The Becker-Döring process: law of large numbers and non-equilibrium potential

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July 31, 2018

Abstract

In this note, we prove a *law of large numbers* for an infinite chemical reaction network for phase transition problems called the stochastic Becker-Döring process. Under a general condition on the rate constants we show the convergence in law and pathwise convergence of the process towards the deterministic Becker-Döring equations. Moreover, we prove that the non-equilibrium potential, associated to the stationary distribution of the stochastic Becker-Döring process, approaches the relative entropy of the deterministic limit model. Thus, the phase transition phenomena that occurs in the infinite dimensional deterministic model is also present in the finite stochastic model.

Keywords: Becker-Döring; infinite-dimensional reaction network; law of large numbers; non-equilibrium potential; entropy.
AMS MSC 2010: 60J75; 60B12; 28D20.

1 Introduction

The Becker-Döring model represents time evolution of spatially homogeneous clusters of particles. Cluster sizes change following two simple rules: they may grow by adding particles one-by-one or shrink by losing particles one-by-one. Denoting a cluster of $i \geq 1$ particles by C_i, we can summarize the model by a simple infinite reaction network:

$$C_1 + C_i \underset{b_{i+1}}{\overset{a_i}{\rightleftharpoons}} C_{i+1}\ , \quad i \geq 1,$$

where a_i and b_{i+1} are the size-dependent reaction rate constants. This model was originally formulated as an infinite set of ordinary differential equations, one for the evolution of the concentration of clusters of each size, denoted by $c_i(t)$, for $t \geq 0$ and $i \geq 1$:

$$\begin{aligned} \frac{dc_1}{dt} &= -2J_1(c) - \sum_{i=1}^{+\infty} J_i(c)\,, \\ \frac{dc_i}{dt} &= J_{i-1}(c) - J_i(c)\,, \qquad i \geq 2\,, \end{aligned} \tag{1}$$

$$J_i(c) = a_i c_1 c_i - b_{i+1} c_{i+1}\,, \qquad i \geq 1\,.$$

We call the system (1) the *deterministic Becker-Döring equations* (DBD). This model was used to represent phase transition phenomena in physics, chemistry, and more recently gained in popularity in biology. The interested reader should refer to the surveys [9, 17] for details.

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In this note, we deal with the stochastic counterpart of this model whose construction follows. Let $(a_i)_{i\geq 1}$ and $(b_i)_{i\geq 2}$ be strictly positive sequences. For $n \geq 1$ and $\rho > 0$, we define the state space

$$\mathcal{E}_\rho^n = \left\{(c_i)_{i\geq 1} \in \mathbb{R}^{\mathbb{N}} \ : \ \forall i \geq 1\,,\, \tfrac{n}{\rho} c_i \in \mathbb{N},\ \sum_{i=1}^{+\infty} i c_i = \rho\right\},$$

where $\mathbb{R}^{\mathbb{N}}$ is the space of real sequences. An important fact is that $\mathcal{E}_\rho^n$ is a finite state space, which can be made clearer with the following equivalent representation: $c \in \mathcal{E}_\rho^n$ iff $c = (\frac{\rho}{n} x_1, \ldots, \frac{\rho}{n} x_n, 0, \ldots)$ with $x_i \in \{1, \ldots, \lfloor \frac{i}{n} \rfloor\}$ and $\sum_{i=1}^n i x_i = n$. Then, we define the following infinitesimal generator as the operator $\mathcal{A}^n$, on the set of borel function $\psi : \mathbb{R}^{\mathbb{N}} \to \mathbb{R}$, bounded on $\mathcal{E}_\rho^n$, by

$$\mathcal{A}^n(\psi)(c) = \frac{n}{\rho} \sum_{i=1}^{+\infty} \Big(A_i(c)[\psi(c + \tfrac{\rho}{n}\Delta_i) - \psi(c)] + B_{i+1}(c)[\psi(c - \tfrac{\rho}{n}\Delta_i) - \psi(c)] \Big) \tag{2}$$

where the transition rates are

$$A_1(c) = a_1 c_1 (c_1 - \tfrac{\rho}{n})\,, \qquad A_i(c) = a_i c_1 c_i\,, i \geq 2\,, \qquad B_i(c) = b_i c_i\,, i \geq 2\,,$$

and jump transitions $\Delta_i = e_{i+1} - e_i - e_1$ with $(e_1, e_2, \ldots)$ the canonical basis of $\mathbb{R}^{\mathbb{N}}$, that is $e_{ik} = 1$ if $k = i$ and 0 otherwise. Remark that for any $c \in \mathcal{E}_\rho^n$, if $A_i(c) > 0$ then $c + \frac{\rho}{n}\Delta_i \in \mathcal{E}_n^\rho$, while if $B_{i+1}(c) > 0$ then $c - \frac{\rho}{n}\Delta_i \in \mathcal{E}_\rho^n$. Finally, fix $(\Omega, \mathcal{F}, \mathbf{P})$ a (sufficiently large) probability space, denote by $\mathbf{E}$ the corresponding expectation and define the following process:

Definition 1.1. *For each $n \geq 1$, the* stochastic Becker-Döring process *(SBD) is a pure jump Markov process c^n on $(\Omega, \mathcal{F}, \mathbf{P})$, with value in $\mathcal{E}_\rho^n$, and infinitesimal generator $\mathcal{A}^n$ given in Eq.* (2)*.*

Since $\mathcal{E}_n^\rho$ is finite, given an initial law $c^{\mathrm{in},\,n} \in \mathcal{E}_\rho^n$, there exists a unique (in law) SBD process c^n with $c^n(0) = c^{\mathrm{in},\,n}$ (in law). Moreover, by construction of $\mathcal{E}_\rho^n$, this yields the so-called mass conservation,

$$\sum_{i=1}^{+\infty} i c_i^n(t) = \rho\,. \tag{3}$$

The parameter n can be seen as the total number of particles, and $\frac{n}{\rho}$ as a volume scaling. We study the behavior of the SBD model when n goes to infinity. As the state space grows together with n, this problem differs from standard results of chemical reaction networks (see discussion). In the first part of our note, we deal with the convergence of the time-dependent SBD process towards the solution of the DBD equations, as $n \to \infty$. In the second part, we elucidate the asymptotic behavior as $n \to \infty$ of the stationary distribution of the SBD process, which is related to the relative entropy associated to the DBD equations.

Summary. In Sec. 2 we introduce additional notations and our main results. In Sec. 3 we prove the Theorem 2.2 for the convergence in law of the SBD process. In Sec. 4, we prove the Theorem 2.3 for pathwise estimates. In Sec. 5 we prove the Theorem 2.4 on the convergence of the stationary distribution. We discuss our results with respect to the literature in section 6. Considered as appendix, in Sec. A we state a criterion for weak compactness of (density) measures and in Sec. B a criterion for tightness of jump processes. Both are used in the proof of Theorem 2.2.

2 Main Results

We naturally embed the sequence of state-space $\mathcal{E}_\rho^n$ into the space

$$X = \big\{(c_i)_{i\geq 1} \in \mathbb{R}^{\mathbb{N}} \ : \ \|c\| \coloneqq \sum_{i=1}^{+\infty} i c_i < +\infty\big\}.$$

The space $(X, \|\cdot\|)$ is complete and separable. For $0 < T \leq +\infty$, we denote by $D([0,T[,X)$ the space of right continuous with left limit X-valued function on $[0,T[$. Equipped with the Skorohod topology, the space $D([0,T[,X)$ is Polish (see [8, Theorem 5.6]). Denote by X^+ the non-negative cone of X. We assume the following on the sequence of initial condition.

Hypothesis. *The sequence of initial condition $\{c^{\mathrm{in,\,n}}\}$ belonging to $\mathcal{E}_\rho^n$ is deterministic, and there exists $c^{\mathrm{in}} \in X^+$ such that*

$$\lim_{n\to+\infty} \|c^{\mathrm{in,\,n}} - c^{\mathrm{in}}\| = 0. \tag{H1}$$

Remark, c^{in} given in (H1) has consequently the same mass ρ, that is $\|c^{\mathrm{in}}\| = \|c^{\mathrm{in,\,n}}\| = \rho$. In our first theorem for the convergence in law of the SBD process, we shall assume that the coagulation reaction rates are linearly bounded.

Hypothesis. *There exists a positive constant K such that*

$$a_i \leq K i, \quad i \geq 1. \tag{H2}$$

For our second theorem for pathwise convergence of the SBD process, we shall impose some monotonicity condition on both the coagulation and the fragmentation reaction rates.

Hypothesis. *There exists a positive constant K such that the reaction rate constants satisfy*

$$\begin{aligned} a_{i+1} - a_i &\leq K, \quad i \geq 1, \\ b_i - b_{i+1} &\leq K, \quad i \geq 2. \end{aligned} \tag{H3}$$

Hypothesis (H3) implies that $a_i \leq i \max(K, a_1)$, for all $i \geq 1$, and it thus stronger than hypothesis (H2). Before stating our limit theorems, we first need to recall the definition of a solution to the DBD equations as stated in [3].

