \log\left[\frac{1+\tau/N}{\tau + \exp\left[-\left(1+\tau/N\right)t\right]}\right]. \tag{31}$$

The coagulation-fragmentation process $\mathbf{X}_r$ has transient product-form distribution and satisfies transient detailed balance (26).

4. Further examples and counter-examples

This section provides additional examples and counter-examples to explore the boundary of the set of reaction networks that have a transient product-form distribution. The first example, the batch-routing network of infinite-server queues is argued not to have a product form. Then, the coagulation process involving three molecules in each reaction and coagulation coefficients $K_{ijk} = ijk$ is shown to have a product form, but other coagulation processes with tree-like are shown not to have product form. The final example considers the tree-like coagulation-fragmentation process with $K_{ij} = ij$. This process is shown not to satisfy transient detailed balance, and therefore does not have a transient product-form distribution.

4.1. Batch-routing queueing networks

A batch-routing queueing network generalizes the single-routing network of Example 2.2 to allow a batch of customers to be transformed into another batch containing the same number of customers. A special case is the network with transition rates (1) and (2). If a non-negative solution $\{c_k\}_{k=1}^N$ exists of the batch traffic equations (12), then the network has a product-form equilibrium distribution (4), see Boucherie and van Dijk[6].

A natural candidate for the transient distribution to have product form is the batch-routing queueing network in which all customers in a transition route independently. The reaction rates for this network are[6]

$$\lambda(\mathbf{g}, \mathbf{g}') = \sum_{g_0=0}^{\infty} \frac{\gamma^{g_0}}{g_0!} e^{-\gamma} \sum_{G} \prod_{i=0}^{N} \binom{g_i}{g_{i1}, \ldots, g_{iN}} \prod_{j=0}^{N} p_{ij}^{g_{ij}}, \tag{32}$$

where

$$G = \left\{ \begin{array}{lll} g_{ij},\ i, j = 0, \ldots, N : & g_{ij} \in \mathbb{N}_0,\ g_{ij} = 0 \text{ if} & p_{ij} = 0, g_{00} = 0, \\ & \sum_{j=0}^{N} g_{ij} = g_i, & i = 0, \ldots, N, \\ & \sum_{i=0}^{N} g_{ij} = g_j', & j = 1, \ldots, N \end{array} \right\}$$

with $\gamma \geq 0$, $p_{ij} \geq 0$, $\sum_{j=0}^{N} p_{ij} = 1$, and $p_{00} = 0$, for all $i, j = 0, \ldots, N$. The interpretation of (32) is that g_i customers depart from queue $i = 1, \ldots, N$; each of these customers join queue $j = 1, \ldots, N$ with probability p_{ij} and leave the system with probability p_{i0}; in addition, a Poisson(γ) number of new arriving customers joins queue j with probability p_{0j}, $j = 1, \ldots, N$. The second summation is over all possible ways to redistribute departing customers $\mathbf{g}$ and newly arrived customers g_0 over the queues such that $\mathbf{g}'$ represents the number of arriving customers at all queues. If $\gamma = p_{0j} = p_{i0} = 0$ for all $1 \leq i, j \leq N$, the network is called closed; we then interpret $\gamma^{g_0} = 0^0 := 1$ for $g_0 = 0$.

Intuitively, as customers under (32) route independently, it may seem that the product-form result for the transient distribution of the single-routing case carries over to the batch-routing case. The reasoning would be that if we start the network empty at time $t = 0$ and a batch arrival occurs after exponential time with mean 1, then at the arrival epoch of the batch the distribution is multidimensional Poisson. At the next transition epoch, we independently sample customers from the queues (random thinning of Poisson random variables) and independently distribute these customers over the queues (adding Poisson random variables), so that the distribution remains Poisson. Adding a Poisson batch of new arrivals to this procedure does not seem to affect the outcome. Unfortunately, this reasoning is, in \al, false. To see this, consider the one-dimensional case, with Poisson batches with mean γ arriving according to a Poisson process with rate 1, but no departures. Clearly, conditioning on the number of arrival epochs, say k, yields a Poisson distribution with mean $k\gamma$. At a fixed time $t > 0$, however, the distribution is a random sum of Poisson random variables with mean γ, but this distribution is not Poisson.