Definition 2.1. *A (global) solution $c = \{c_i\}$ to the Deterministic Becker-Döring equations* (1) *is a function $c \coloneqq [0,+\infty) \to X$ such that*

1. *For all $t \geq 0$, $c(t) \in X^+$;*
2. *Each c_i is continuous and $\sup_{t\geq 0} \|c(t)\| < +\infty$;*
3. *For all $t \geq 0$, $\displaystyle\int_0^t \sum_{i=1}^{+\infty} [a_i c_1(s) c_i(s) + b_{i+1} c_{i+1}(s)] ds < +\infty$;*
4. *And satisfies, for all $t \geq 0$,*

$$c_i(t) = c_i(0) + \int_0^t [J_{i-1}(c(s)) - J_i(c(s))] ds, \qquad i \geq 2,$$
$$c_1(t) = c_1(0) - \int_0^t \left[J_2(c(s)) + \sum_{i=2}^{+\infty} J_i(c(s)) \right] ds.$$

Importantly, by [3, Corollary 2.6, Proposition 3.1], a solution c to the DBD equations is actually continuous from $[0,+\infty)$ to $(X, \|\cdot\|)$ and preserve the mass, that is, if $\|c(0)\| = \rho$, for all $t \geq 0$

$$\sum_{i=1}^{+\infty} i c_i(t) = \rho\,. \tag{4}$$

Existence of global solution to Eq. (1), in the sense of Definition 2.1, have been established in [3, Corollary 2.3] under the fairly general hypothesis (H2). This existence result is optimal in the sense that without any extra hypothesis on the strength of b_i (nor extra-regularity on the initial condition) the DBD equations may not have solution if hypothesis (H2) is not true (see [3, Theorem 2.7]). We can now state our first theorem.

Theorem 2.2. *Under Hypothesis* (H1) *and* (H2)*. If $\{c^n\}$ is the sequence of SBD processes with $c^n(0) = c^{\text{in},\,n}$ then, the sequence $\{c^n\}$ is relatively compact in $D([0,+\infty), X)$ and any limit point c is, almost surely, a solution to the DBD equations with initial condition $c(0) = c^{\text{in}}$.*

Uniqueness of solution to Eq. (1) requires extra hypotheses, either on the initial condition or on the constant rates (see for instance [3, Theorems 3.6 and 3.7]). When uniqueness holds, it is a direct consequence of Theorem 2.2 that the full sequence $\{c^n\}$ converges (in law) to the unique solution of the DBD system (1). To our knowledge, the best uniqueness result with the largest class of coagulation rates was obtained in [15, Theorem 2.1], under the hypothesis (H3). Under this hypothesis, we obtain our second theorem.

Theorem 2.3. *Under Hypothesis* (H1) *and* (H3)*. If $\{c^n\}$ is the sequence of SBD processes with $c^n(0) = c^{\text{in},\,n}$ and $\{c\}$ the unique solution of the DBD equations satisfying $c(0) = c^{\text{in}}$ then,*

$$\lim_{n\to+\infty} \mathbf{E} \sup_{t\in[0,T]} \|c^n(t) - c(t)\| = 0.$$

We turn now to the study of the behavior of the stationary distribution of the SBD process, as $n \to \infty$. The SBD process being a Markov chain in a finite state space, it is clear that it has an unique invariant measure on each irreducible component of the state space, that is on each $\mathcal{E}_\rho^n$. Moreover, the SBD process has the detailed balance property: it is reversible with respect to its invariant measure. To see that, let us define, for any $c \in \mathcal{E}_\rho^n$,

$$\Pi^n(c) = \frac{1}{B_n^z} \prod_{i=1}^{n} \frac{(\frac{n}{\rho} Q_i z^i)^{\frac{n}{\rho} c_i}}{(\frac{n}{\rho} c_i)!} e^{-\frac{n}{\rho} Q_i z^i}\,, \tag{5}$$

and $\Pi^n(c) = 0$ for all $c \notin \mathcal{E}_\rho^n$, where Q_i is defined by, for all $i \geq 1$,

$$Q_1 = 1, \qquad Q_i = \prod_{j=1}^{i-1} \frac{a_j}{b_{j+1}}, \quad i \geq 2\,,$$

and where $z > 0$ is arbitrary and B_n^z is the following normalizing constant

$$B_n^z = \sum_{c\in\mathcal{E}_\rho^n} \prod_{i=1}^{n} \frac{(\frac{n}{\rho} Q_i z^i)^{\frac{n}{\rho} c_i}}{(\frac{n}{\rho} c_i)!} e^{-\frac{n}{\rho} Q_i z^i}\,. \tag{6}$$

On can easily check that Π^n satisfies the reversibility condition: for all $c \in \mathcal{E}_\rho^n$, for all $i \geq 1$,

$$A_i(c)\Pi(c) = B_{i+1}(c + \tfrac{\rho}{n}\Delta_i)\Pi(c + \tfrac{\rho}{n}\Delta_i)\,,$$

and thus Π^n is the unique invariant distribution of the SBD process on $\mathcal{E}_\rho^n$. To understand the limiting behavior of the stationary distribution, it is convenient to write down the so-called non-equilibrium potential, for $c \in \mathcal{E}_\rho^n$,

$$-\frac{\rho}{n}\ln \Pi^n(c) = \sum_{i=1}^{n}\left\{-c_i \ln\left(\frac{n}{\rho}Q_i z^i\right) + \frac{\rho}{n}\ln\frac{n}{\rho}c_i! + Q_i z^i\right\} + \frac{\rho}{n}\ln B_n^z$$

Before stating our theorem on the limit of the stationary distribution, we first recall the main results on the long term behavior of the DBD equations. From Eq. (1), it is not difficult to see that any equilibrium of the DBD equations must be of the form c^z, defined by, for any $z > 0$,

$$c_i^z := Q_i z^i\,, \quad i \geq 1\,.$$

Thus, there is a family of potential candidates for the equilibrium of the DBD equations. Owing to the mass conservation Eq. (4), that holds for all finite times, it is natural to expect the equilibrium to satisfy the same relation namely,

$$\|c^z\| = \rho. \tag{7}$$

We naturally define the radius of convergence of the series $\|c^z\| = \sum_{i=1}^{+\infty} iQ_i z^i$,

$$z_s := \left(\limsup Q_i^{1/i}\right)^{-1}\,, \tag{8}$$

and the supremum value of $\|c^z\|$, which leads to the notion of critical mass

$$\rho_s := \sup_{z<z_s}\|c^z\|\,.$$

Indeed, by monotonicity of $\|c^z\|$ in z, either Eq. (7) has a unique solution z, or does not have any solution, depending on the mass ρ being less or greater than the critical threshold ρ_s, respectively. For any $\rho < \rho_s$, we define $z(\rho)$ as the unique solution of Eq. (7), that is

$$\|c^{z(\rho)}\| = \rho\,, \quad z < z_s\,. \tag{9}$$

It turns out that a dichotomy occurs in the large time behaviour of the DBD equations, which is at the corner stone of the phase transition phenomena of the Becker-Döring model. Under some additional technical assumptions (see [3, 16]), if $\rho \leq \rho_s$, any time-dependent solution of the DBD equations converges strongly towards $c^{z(\rho)}$, as $t \to \infty$. However, if $\rho > \rho_s$, any time-dependent solution of the DBD equations converges towards c^{z_s} in the $weak-*$ topology on X (pointwise convergence, component by component). In the latter case, the steady-state c^{z_s} has a mass strictly inferior than the initial condition. The quantity $\rho - \rho_s$ is interpreted as the mass which leaves the initial phase and undergoes a phase transition.

The long-time results of the DBD equations are proved thanks to the help of the following function $\mathcal{H}$, that turns to be a Lyapunov for DBD equations, for any $z > 0$,

$$\mathcal{H}(c|c^z) = \sum_{i=1}^{+\infty}\left\{c_i\left(\ln\frac{c_i}{Q_i z^i} - 1\right) + Q_i z^i\right\}\,. \tag{10}$$

The function $\mathcal{H}$ is also called the relative entropy, as, for $z \leq z_s$, and $c \in X^+$, with $\|c\| \leq \rho$, one have $\mathcal{H}(c|c^z) = 0$ if, and only if, $c = c^z$, and $\mathcal{H}(c|c^z) > 0$ otherwise. We refer to [6] for recent results and discussion of the use of entropy techniques for the DBD equations.

Our theorem on the limit of the stationary distribution (5), as $n \to \infty$, shows that a similar dichotomy holds for the stationary state of SBD process.

Theorem 2.4. *Assume $0 < z_s < +\infty$. Let $\{c^n\}$ a sequence belonging to $\mathcal{E}^n_\rho$.*

1. *If $0 < \rho \leq \rho_s$, and if $\liminf_{i\to+\infty} Q_i^{1/i} > 0$ and $c^n \to c \in X$ strongly, as $n \to \infty$, then*

$$\lim_{n\to+\infty} -\frac{\rho}{n} \ln \Pi^n(c^n) = \mathcal{H}(c|c^{z(\rho)}),$$

where $c^{z(\rho)} = \{Q_i z(\rho)^i\}$ such that $\|c^{z(\rho)}\| = \rho$.

2. *If $\rho > \rho_s$, and if $\lim Q_i^{1/i}$ exists, and $c^n \rightharpoonup c \in X$ $weak-*$, then*

$$\lim_{n\to+\infty} -\frac{\rho}{n} \ln \Pi^n(c^n) = \mathcal{H}(c|c^{z_s}) \,,$$

where $c^{z_s} = \{Q_i z_s^i\}$ and $\|c^{z_s}\| = \rho_s$.

3 Limit theorem: The general case

In this section we prove Theorem 2.2. We recall that the SBD process (see Definition 1.1) c^n is a continuous-time Markov chain with value in a finite state-space, whose infinitesimal generator is given by Eq. (2). We remind that, classically, for any Borel function $\psi : \mathbb{R}^\mathbb{N} \to \mathbb{R}$ bounded on $\mathcal{E}^n_\rho$, we have

$$\psi(c^n(t)) - \psi(c^{\mathrm{in,n}}) - \int_0^t \mathcal{A}^n \psi(c^n(s)) ds \tag{11}$$

is an L^2-martingale starting from 0 whose previsible quadratic variation is

$$\frac{n}{\rho} \int_0^t \sum_{i=1}^{+\infty} \Big(A_i(c^n(s))[\psi(c^n(s) + \tfrac{\rho}{n}\Delta_i) - \psi(c^n(s))]^2 + B_{i+1}(c^n(s))[\psi(c^n(s) - \tfrac{\rho}{n}\Delta_i) - \psi(c^n(s))]^2 \Big) ds. \tag{12}$$

The proof of Theorem 2.2 follows a general scheme in infinite dimensional settings, which we briefly sketch here:

(i) Moment estimates (Proposition 3.3): this is an important step as it provides a superlinear moment, crucial to control the infinite sums that arise in the limit $n \to \infty$.

(ii) Compactness (Proposition 3.4): we prove compactness in $D([0, +\infty), X)$ for the Skorohod topology, using essentially the mass conservation (3) and moment estimates from step (i).

(iii) Vanishing martingale (lemma 3.7): we classically control the sequence of martingales through their predictable quadratic variation process and Doob's inequality.

(iv) Continuity property (lemma 3.8): We use the classical fact (in the study of DBD equations) that the integral form of the *truncated* version of the DBD equations defines a continuous maps on $C([0, +\infty), X)$.

(v) Convergence of the truncation (lemma 3.9): the truncated version of the right-hand side of the DBD is arbitrary close to the one of the original DBD, uniformly with respect to the sequence of SBD processes. This estimate is possible thanks to step (ii).

(vi) The final step combines steps (i), (iii), (iv) and (v) to show that any limit point of the sequence of SBD processes satisfies the integral form of the DBD equations.

3.1 Regularities results

In this section we collect some important estimates. The next two lemma concern generalized moment propagation and the control of the fragmentation term, the latter playing the role of a diffusive term. Analogous results are known in the deterministic context, see [3, Theorem 2.2], and yield the important estimate in Lemma 3.2. Let $\{g_i\}$ be an arbitrary real sequence, and consider the measurable function defined by $\psi(\eta) := \sum_{i=1}^{+\infty} g_i \eta_i$ on $\mathcal{E}_\rho^n$. Note that ψ is necessarily bounded on $\mathcal{E}_\rho^n$. Hence, using the martingale (11), we have, for all $t \geq 0$, $n \geq 1$ and $N \geq 2$,

$$\mathbf{E}\left[\sum_{i=N}^{+\infty} g_i c_i^n(t)\right] + \mathbf{E}\left[\int_0^t \sum_{i=N}^{+\infty} (g_{i+1} - g_i) B_{i+1}(c^n(s)) ds\right] = \mathbf{E}\left[\sum_{i=N}^{+\infty} g_i c_i^n(0)\right] + \mathbf{E}\left[\int_0^t \left\{ g_N (A_{N-1}(c^n(s)) - B_N(c^n(s))) + \sum_{i=N}^{+\infty} (g_{i+1} - g_i) A_i(c^n(s)) \right\} ds\right] . \quad (13)$$

We deduce the following estimate.

Lemma 3.1. *Let $N \geq 2$ and $\{g_i\}$ be a non-negative non-decreasing sequence such that for some positive constant K_0 we have, for all $i \geq 1$, $a_i(g_{i+1} - g_i) \leq K_0 g_i$. Then, if*

$$\sup_{n\geq 1} \mathbf{E}\left[\sum_{i=N}^{+\infty} g_i c_i^n(0)\right] < +\infty \,,$$

there exists, for each $T > 0$, a constant K_T such that, for all $n \geq 1$,

$$\mathbf{E}\left[\sum_{i=N}^{+\infty} g_i c_i^n(t)\right] + \mathbf{E}\left[\int_0^T \left\{ g_N B_N(c^n(s)) + \sum_{i=N}^{+\infty} (g_{i+1} - g_i) B_{i+1}(c^n(s)) \right\} ds\right] \leq K_T . \quad (14)$$

The constant K_T also depends on N, $\{g_i\}$, K_0, the mass ρ and the kinetic rate a_{N-1}.

Proof. By Eq. (13) and the mass conservation Eq. (3), for all $t \geq 0$,

$$\mathbf{E}\left[\sum_{i=N}^{+\infty} g_i c_i^n(t)\right] \leq \mathbf{E}\left[\sum_{i=N}^{+\infty} g_i c_i^n(0)\right] + g_N a_{N-1} \rho^2 t + K_0 \rho \int_0^t \mathbf{E}\left[\sum_{i=N}^{+\infty} g_i c_i^n(s)\right] ds. \quad (15)$$

We first apply Grönwall lemma to bound uniformly in time on $[0, T]$ the left hand side of Eq. (15). Then, we use this bounds into (13) to conclude on the bound (14). Note that the (Fubini-Tonelli) inversion of expectation and time integral follows by the fact that any right-continuous left-limit process is progressive. □

Lemma 3.2. *Under Hypothesis* (H2)*, for each $T > 0$, there exists a constant K_T, such that,*

$$\sup_{n\geq 1} \mathbf{E}\left[\int_0^T \sum_{i=1}^{+\infty} \{A_i(c^n(s)) + B_{i+1}(c^n(s))\} ds\right] \leq K_T \quad (16)$$

Proof. Note that by Hypothesis (H2), with $g_i = i$ for all $i \geq 1$, we have $a_i(g_{i+1} - g_i) \leq K g_i$. Hence Eq. (16) readily follows from Lemma 3.1 with $N = 2$ and the mass conservation Eq. (3). □

The next result proves the existence of a finite super-linear moment, which allows us to control the formation of large clusters and to obtain compactness properties. Again, such bound is known for deterministic coagulation-fragmentation models, see for instance [15, 13]. We slightly improve the latter results, as no further assumption on the initial condition is needed here. Denote by $\mathcal{U}$ the set of non-negative convex functions ϕ, continuously differentiable with piecewise continuous second derivative, such that $\phi(x) = \frac{x^2}{2}$ for $x \in [0,1]$, ϕ' is concave, $\phi'(x) \leq x$ for $x \geq 0$, and

$$\lim_{x \to +\infty} \frac{\phi(x)}{x} = +\infty. \tag{17}$$

In appendix Sec. A we collect some properties related to the functions belonging to $\mathcal{U}$.

Proposition 3.3. *Under Hypothesis* (H1)*, there exists $\phi \in \mathcal{U}$ such that the sequence of initial condition $\{c^{\mathrm{in,\,n}}\}$ satisfies*

$$\sup_{n\geq 1} \sum_{i=1}^{+\infty} \phi(i) c_i^{\mathrm{in,\,n}} < +\infty\,. \tag{18}$$

Moreover, under Hypothesis (H2)*, if $\{c^n\}$ is the sequence of SBD processes with $c^n(0) = c^{\mathrm{in,\,n}}$, for all $T > 0$, there exists a constant K_T such that*

$$\sup_{n\geq 1} \mathbf{E}\left[\sup_{t\in[0,T]} \sum_{i=1}^{+\infty} \phi(i) c_i^n(t) \right] \leq K_T. \tag{19}$$