Note that the *discrete-time* reaction process $\mathbf{X^d}$ with state-space $S = \mathbb{N}_0^N$ starting empty at $t = 0$ with transition *probabilities*

$$p(\mathbf{n}, \mathbf{n}') = \sum_{\{\mathbf{g}, \mathbf{g}', \mathbf{m} \in \mathbb{N}_0^N : \mathbf{m}+\mathbf{g}=\mathbf{n},\ \mathbf{m}+\mathbf{g}'=\mathbf{n}'\}} \prod_{k=1}^{N} \binom{m_k + g_k}{g_k} p_k^{g_k} (1 - p_k)^{m_k} \lambda(\mathbf{g}, \mathbf{g}')$$

where p_k is the probability that a particle of type k is selected in the reaction, and with λ given in (32), has transient Poisson distribution for all $t = 0, 1, 2, \ldots$:

$$P(\mathbf{X^d}(t) = \mathbf{n}) = \prod_{k=1}^{N} \frac{c_k(t)^{n_k}}{n_k!} \mathrm{e}^{-c_k(t)}, \quad \mathbf{n} \in S,$$

which can readily be concluded as the conditioning argument used above is valid in discrete-time, or by insertion in the Chapman–Kolmogorov equations. The means $c_k(t)$ are, for $k = 1, \ldots, N$, recursively obtained from

$$c_k(0) = 0,$$
$$c_k(t+1) = c_k(t)(1 - p_k) + \gamma p_{0k} + \sum_{i=1}^{N} c_i(t) p_i p_{ik}, \quad t = 0, 1, 2, \ldots$$

For the continuous-time batch-routing case, the intuitive reasoning for the one-dimensional case seems to exclude transient product form. The following lemma provides a counter-example that illustrates that the transient distribution of the closed two queue network containing two customers is not a product-form distribution unless the system starts in equilibrium.

Lemma 4.1 (Counter-example). Consider a closed batch-routing queueing network with two queues and two customers. If $\lambda(2\mathbf{e}_1, 2\mathbf{e}_2) > 0$ or $\lambda(2\mathbf{e}_2, 2\mathbf{e}_1) > 0$, and the system does not start in equilibrium, then $P(\mathbf{n}, t)$ does not have product form for all $t \geq 0$.

Proof. The differential equations (7) applied to states $2\mathbf{e}_1$, $\mathbf{e}_1 + \mathbf{e}_2$, and $2\mathbf{e}_2$, respectively, reduce to

$$\frac{1}{B(t)}\frac{dB(t)}{dt} + \frac{2}{c_1(t)}\frac{dc_1(t)}{dt} = 2\lambda(\mathbf{e}_2, \mathbf{e}_1)\frac{c_2(t)}{c_1(t)} + \lambda(2\mathbf{e}_2, 2\mathbf{e}_1)\frac{c_2(t)^2}{c_1(t)^2} - 2\lambda(\mathbf{e}_1, \mathbf{e}_2) - \lambda(2\mathbf{e}_1, 2\mathbf{e}_2), \tag{33}$$

$$\frac{2}{B(t)}\frac{dB(t)}{dt} + \frac{2}{c_1(t)}\frac{dc_1(t)}{dt} + \frac{2}{c_2(t)}\frac{dc_2(t)}{dt} = 2\lambda(\mathbf{e}_1, \mathbf{e}_2)\frac{c_1(t)}{c_2(t)} + 2\lambda(\mathbf{e}_2, \mathbf{e}_1)\frac{c_2(t)}{c_1(t)} - 2\lambda(\mathbf{e}_2, \mathbf{e}_1) - 2\lambda(\mathbf{e}_1, \mathbf{e}_2), \tag{34}$$

$$\frac{1}{B(t)}\frac{dB(t)}{dt} + \frac{2}{c_2(t)}\frac{dc_2(t)}{dt} = 2\lambda(\mathbf{e}_1, \mathbf{e}_2)\frac{c_1(t)}{c_2(t)} + \lambda(2\mathbf{e}_1, 2\mathbf{e}_2)\frac{c_1(t)^2}{c_2(t)^2} - 2\lambda(\mathbf{e}_2, \mathbf{e}_1) - \lambda(2\mathbf{e}_2, 2\mathbf{e}_1). \tag{35}$$

Adding (33) to (35) and subtracting (34), we obtain

$$\lambda(2\mathbf{e}_1, 2\mathbf{e}_2)\left(\frac{c_2(t)^2}{c_1(t)^2} - 1\right) + \lambda(2\mathbf{e}_2, 2\mathbf{e}_1)\left(\frac{c_1(t)^2}{c_2(t)^2} - 1\right) = 0,$$

which implies that $c_2(t)/c_1(t)$ is constant for all $t \geq 0$. This is only possible if the system starts in equilibrium. □

4.2. Coagulation processes with tree-like polymers involving at least three molecules

Analogous to batch-routing queueing networks we can consider d-coagulation processes, where in each transition d molecules react. We first extend the pure 2-coagulation process (15) with $K_{ij} = ij$, $F_{ij} = 0$, to $d = 3$: the only non-zero values of $\lambda(\mathbf{g}, \mathbf{g}')$ are given by

$$\lambda(\mathbf{e}_i + \mathbf{e}_j + \mathbf{e}_k, \mathbf{e}_{i+j+k}) = ijk, \quad 1 \leq i, j, k \leq i + j + k \leq N, \tag{36}$$

for some $N \in \mathbb{N}$. The state space of this system is $S := \{\mathbf{n} \in \mathbb{N}_0^N \mid \sum_{k=1}^N kn_k = N\}$.

Lemma 4.2. If there are $\xi_1, \dots, \xi_N \geq 0$ and differentiable functions $c_1, \dots, c_N : [0, \infty) \to [0, \infty)$ such that for all $t \geq 0$ and all $\ell = 1, \dots, N$,

$$\frac{dc_\ell(t)}{dt} = \frac{1}{6} \sum_{i,j,k \in \mathbf{N}: i+j+k=\ell} ijkc_i(t)c_j(t)c_k(t) - \frac{\ell}{6}(N-\ell)(N-2\ell)c_\ell(t),$$

$$c_\ell(0) = \xi_\ell,$$

with initial distribution (6) with $B_0 = N!$, then the reaction network has transient product form (10) with $B(t) = N!$.

Proof. The crux of the proof is to show that the right-hand side of (7) is linear in each n_k; the differential equations can then be decomposed into an equation for each $k = 1, \dots, N$. To this end, write

$$S(\mathbf{n}) := \sum_{(\mathbf{g},\mathbf{g}'):\lambda(\mathbf{g},\mathbf{g}')>0} \lambda(\mathbf{g}, \mathbf{g}') \prod_{k=1}^{N} \binom{n_k}{g_k},$$

and observe that the right-hand side of (7) equals

$$\sum_{i=1}^{N} \frac{n_i}{c_i(t)} \sum_{\mathbf{g}':\lambda(\mathbf{g}',\mathbf{e}_i)>0} \lambda(\mathbf{g}', \mathbf{e}_i) \prod_{k=1}^{N} \frac{c_k(t)^{g'_k}}{g'_k!} - S(\mathbf{n}). \tag{37}$$

Note that

$$S(\mathbf{n}) = \sum_i \lambda(3\mathbf{e}_i, \mathbf{e}_{3i}) \binom{n_i}{3} + \sum_i \sum_{j \neq i} \lambda(2\mathbf{e}_i + \mathbf{e}_j, \mathbf{e}_{2i+j}) \binom{n_i}{2} \binom{n_j}{1}$$
$$+ \sum_i \sum_{j \neq i} \sum_{k \neq j, k \neq i} \lambda(\mathbf{e}_i + \mathbf{e}_j + \mathbf{e}_k, \mathbf{e}_{i+j+k}) n_i n_j n_k. \tag{38}$$