Proof. Let the punctual measure ν^n on $[0,+\infty)$ defined by, for any Borelian set A,

$$\nu^n(A) = \sum_{i=1}^{+\infty} c_i^{\mathrm{in,n}} \delta_i(A).$$

In particular, for all $n \geq 1$,

$$\int_0^{+\infty} x \nu^n(dx) = \rho\,.$$

By hypothesis (H1), the set $\{x \cdot \nu^n\}$ is relatively weakly compact in the space of Borel measures on $\mathbb{R}^+$ (recall that this topology is given by the sequential characterization of convergence of measure against bounded continuous functions). Then, by Theorem A.3 (with $g(x) = x$), there exists $\phi \in \mathcal{U}$ such that Eq. (18) holds. We denote by K_1 the constant arising in Eq. (18). Now, using Eqs. (11)-(12) with ψ given by $\psi(\eta) = \sum_{i=1}^{+\infty} \phi(i)\eta_i$ for $\eta \in \mathcal{E}_\rho^n$, we have (recall that for any $n \geq 1$ we deal with finite sums) for $t \geq 0$

$$\begin{aligned} &\sum_{i=1}^{+\infty} \phi(i) c_i^n(t) + \int_0^t \sum_{i=1}^{+\infty} B_{i+1}(c^n(s)) \left(\phi(i+1) - \phi(i) - \phi(1)\right) dt \\ &\quad = \sum_{i=1}^{+\infty} \phi(i) c_i^n(0) + \int_0^t \sum_{i=1}^{+\infty} A_i(c^n(s)) \left(\phi(i+1) - \phi(i) - \phi(1)\right) dt + M_\phi^n(t)\,, \end{aligned} \tag{20}$$

where M_ϕ^n is a square-integrable martingale starting from 0 and

$$\mathbf{E}|M_\phi^n(t)|^2 = \frac{\rho}{n} \mathbf{E}\left[\int_0^t \sum_{i=1}^{+\infty} [A_i(c^n(s)) + B_{i+1}(c^n(s))](\phi(i+1) - \phi(i) - \phi(1))^2 ds \right].$$

Since ϕ belongs to $\mathcal{U}$, by Prop. A.1, $\phi(i+1) - \phi(i) - \phi(1) \geq 0$ for all $i \geq 1$ and there is a positive constant K_2 such that, for all $i \leq n$,

$$\frac{\phi(i+1) - \phi(i) - \phi(1)}{n} \leq K_2.$$

Then, we obtain

$$\mathbf{E}|M_\phi^n(t)|^2 \leq \rho K_2 \mathbf{E}\left[\int_0^t \sum_{i=1}^{+\infty} [A_i(c^n(s)) + B_{i+1}(c^n(s))](\phi(i+1) - \phi(i) - \phi(1))ds\right]. \quad (21)$$

From Eq. (20), since ϕ is non-negative, we deduce

$$\mathbf{E}\int_0^t \sum_{i=1}^{+\infty} B_{i+1}(c^n(s))\left(\phi(i+1) - \phi(i) - \phi(1)\right)ds \leq K_1 + \mathbf{E}\int_0^t \sum_{i=1}^{+\infty} A_i(c^n(s))\left(\phi(i+1) - \phi(i) - \phi(1)\right)ds,$$

which in Eq. (21) yields

$$\mathbf{E}|M_\phi^n(t)|^2 \leq \rho K_1 K_2 + 2\rho K_2 \mathbf{E}\left[\int_0^t \sum_{i=1}^{+\infty} A_i(c^n(s))(\phi(i+1) - \phi(i) - \phi(1))ds\right]. \quad (22)$$

Moreover, by hypothesis (H2), the mass conservation in (3) and Prop. A.1, for all $t \in [0,T]$,

$$\begin{aligned}\mathbf{E}\left[\int_0^t \sum_{i=1}^{+\infty} A_i(c^n(s))\left(\phi(i+1) - \phi(i) - \phi(1)\right)ds\right] &\leq mK\rho \int_0^t \mathbf{E}\left[\sum_{i=1}^{+\infty} c_i^n(s)(i\phi(1) + \phi(i))\right] ds \\ &\leq mK\rho^2\phi(1)T + mK\rho\int_0^t \mathbf{E}\left[\sup_{\tau\in[0,s]} \sum_{i=1}^{+\infty} \phi(i)c_i^n(\tau)\right] ds, \quad (23)\end{aligned}$$

where m is a constant depending on ϕ. Using Eq. (23) into Eq. (22), and Doob's inequality, there is a some positive constant K_3 such that,

$$\mathbf{E}\sup_{s\in[0,t]} |M_\phi^n(s)| \leq K_3\left(1 + \int_0^t \mathbf{E}\left[\sup_{\tau\in[0,s]} \sum_{i=1}^{+\infty} \phi(i)c_i^n(\tau)\right] ds\right). \quad (24)$$

Now, we use again Eq. (20), but taking first supremum in time and then expectation, which entails

$$\mathbf{E}\left[\sup_{s\in[0,t]} \sum_{i=1}^{+\infty} \phi(i)c_i^n(s)\right] \leq K_1 + \mathbf{E}\left[\int_0^t \sum_{i=1}^{+\infty} A_i(c^n(s))\left(\phi(i+1) - \phi(i) - \phi(1)\right)\right] ds + \mathbf{E}\sup_{s\in[0,t]} |M_\phi^n(s)|. \quad (25)$$

We conclude using Eqs (23) and (24) into (25) and the Grönwall lemma. □

3.2 Compactness

In this section, we use a tightness criterion for the sequence of SBD processes $\{c^n\}$ in order to prove to the next proposition.

Proposition 3.4. *Under Hypotheses* (H1) *and* (H2)*, the sequence* $\{c^n\}$ *of SBD processes with* $c^n(0) = c^{\mathrm{in,\,n}}$ *is relatively compact in* $D([0,+\infty), X)$*. Any limit point* c *satisfies (almost surely) points 1 to 3 of the Definition 2.1 of a solution to the DBD equations, and is continuous from* $[0,+\infty)$ *to* X*.*

The compactness part of the proof is a direct application of a classical tightness criteria [8, Chap. 3 Corollary 7.4], which consists in verifying two points: first, the compact containment (lemma 3.5) and second, a control on the modulus of continuity (lemma 3.6).

Lemma 3.5. *For all $\varepsilon > 0$ and $t \geq 0$, there exists a compact subset $\Gamma_{\varepsilon,t}$ such that*

$$\mathbf{P}\left\{c^n(t) \notin \Gamma^c_{\varepsilon,t}\right\} \leq \varepsilon.$$

Proof. Let $\varepsilon > 0$ and $t \geq 0$. Fix $T \geq t$ and

$$\Gamma_{\varepsilon,T} = \left\{ c \in X^+ \,|\, \|c\| \leq \rho,\ \sum_{i=1}^{+\infty} \phi(i) c_i \leq \frac{K_T}{\varepsilon} \right\},$$

where ϕ is given by Proposition 3.3 and K_T is the constant in Eq. (19). Clearly, $\Gamma_{\varepsilon,T}$ is a compact subset of X. Since by the mass conservation Eq. (3), $\|c^n(t)\| = \rho$, we have

$$\mathbf{P}\left\{c^n(t) \notin \Gamma^c_{\varepsilon,T}\right\} = \mathbf{P}\left\{ \sum_{i=1}^{+\infty} \phi(i) c_i(t) > \frac{K_T}{\varepsilon} \right\}.$$

Then, by Chebychev's inequality and Proposition 3.3,

$$\mathbf{P}\left\{c^n(t) \notin \Gamma^c_{\varepsilon,T}\right\} \leq \varepsilon\,.$$

□

Let us define the modulus of continuity on X. For $\delta > 0$ and $T > 0$, the set Π_δ denotes the set of all partitions $\{t_k\}$ of $[0,T]$ such that for some K we have $0 = t_0 < t_1 < \cdots < t_{K-1} < T \leq t_K$ with $\min_{k=0,\ldots,K} |t_{k+1} - t_k| > \delta$, and we define

$$w(c,\delta,T) = \inf_{\{t_k\}\in\Pi_\delta} \max_k \sup_{s,t\in[t_k,t_{k+1}[} \|c_t - c_s\|,$$

for all $c \in D([0,+\infty), X)$.

Lemma 3.6. *For all $T \geq 0$ and $\varepsilon > 0$, there exists $\delta > 0$ such that*

$$\limsup_{n\to+\infty} \mathbf{P}\{w(c^n, T, \delta) \geq \varepsilon\} \leq \varepsilon.$$

Proof. Fix $T \geq 0$ and $\varepsilon > 0$. We define for each $N \geq 2$, $\delta > 0$ and $c \in D([0,T[, X)$,

$$w^N(c,\delta,T) = \inf_{\{t_k\}\in\Pi_\delta} \max_k \sup_{s,t\in[t_k,t_{k+1}[} \sum_{i=2}^{N} i|c_i(t) - c_i(s)|.$$

which is the modulus of continuity of the components 2 to N seen as an $\mathbb{R}^{N-1}$-valued process equipped with the 1-norm (with a factor i). Fix $N \geq 2$. The infinitesimal transition rate of $\{c_2^n, \ldots, c_N^n\}$ is given by

$$\lambda_N^n(t) \coloneqq \frac{n}{\rho} \sum_{i=2}^{N+1} \left[A_{i-1}(c^n(s)) + B_i(c^n(s))\right],$$

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

$$\lambda_N^n(t) \leq \frac{n}{\rho} \sup_{i\leq N+1} (a_{i-1} + b_i) \sum_{i=2}^{N+1} \left[c_1^n(s) c_{i-1}^n(s) + c_i^n(s)\right],$$

Thus, by the mass conservation Eq. (3), there exists a constant K_N depending on the $(N+1)$ first rate constants and ρ such that $\lambda_N^n(t) \leq K_N \frac{n}{\rho}$ for all $t \geq 0$ and $n \geq 1$. Moreover, any transition jump satisifies

$$\sum_{i=2}^{N} i|c_i^n(t) - c_i^n(t^-)| \leq 2N\frac{\rho}{n}\,.$$

Hence, by lemma B.1, there exists $\delta > 0$ such that

$$\limsup_{n\to+\infty} \mathbf{P}\{w^N(c^n,\delta,T) \geq \tfrac{\varepsilon}{2}\} \leq \tfrac{\varepsilon}{2}. \tag{26}$$

Now, for all $t, s \leq T$ and $n \geq 1$, we have

$$\sum_{i=N+1}^{+\infty} i|c_i^n(t) - c_i^n(s)| \leq 2 \sup_{j\geq N+1} \frac{j}{\phi(j)} \sup_{u\in[0,T]} \sum_{i=1}^{+\infty} \phi(i) c_i^n(u),$$

and

$$|c_1^n(t) - c_1^n(s)| = \left|\sum_{i=2}^{+\infty} i(c_i^n(t) - c_i^n(s))\right| \leq \sum_{i=2}^{N} i|c_i^n(t) - c_i^n(s)| + \sum_{i=N+1}^{+\infty} i|c_i^n(t) - c_i^n(s)|\,.$$

Thus,

$$\|c^n(t) - c^n(s)\| \leq 2\sum_{i=2}^{N} i|c_i^n(t) - c_i^n(s)| + 4 \sup_{j\geq N+1} \frac{j}{\phi(j)} \sup_{u\in[0,T]} \sum_{i=1}^{+\infty} \phi(i) c_i^n(u)\,,$$

so that

$$\mathbf{P}\{w(c^n,\delta,T) \geq \varepsilon\} \leq \mathbf{P}\{w^N(c^n,\delta,T) \geq \tfrac{\varepsilon}{2}\} + \mathbf{P}\left\{4 \sup_{j\geq N+1} \frac{j}{\phi(j)} \sup_{u\in[0,T]} \sum_{i=1}^{+\infty} \phi(i) c_i^n(u) \geq \tfrac{\varepsilon}{2}\right\} \tag{27}$$

Using Eq. (26), Lemma 3.3 and Chebychev's inequality into the above Eq. (27) we have for all $n \geq 1$,

$$\mathbf{P}\{w(c^n,\delta,T) \geq \varepsilon\} \leq \frac{\varepsilon}{2} + \frac{8}{\varepsilon} \sup_{j\geq N} \frac{j}{\phi(j)} K_T \tag{28}$$

where K_T is the constant in Eq. (19). By property of ϕ in Eq. (17), we can choose N large enough such that the second term in the right hand side of Eq. (28) is less than $\varepsilon/2$, to conclude the proof. □

Proof of Prop. 3.4. Using Lemma 3.5 and Lemma 3.6, we deduce from the tightness criteria [8, Chap. 3 Corollary 7.4] that $\{c^n\}$ is relatively compact in $D([0,+\infty), X)$. Let c be a limit point of $\{c^n\}$. Then a subsequence, still denoted by $\{c^n\}$, converges in distribution to c. We shall prove that points 1 to 3 of Definition 2.1 are valid a.s. for c. Since for all $i \geq 1$, the map $\eta \mapsto \psi(\eta) = \eta_i$ is continuous from $D([0,+\infty), X)$ into $D([0,+\infty), \mathbb{R})$, each sequence $\{c_i^n\}$ converges in law to $\{c_i\}$. Moreover, for all $i \geq 1$, and $T > 0$,

$$\sup_{t\leq T} |c_i^n(t) - c_i^n(t^-)| \leq \frac{\rho}{n} \to 0\,, \quad n \to \infty\,,$$

which by [8, Chap. 3 Theorem 10.2] ensures all c_i are almost surely continuous. Remark that $F_\rho \coloneqq \{\eta \in D([0,+\infty), X) \,|\, \eta_i(t) \geq 0,\ \|\eta(t)\| \leq \rho,\ \text{for all } t \geq 0\}$ is a close subset of

$D([0,+\infty),X)$. Thus, by Portemanteau theorem, $1 = \limsup \mathbf{P}\{c^n \in F\} \leq \mathbf{P}\{c \in F\}$. We prove that point 3 of Definition 2.1 holds using that, for all finite N,

$$\begin{aligned}\mathbf{E}&\left[\int_0^t \sum_{i=1}^{N} [a_i c_1(s) c_i(s) + b_{i+1} c_{i+1}(s)] ds\right] \\ &= \lim_{n\to\infty} \mathbf{E}\left[\int_0^t \sum_{i=1}^{N} [a_i c_1^n(s) c_i^n(s) + b_{i+1} c_{i+1}^n(s)] ds\right] \\ &\leq \limsup_{n\to\infty} \mathbf{E}\left[\int_0^t \sum_{i=1}^{\infty} [a_i c_1^n(s) c_i^n(s) + b_{i+1} c_{i+1}^n(s)] ds\right] < \infty\,, \qquad (29)\end{aligned}$$

where the first line of Eq. (29) is a consequence of the convergence in law of $\{c^n\}$ towards c, and the second line of Eq. (29) comes from Lemma 3.2. We conclude by the monotone convergence theorem, that for all $t \geq 0$,

$$\mathbf{E}\left[\int_0^t \sum_{i=1}^{+\infty} [a_i c_1(s) c_i(s) + b_{i+1} c_{i+1}(s)] ds\right] < +\infty.$$

We end the proof by (countable, $i \geq 1$ and $t \in \mathbb{N}$ for instance) construction of a set of probability 0 for which properties 1 to 3 of Definition 2.1 hold. Finally, the same strategy as Eq. (29) shows that c also satisfies the inequality (using Proposition 3.3)

$$\sup_{t\in[0,T]} \sum_{i=1}^{+\infty} \phi(i) c_i(t) < \infty\,, \quad a.s. \qquad (30)$$

Thus, the continuity of each c_i and Eq. (30) yields that c is actually continuous from $[0,+\infty)$ to X. □

3.3 Identification of the limit

Thanks to Proposition 3.4 it remains to prove that any limit point c satisfies, almost surely, point 4 of the Definition 2.1. To prepare the proof, let us introduce few notations. We define, for $\eta \in X$,

$$\mathcal{A}_1(\eta) := -2J_1(\eta) - \sum_{i=2}^{+\infty} J_i(\eta), \text{ and } \mathcal{A}_i(\eta) := J_{i-1}(\eta) - J_i(\eta), \quad i \geq 2\,,$$

where we recall that J_i are defined in Eq. (1). We also define, for $t \geq 0$ and $\eta \in D([0,+\infty),X)$,

$$M_i(\eta,t) := \eta_i(t) - \eta_i(0) - \int_0^t \mathcal{A}_i(\eta(s)) ds\,.$$

Thus, point 4 of Definition 2.1 is equivalent to $M_i(c,t) = 0$, for all $i \geq 1$ and $t \geq 0$. Let ψ_i the continuous function on X defined by $\psi_i(\eta) = \eta_i$. We define, for each $i \geq 1$,

$$\mathcal{A}_i^n(\eta) := \mathcal{A}^n \psi_i(\eta) = \frac{\rho}{n} a_1 \eta_1 (2e_{i1} - e_{i2}) + \mathcal{A}_i(\eta)\,,$$

where $e_{ik} = 1$ if $i = k$ and 0 otherwise. We then finally define

$$M_i^n(t) := c_i^n(t) - c_i^n(0) - \int_0^t \mathcal{A}_i^n(c^n(s)) ds.$$

Using the martingale representation Eq. (11)-(12), we deduce that M_i^n is an L^2 martingale starting from 0 and satisfies, for all $i \geq 2$,

$$\mathbf{E}\left[|M_i^n(T)|^2\right] = \mathbf{E}\left[\frac{\rho}{n}\int_0^T \left(A_{i-1}(c^n(s)) + B_i(c^n(s)) + A_i(c^n(s)) + B_{i+1}(c^n(s))\right) ds\right], \quad (31)$$

and for $i = 1$,

$$\mathbf{E}\left[|M_1^n(T)|^2\right] = \mathbf{E}\left[\frac{\rho}{n}\int_0^T \sum_{i=1}^{+\infty}(1+e_{1i})\left(A_i(c^n(s)) + B_{i+1}(c^n(s))\right) ds\right]. \quad (32)$$

With these notations we have that for all $i \geq 1$ and $t \geq 0$,

$$M_i(c^n, t) = M_i^n(t) + \frac{\rho}{n}\int_0^t a_1 c_1^n(s)(2e_{i1} - e_{i2})ds\,, \quad (33)$$

and we are ready to prove the

Lemma 3.7. *For all* $i \geq 1$ *and* $T > 0$*,*

$$\lim_{n\to+\infty} \mathbf{E} \sup_{t\in[0,T]} |M_i(c^n, t)| = 0.$$

Proof. Using Eqs (31) and (32), and Lemma 3.2, we have, for $T > 0$ and all $i \geq 1$,

$$\mathbf{E}\left[|M_i^n(T)|^2\right] \leq 2\frac{\rho}{n}K_T\,,$$

where K_T is the constant in Eq. (16). By Doob's inequality,

$$\mathbf{E}\left[\sup_{t\in[0,T]} |M_i^n(t)|\right] \leq 2\sqrt{2\frac{\rho}{n}K_T}. \quad (34)$$