The first term on the right-hand side of (38) equals

$$\sum_{(i,j,k):i=j=k} ijkn_i(n_j - \delta_{ji})(n_k - 2\delta_{ki}). \tag{39}$$

The third term on the right-hand side of (38) equals

$$\frac{1}{6} \sum_{(i,j,k):i \neq j \neq k \neq i} ijkn_i(n_j - \delta_{ji})(n_k - 2\delta_{ki}); \tag{40}$$

the factor $\frac{1}{6}$ arises because of symmetry. By expanding $n_i - 1 = (n_i - 1)/3 + (n_i - 2)/3 + n_i/3$ and relabeling, the second term on the right-hand side of (38) can be written as

$$\sum_i \sum_{j \neq i} i^2 j \frac{1}{2} n_i n_j (n_i - 1)$$
$$= \frac{1}{6} \sum_i \sum_{j \neq i} i^2 jn_i n_j (n_i - 1) + \frac{1}{6} \sum_i \sum_{j \neq i} i^2 jn_i n_j (n_i - 2) + \frac{1}{6} \sum_i \sum_{j \neq i} i^2 jn_i n_j (n_i)$$

$$= \frac{1}{3} \sum_{(i,j,k):i=j\neq k} i^2 k \frac{n_i n_k (n_i - 1)}{2}$$

$$+ \frac{1}{3} \sum_{(i,j,k):i=k\neq j} i^2 j \frac{n_i n_j (n_i - 2)}{2} + \frac{1}{3} \sum_{(i,j,k):i\neq j=k} i j^2 n_j n_i n_j$$

$$= \frac{1}{6} \sum_{(i,j,k):i=j\neq k \text{ or } i=k\neq j \text{ or } i\neq j=k} ijk n_i (n_j - \delta_{ji})(n_k - 2\delta_{ki}). \tag{41}$$

Combining equations (39), (40), and (41), and using conservation of mass, we obtain

$$\begin{aligned} S(\mathbf{n}) &= \frac{1}{6} \sum_i \sum_j \sum_k ijk n_i (n_j - \delta_{ji})(n_k - 2\delta_{ki}) \\ &= \frac{1}{6} \sum_i i n_i \left(\sum_j j n_j - i \right) \left(\sum_k k n_k - 2i \right) \\ &= \frac{1}{6} \sum_i i(N - i)(N - 2i) n_i. \end{aligned} \tag{42}$$

In addition, in (37) we can write

$$\sum_{\mathbf{g}':\lambda(\mathbf{g}',\mathbf{e}_\ell)>0} \lambda(\mathbf{g}', \mathbf{e}_\ell) \prod_{k=1}^{N} \frac{c_k(t)^{g'_k}}{g'_k!} = \frac{1}{6} \sum_{i,j,k\in\mathbf{N}:i+j+k=\ell} ijk c_i(t) c_j(t) c_k(t); \tag{43}$$

(the term 1/6 is explained by observing that terms of the form $\lambda(2\mathbf{e}_n + \mathbf{e}_m, \mathbf{e}_{2n+m}) \frac{c_n(t)^2 c_m(t)}{2}$, $n \neq m$, occur three times in the right-hand side sum, and terms of the form $\lambda(\mathbf{e}_n + \mathbf{e}_m + \mathbf{e}_p, \mathbf{e}_{n+m+p}) c_n(t) c_m(t) c_p(t)$, $n \neq m \neq p \neq n$, occur six times in the right-hand side sum).

Combining (7), (37), (42), and (43) completes the proof. □

The appearance of a transient product-form distribution in this higher dimensional coagulating system turns out to be rather exceptional. The reason it holds is that the term $S(\mathbf{n})$ in (37), a cubic polynomial in $n_1, \ldots, n_N$, turns out to reduce to a linear polynomial because of the conservation-of-mass property, and as a result the traffic equations (7) reduce to N state-independent differential equations. This reasoning cannot be extended to d-coagulating systems of the form

$$\lambda(\mathbf{g}, \mathbf{e}_{\sum_k g_k}) = \prod_k k^{g_k},$$

with $d \geq 4$, neither to d-coagulating systems of the form

$$\lambda(\mathbf{g}, \mathbf{e}_{\sum_k g_k}) = \sum_k k g_k,$$

with $d \geq 3$, which contains the case $\lambda(\mathbf{e}_i + \mathbf{e}_j + \mathbf{e}_k, \mathbf{e}_{i+j+k}) = i + j + k$. For these systems, $S(\mathbf{n})$ is not linear in $\mathbf{n}$. The derivations required to show this are long but elementary, and may be obtained from the authors.

4.3. Coagulation fragmention with $K_{ij} = ij$

It is shown in Refs.[10,11] that the clustering process $\mathbf{X}$ with $K_{ij} = ij$ and with mono-dispersed initial conditions $\mathbf{X}(0) = N\mathbf{e}_1$ has transient product form

$$P(\mathbf{n}, t) = N! \prod_{k=1}^{N} \frac{a_k(t)^{n_k}}{n_k!},$$

for all $\mathbf{n} \in S = \{\mathbf{n} \in \mathbb{N}_0^N \mid \sum_{k=1}^{N} kn_k = N\}$, with

$$a_k(t) = \sum_{j=1}^{k} \frac{(-1)^{j-1}}{j} \sum_{n_1+\cdots+n_j=k,\ n_i \geq 1} q_{n_1}(t) \cdots q_{n_j}(t),$$

$$q_n(t) = \frac{1}{n!} \exp\left[-\frac{1}{2}nt(1 - n/N)\right].$$

Thus, for transient detailed balance, it must be that functions $G(t)$ and $\mu(\mathbf{e}_{i+j}, \mathbf{e}_i + \mathbf{e}_j)$ exist that satisfy

$$G(t)a_i(t)a_j(t)(ij) = a_{i+j}(t)\mu(\mathbf{e}_{i+j}, \mathbf{e}_i + \mathbf{e}_j), \quad 1 \leq i, j \leq i + j \leq N. \tag{44}$$

For the case $N = 3$, i.e., the system starting with three mono-mers, we have only three cases for i, j: $i = j = 1$ or $i = 1, j = 2$ or $i = 2, j = 1$. Insertion of $a_1(t)$ and $a_2(t)$ in (44) readily shows that functions $G(t)$ and $\mu(\mathbf{e}_{i+j}, \mathbf{e}_i + \mathbf{e}_j)$ do not exist. As the equation (44) for the three cases for i, j: $i = j = 1$ or $i = 1, j = 2$ or $i = 2, j = 1$ must be part of the system of equations (44) for all N, we conclude that the clustering process with $K_{ij} = ij$ does not satisfy transient detailed balance.

5. Concluding remarks

This paper has explored the boundary of the set of reaction networks that have an exact transient (truncated) multidimensional Poisson or product-form distribution for the number of particles of different types. From the notion of transient detailed balance that generalizes detailed balance to the transient distribution, we have obtained the novel transient product-form distribution of the coagulation-fragmentation process for polymers with a tree-like structure as a delayed version of that distribution for the pure coagulation process. Except for a few additional linear systems that have transient product form reported in this paper, via counter-examples we have indicated that appearance of the specific (truncated) multidimensional Poisson or product-form distribution seems to be restricted to these linear systems.

Detailed balance has had a huge impact on the analysis of the equilibrium distribution of stochastic networks. The results of the paper have been based on the particular multidimensional Poisson or product-form transient distribution. This is a natural first candidate for the transient distribution, as is also apparent from the results reported in Boucherie and Taylor[8]. It is interesting for further studies to investigate other candidates for the transient distribution. A starting point might be transient detailed balance.

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