Using the mass conservation Eq. (3) and Eq. (34) into Eq. (33), we end the proof. □

The next step is to show that the applications M_i are continuous. The case $i = 1$ yields an infinite sum and must be treated separately. Classically in the study of the DBD equations, this infinite sum is truncated and an extra-moment helps to conclude. We shall proceed similarly. Let us define, for $N \geq 3$, and $\eta \in D([0, +\infty), X)$,

$$\tilde{M}_1^N(\eta, t) := \eta_1(t) - \eta_1(0) - \int_0^t \left[2J_1(\eta(s) + \sum_{i=2}^{N-1} J_i(\eta(s))\right] ds.$$

Lemma 3.8. *For all* $t \geq 0$ *and* $N \geq 3$*, the maps defined on* $D([0, +\infty), X)$ *by*

$$\eta \mapsto \tilde{M}_1^N(\eta, t) \text{ and } \eta \mapsto M_i(\eta, t)$$

for $i \geq 2$ *are continuous at any* $c \in C([0, +\infty), X)$*.*

Proof. Let $\eta \in C([0, +\infty), X)$ and $\{\eta^n\}$ a sequence belonging to $D([0, +\infty), X)$ converging to η for the Skorohod topology. Hence, each sequence $\{\eta_i^n\}$ converges to η_i in $D([0, +\infty), \mathbb{R})$. Then, for all $i \geq 1$, we have $\eta_i^n(t) \to \eta_i(t)$ as $n \to +\infty$, for all $t \geq 0$, by [8, Chap. 3 Prop. 5.2]. Since η_i^n is bounded, by the dominated convergence theorem, the sequences of time integrals $\int_0^t \eta_i^n(s)ds$ and $\int_0^t \eta_1^n\eta_i^n(s)ds$ are converging to, respectively, $\int_0^t \eta_i(s)ds$ and $\int_0^t \eta_1\eta_i(s)ds$ These conclude the proof as $\tilde{M}_1^N$ and M_i are finite sums. □

We now show that the truncation $\tilde{M}_1^N$ converges to M_1, as $N \to \infty$, along the sequence of SBD processes and any of its limit points.

Lemma 3.9. *We have the following two limits:*

$$\lim_{N\to+\infty}\sup_{n\geq 1}\mathbf{E}\sup_{t\in[0,T]}|M_1(c^n,t)-\tilde{M}_1^N(c^n,t)|=0,$$

and if c is a limit point of $\{c^n\}$,

$$\lim_{N\to+\infty}\mathbf{E}\sup_{t\in[0,T]}|M_1(c,t)-\tilde{M}_1^N(c,t)|=0.$$

Proof. Let $T>0$, $N\geq 3$. For all $t\in[0,T]$,

$$|M_1(c^n,t)-\tilde{M}_1^N(c^n,t)|\leq\int_0^T\sum_{i=N}^{+\infty}[A_i(c^n(s))+B_{i+1}(c^n(s))]ds. \tag{35}$$

We shall first deal with the fragmentation term. From Eq. (13), taking $g_i=i$ and using Hypothesis (H2) and the mass conservation (3), we get

$$\mathbf{E}\int_0^T\sum_{i=N}^{+\infty}B_{i+1}(c^n(s))ds\leq\mathbf{E}\sum_{i=N}^{+\infty}ic_i^n(0)$$
$$+\mathbf{E}\int_0^t\left[N(A_{N-1}(c^n(s))-B_N(c^n(s)))+K\rho\sum_{i=N}^{+\infty}ic_i^n(s)\right]ds. \tag{36}$$

Note that from Eq. (11), we deduce that

$$\sum_{i=N}^{+\infty}c_i^n(t)-\sum_{i=N}^{+\infty}c_i^n(0)-\int_0^t(A_{N-1}(c^n(s))-B_N(c^n(s)))$$

is a martingale starting from 0. Hence we obtain, from Eq. (36),

$$\mathbf{E}\int_0^T\sum_{i=N}^{+\infty}B_{i+1}(c^n(s))ds\leq\mathbf{E}\sum_{i=N}^{+\infty}ic_i^n(0)+\mathbf{E}\sum_{i=N}^{+\infty}ic_i^n(t)+K\rho\mathbf{E}\int_0^t\sum_{i=N}^{+\infty}ic_i^n(s)ds. \tag{37}$$

Using the extra-moment estimate in (19), we deduce from Eq. (37),

$$\mathbf{E}\int_0^T\sum_{i=N}^{+\infty}B_{i+1}(c^n(s))ds\leq K_T\sup_{i\geq N}\frac{i}{\phi(i)} \tag{38}$$

for some new constant K_T. Using Hypothesis (H2) and the extra-moment estimate Eq. (19), the coagulation term is directly controlled by

$$\mathbf{E}\int_0^T\sum_{i=N}^{+\infty}A_i(c^n(s))ds\leq K_T\sup_{i\geq N}\frac{i}{\phi(i)} \tag{39}$$

for some new constant K_T. The first part of the lemma is then proved using Eqs. (38) and (39) into Eq. (35), letting $N\to\infty$ and using the property of ϕ.

The proof of the second part of the lemma goes along similar lines. Let c a limit point of $\{c^n\}$. We have

$$|M_1(c,t)-\tilde{M}_1^N(c,t)|\leq\int_0^T\sum_{i=N}^{+\infty}[A_i(c(s))+B_{i+1}(c(s))]ds.$$

The coagulation term is controlled as previously, due to the control in Eq. (30). The fragmentation term requires an extra step as follows. From Eq. (38), it is clear that, for any $R > N$,

$$\mathbf{E}\int_0^T \sum_{i=N}^{R} B_{i+1}(c^n(s))ds \leq K_T \sup_{i\geq N} \frac{i}{\phi(i)}\,.$$

Thus, by convergence in law of c^n towards c, we obtain, as $n \to \infty$,

$$\mathbf{E}\int_0^T \sum_{i=N}^{R} B_{i+1}(c(s))ds \leq K_T \sup_{i\geq N} \frac{i}{\phi(i)}\,.$$

Then, by monotone convergence theorem, as $R \to \infty$,

$$\mathbf{E}\int_0^T \sum_{i=N}^{\infty} B_{i+1}(c(s))ds \leq K_T \sup_{i\geq N} \frac{i}{\phi(i)}\,,$$

and we obtain the second limit of our lemma using Eq. (30). ☐

Proof of the Theorem 2.2. Let c be a limit point of $\{c^n\}$ in $D([0,+\infty),X)$. By Proposition 3.4, c is almost surely continuous in time. Thus, by Lemma 3.8, for all $t \geq 0$ and for each $i \geq 2$, $M_i(c^n,t)$ converges in distribution to $M_i(c,t)$. Then, by Fatou's lemma, for all $t \geq 0$, and $i \geq 2$,

$$\mathbf{E}|M_i(c,t)| \leq \liminf_{n\to+\infty} \mathbf{E}|M_i(c^n,t)| = 0\,,$$

where the last equality is due to lemma 3.7. Then, as $t \mapsto M_i(c,t)$ is continuous in time, we have, almost surely, for all $t \geq 0$ and $i \geq 2$,

$$M_i(c,t) = 0. \tag{40}$$

We turn now to the case $i = 1$. For all $t \geq 0$, we have

$$\begin{aligned}\mathbf{E}|M_1(c,t)| \leq \mathbf{E}|M_1(c,t) - \tilde{M}_1^N(c,t)| + \mathbf{E}|\tilde{M}_1^N(c,t) - \tilde{M}_1^N(c^n,t)| \\ + \mathbf{E}|\tilde{M}_1^N(c^n,t) - M_1(c^n,t)| + \mathbf{E}|M_1(c^n,t)|.\end{aligned} \tag{41}$$

The last term of the right hand side of Eq. (41) goes to zero as $n \to +\infty$ by Lemma 3.7. Then, we observe that (taking only the expectation in c)

$$\eta \to \mathbf{E}|\tilde{M}_1^N(c,t) - \tilde{M}_1^N(\eta,t)|$$

defined on $D([0,+\infty),X)$ is continuous at any $\eta \in C([0,+\infty),X)$. Then,

$$\lim_{n\to+\infty} \mathbf{E}|\tilde{M}_1^N(c,t) - \tilde{M}_1^N(c^n,t)| = 0.$$

Finally, we first take the limit in $n \to +\infty$ in Eq. (41), and then in N, to obtain, by Lemma 3.9, for all $t \geq 0$,

$$\mathbf{E}|M_1(c,t)| = 0\,.$$

From point 3. of the definition 2.1, $t \mapsto M_1(c,t)$ is continuous in time and we have, almost surely, for all $t \geq 0$, $M_1(c,t) = 0$. Together with Eq. (40), we conclude that c is almost surely a solution of the DBD equations (1), in the sense of definition 2.1. ☐

4 Limit theorem: Pathwise convergence

Let c^n be the SBD process defined in Definition 1.1, and let c be the unique solution (under Hypothesis (H2)) to the DBD equations with initial condition c^{in}. To prove our Theorem 2.3, we follow the proof in [15] of uniqueness of solutions to the DBD equations. For that we introduce some notations. We define

$$E_i^n(t) = \sum_{j=i}^{+\infty} [c_j^n(t) - c_j(t)]\,,$$

for all $t \geq 0$, $i \geq 1$ and $n \geq 1$. Remark that by Eq. (3)-(4), we have $|E_i^n(t)| \leq 2\rho$. Then, from the DBD Eq. (1) and the martingale given in Eq. (11), we deduce that, for all $i \geq 2$,

$$E_i^n(t) - E_i^n(0) - \int_0^t (J_{i-1}(c^n(s)) - J_{i-1}(c(s)))\,ds + e_{i2}\frac{\rho}{n}\int_0^t a_1 c_1^n(s) ds$$

is a martingale. We aim to prove in this section that, for all $i \geq 1$,

$$\lim_{n\to+\infty} \mathbf{E} \sup_{t\in[0,T]} |E_i^n(t)| = 0 \tag{42}$$

Writing an equation on $|E_i^n(t)|$ yields several problems around 0 from the lack of smoothness. Hence we shall work with smooth functions φ, sufficiently close to $|\cdot|$. Let φ a continuously differentiable function on $[-2\rho, 2\rho]$. Applying Ito's formula, we obtain, for any $N \geq 2$,

$$\begin{aligned}\sum_{i=2}^N \varphi(E_i^n(t)) = \sum_{i=2}^N \varphi(E_i^n(0)) - \int_0^t \sum_{i=2}^N \varphi'(E_i^n(s)) J_{i-1}(c(s))) ds \\ + \frac{n}{\rho}\int_0^t \sum_{i=2}^N \Big\{ A_{i-1}(c^n(s))\left[\varphi(E_i^n(s) + \tfrac{\rho}{n}) - \varphi(E_i^n(s))\right] \\ + B_i(c^n(s))\left[\varphi(E_i^n(s) - \tfrac{\rho}{n}) - \varphi(E_i^n(s))\right]\Big\} ds + O_\varphi^N(t)\,,\end{aligned} \tag{43}$$

where O_φ^N is an L^2-martingale with

$$\begin{aligned}\mathbf{E}|O_\varphi^N(t)|^2 = \frac{n}{\rho}\mathbf{E}\int_0^t \sum_{i=2}^N \Big\{ A_{i-1}(c^n(s))\left[\varphi(E_i^n(s) + \tfrac{\rho}{n}) - \varphi(E_i^n(s))\right]^2 \\ + B_i(c^n(s))\left[\varphi(E_i^n(s) - \tfrac{\rho}{n}) - \varphi(E_i^n(s))\right]^2\Big\} ds\,.\end{aligned} \tag{44}$$

We collect a first estimate in the next lemma for a certain class of functions.

Lemma 4.1. *Assume hypothesis* (H3)*, and let φ a non-negative convex function, continuously differentiable on $[-2\rho, 2\rho]$, having finite right and left second derivatives, such that there exists $\varepsilon > 0$ for which $|x| \leq \varphi(x) \leq |x| + \varepsilon$ for all $x \in \mathbb{R}$. For any $N \geq 2$ and $T > 0$, there exists a constants K' independent on φ, N, n and ε such that,*

$$\begin{aligned}\mathbf{E}\sup_{t\in[0,T]} \sum_{i=2}^N \varphi(E_i^n(t)) \leq \exp(K'(\|\varphi'\|_\infty + 1)T)\Big\{\mathbf{E}\sum_{i=2}^N |E_i^n(0)| \\ b_N \mathbf{E}\int_0^T |E_{N+1}^n(t)| dt + K'(\|\varphi'\|_\infty + 1)\mathbf{E}\int_0^T \sum_{i=N+1}^{+\infty} |E_i^n(t)| dt \\ + K'(1+b_N)T\varepsilon + K'\|\varphi'\|_\infty T\frac{1}{n} + \frac{\rho}{n}\|\varphi''\|_\infty K' + 2\sqrt{\frac{\rho}{n}K'\|\varphi''\|_\infty}\Big\}.\end{aligned} \tag{45}$$

where $\|\cdot\|_\infty$ *is the norm of the supremum on* $[-2\rho, 2\rho]$.

Proof. Let $N \geq 2$ and $T > 0$. Since φ is C^1 with finite right and left second derivatives, by Taylor's expansion, we deduce from Eq. (43) and the mass conservation Eq. (3), that, for all $t \leq T$,

$$\sum_{i=2}^{N} \varphi(E_i^n(t)) \leq \sum_{i=2}^{N} \varphi(E_i^n(0)) + \int_0^t \sum_{i=2}^{N} \varphi'(E_i^n(s))[J_{i-1}(c^n(s)) - J_{i-1}(c(s))]ds$$
$$+ \frac{\rho^2}{n} a_1 \|\varphi'\|_\infty T + \frac{\rho}{n}\|\varphi''\|_\infty \int_0^t \sum_{i=2}^{N} \{A_{i-1}(c^n(s)) + B_i(c^n(s))\}\, ds + O_\varphi^N(t) \quad (46)$$

We now write

$$J_{i-1}(c^n) - J_{i-1}(c) = a_{i-1}c_{i-1}\left(c_1^n - c_1\right) + a_{i-1}c_1^n\left(E_{i-1}^n - E_i^n\right) - b_i\left(E_i^n - E_{i+1}^n\right) .$$

Then, by convexity of φ, we have for all $i \geq 2$, $s \geq 0$,

$$\varphi'(E_i^n(s))[J_{i-1}(c^n(s)) - J_{i-1}(c(s))] \leq \|\varphi'\|_\infty a_{i-1}c_{i-1}(s)|c_1^n(s) - c_1(s)|$$
$$+ a_{i-1}c_1^n\left(\varphi(E_{i-1}^n(s)) - \varphi(E_i^n(s))\right) + b_i\left(\varphi(E_{i+1}^n(s)) - \varphi(E_i^n(s))\right). \quad (47)$$

Summing Eq. (47) from $i = 2$ to N and reordering sums yields to

$$\sum_{i=2}^{N} \varphi'(E_i^n(s))[J_{i-1}^n(c^n(s)) - J_{i-1}(c(s))] \leq \|\varphi'\|_\infty |c_1^n(s) - c_1(s)| \sum_{i=1}^{N-1} a_i c_i(s)$$
$$+ a_1 c_1^n \varphi(E_1^n(s)) - a_{N-1}c_1^n\varphi(E_N^n(s)) + \sum_{i=2}^{N-1}(a_i - a_{i-1})c_1^n(s)\varphi(E_i^n(s))$$
$$+ b_N\varphi(E_{N+1}^n(s)) - b_2\varphi(E_2^n(s)) + \sum_{i=2}^{N-1}(b_i - b_{i+1})\varphi(E_{i+1}^n(s)). \quad (48)$$

Using Hypothesis (H3), the mass conservation (3) and dropping non-positives terms into Eq. (48) entails

$$\sum_{i=2}^{N} \varphi'(E_i^n(s))[J_{i-1}^n(c^n(s)) - J_{i-1}(c(s))] \leq \max(K, a_1)\rho\|\varphi'\|_\infty |c_1^n(s) - c_1(s)|$$
$$+ a_1\rho\varphi(E_1^n(s)) + b_N\varphi(E_{N+1}^n(s)) + K(\rho+1)\sum_{i=2}^{N}\varphi(E_i^n(s)). \quad (49)$$

Note that using the mass conservation (3)-(4), we deduce that $c_1^n - c_1 = -E_2 - \sum_{i=2}^{+\infty} E_i^n$, so that

$$|c_1^n(s) - c_1(s)| \leq |E_2^n(s)| + \sum_{i=2}^{+\infty}|E_i^n(s)| \leq 2\sum_{i=2}^{N}\varphi(E_i^n(s)) + \sum_{i=N+1}^{+\infty}|E_i^n(s)|\,, \quad (50)$$

as $|x| \leq \varphi(x)$. Since $E_1^n = c_1^n - c_1 + E_2^n$ and $\varphi(x) \leq |x| + \varepsilon$, we have, by Eq. (50),

$$\varphi(E_1^n(s)) \leq \varepsilon + |E_1^n(s)| \leq \varepsilon + 3\sum_{i=2}^{N}\varphi(E_i^n(s)) + \sum_{i=N+1}^{+\infty}|E_i^n(s)|. \quad (51)$$

Combining Eqs. (51) and (50) into (49), we deduce that there exists a constant K' independent on φ, N, n and ε, such that, for all $s \geq 0$,

$$\sum_{i=2}^{N} \varphi'(E_i^n(s))[J_{i-1}^n(c^n(s)) - J_{i-1}(c(s))] \leq b_N \varphi(E_{N+1}^n(s)) + a_1\rho\varepsilon + K'(\|\varphi'\|_\infty + 1)\left(\sum_{i=2}^{N} \varphi(E_i^n(s)) + \sum_{i=N+1}^{+\infty} |E_i^n(s)|\right). \quad (52)$$

Taking supremum in time, then expectation, we deduce from (52) and (46) that there exists a constant (again denoted by K') independent on φ, N, n and ε, such that, for all $t \in [0, T]$

$$\mathbf{E} \sup_{s\in[0,t]} \sum_{i=2}^{N} \varphi(E_i^n(s)) \leq \mathbf{E} \sum_{i=2}^{N} \varphi(E_i^n(0)) + K'(\|\varphi'\|_\infty + 1) \int_0^t \mathbf{E} \sup_{s\in[0,\tau]} \sum_{i=2}^{N} |E_i^n(\tau)| d\tau + b_N \mathbf{E} \int_0^t |E_{N+1}^n(s)| ds + K'(\|\varphi'\|_\infty + 1)\mathbf{E} \int_0^t \sum_{i=N+1}^{+\infty} |E_i^n(s)| ds + \mathbf{E} \sup_{s\in[0,t]} |O_\varphi^N(s)| + K'(1 + b_N)T\varepsilon + K'\|\varphi'\|_\infty T \frac{1}{n} + \frac{\rho}{n}\|\varphi''\|_\infty \int_0^t \sum_{i=2}^{N} \{A_{i-1}(c^n(s)) + B_i(c^n(s))\} ds. \quad (53)$$

We observe that by Doob's inequality and Eq. (44), we have

$$\mathbf{E}\left[\sup_{t\in[0,T]} |O_\varphi^N(t)|\right] \leq 2\sqrt{\frac{\rho}{n}\|\varphi'\|_\infty^2 \mathbf{E} \int_0^T \sum_{i=2}^{N} \{A_{i-1}(c^n(s)) + B_i(c^n(s))\} ds} \quad (54)$$

Using Lemma 3.2, we deduce from Eq. (53)-(54) that

$$\mathbf{E} \sup_{s\in[0,t]} \sum_{i=2}^{N} \varphi(E_i^n(s)) \leq \mathbf{E} \sum_{i=2}^{N} \varphi(E_i^n(0)) + K'(\|\varphi'\|_\infty + 1) \int_0^t \mathbf{E} \sup_{s\in[0,\tau]} \sum_{i=2}^{N} |E_i^n(\tau)| d\tau + b_N \mathbf{E} \int_0^t |E_{N+1}^n(s)| ds + K'(\|\varphi'\|_\infty + 1)\mathbf{E} \int_0^t \sum_{i=N+1}^{+\infty} |E_i^n(s)| ds + K'(1 + b_N)T\varepsilon + K'\|\varphi'\|_\infty T \frac{1}{n} + \frac{\rho}{n}\|\varphi''\|_\infty K_T + 2\sqrt{\frac{\rho}{n} K_T \|\varphi''\|_\infty},$$

where K_T is the constant in (16). We conclude that Eq (45) holds by the Grönwall lemma. □

To be able to pass in the limit n goes to $+\infty$ and then ε to 0 into Eq. (45), we need the next lemma:

Lemma 4.2. *Under Hypothesis* (H3)*, we have the following limits*

$$\lim_{N\to+\infty} \sup_{n\geq1} \mathbf{E} \sup_{t\in[0,T]} \sum_{i=N+1}^{+\infty} |E_i^n(t)| = 0 \quad (55)$$

$$\lim_{N\to+\infty} \sup_{n\geq1} b_N \mathbf{E} \int_0^T |E_{N+1}^n(t)| dt = 0 \quad (56)$$

Proof. We first observe that

$$\sum_{i=N}^{+\infty} |E_i^n(t)| \leq \sum_{j=N}^{+\infty} \sum_{i=N}^{j} (c_j^n(t) + c_j(t)) \leq 2 \sum_{j=N}^{+\infty} j c_j^n(t) + 2 \sum_{j=N}^{+\infty} j c_j(t)\,. \tag{57}$$

From Prop. 3.3, there exists $\phi \in \mathcal{U}$ and a constant K_T such that

$$\mathbf{E}\left[\sup_{t\in[0,T]} \sum_{j=N}^{+\infty} j c_j^n(t)\right] \leq \sup_{i\geq N} \frac{i}{\phi(i)} K_T, \tag{58}$$

Similarly, by [13, Theorem 2.5 and 4.1], there exists $\tilde{\phi} \in \mathcal{U}$ such that

$$\sup_{t\in[0,T]} \sum_{i=1}^{+\infty} \tilde{\phi}(i) c_i(t) < K_T,$$

for some new constant K_T. Thus, we also have

$$\sup_{t\in[0,T]} \sum_{i=1}^{+\infty} i c_i(t) \leq \sup_{i\geq N} \frac{i}{\tilde{\phi}(i)} K_T. \tag{59}$$

Using Eqs. (58)-(59) into Eq. (57), together with the properties of ϕ and $\tilde{\phi}$ in $\mathcal{U}$, we deduce that Eq. (55) holds.

We now prove the second limit of the lemma. By Hypothesis (H3), we obtain

$$\mathbf{E}\int_0^T b_N \sum_{i=N+1}^{+\infty} c_i^n(t)dt \leq \mathbf{E}\int_0^T \sum_{i=N+1}^{+\infty} b_i c_i^n(t)dt + K\mathbf{E}\int_0^T \sum_{i=N+1}^{+\infty} i c_i^n(t)dt.$$

Hence, by (58) and the estimate obtained in Eq. (38), the right hand side goes to 0 uniformly in n, as N to $+\infty$. Moreover, from point 2 and 3 of Definition 2.1,

$$\lim_{N\to\infty} \int_0^T \sum_{i=N+1}^{+\infty} b_i c_i(t)dt + K\mathbf{E}\int_0^T \sum_{i=N+1}^{+\infty} i c_i(t)dt = 0\,.$$

which allows us to conclude that Eq. (56) holds. □

Proof of the Theorem 2.3. We are now ready to prove our theorem. We first construct a sequence of function $\{\varphi_\varepsilon\}$ satisfying hypothesis of Lemma 4.1 together with uniformly bounded first derivative. For instance, we can define $\varphi_\varepsilon(x) = \frac{1}{2\varepsilon}x^2 + \frac{\varepsilon}{2}$ for $|x| \leq \varepsilon$ and $\varphi_\varepsilon(x) = |x|$ for $|x| \geq \varepsilon$. Thus $|\varphi_\varepsilon'\|_\infty \leq 1$. By Lemma 4.1 with φ_ε in Eq. (45), using Hypothesis (H1), we have

$$\begin{aligned}\lim_{\varepsilon\to 0}\lim_{n\to 0} \mathbf{E} \sup_{t\in[0,T]} \sum_{i=2}^{N} \varphi_\varepsilon(E_i^n(s)) \leq \exp(2K'T)\Bigg\{ &\sup_{n\geq 0} b_N \mathbf{E}\int_0^T |E_{N+1}^n(s)|ds \\ &+ 2K' \sup_{n\geq 1}\int_0^T \mathbf{E}\sum_{i=N}^{+\infty} |E_i^n(s)|\Bigg\}.\end{aligned} \tag{60}$$

Then, using Eqs. (55) and (56) into Eq. (60) we have

$$\lim_{N\to+\infty}\lim_{\varepsilon\to 0}\lim_{n\to 0} \mathbf{E} \sup_{t\in[0,T]} \sum_{i=2}^{N} \varphi_\varepsilon(E_i^n(s)) = 0. \tag{61}$$

Since $\varphi_\varepsilon(x) \geq |x|$ for all $x \geq 0$, for each $i \geq 2$, there exists N large enough such that

$$\mathbf{E} \sup_{t\in[0,T]} |E_i^n(t)| \leq \mathbf{E} \sup_{t\in[0,T]} \sum_{i=2}^{N} \varphi_\varepsilon(E_i^n(t))\,.$$

Thus, we deduce from Eq (61) that Eq. (42) holds for any $i \geq 2$. For $i = 1$, we have

$$|E_1^n(t)| \leq |c_1^n(t) - c_1(t)| + |E_2^n(t)| \leq 2|E_2^n(t)| + \sum_{i=2}^{N} |E_i^n(t)| + \sum_{i=N+1}^{+\infty} |E_i^n(t)|\,.$$

Hence from Eq. (55) and Eq. (42) for $i \geq 2$, we conclude that Eq (42) holds for $i = 1$ as well. Finally, we easily deduce

$$\lim_{n\to+\infty} \mathbf{E} \sup_{t\in[0,T]} |c_i^n(t) - c_i(t)| \leq \lim_{n\to+\infty} \mathbf{E} \sup_{t\in[0,T]} |E_i^n(t) - E_{i+1}^n(t)| = 0\,.$$

Since

$$\mathbf{E} \sup_{t\in[0,T]} \|c^n(t) - c(t)\| \leq \mathbf{E} \sup_{t\in[0,T]} \sum_{i=1}^{N} i|c_i^n(t) - c_i(t)| + \sup_{i\geq N} \frac{i}{\phi(i)} K_T + \sup_{i\geq N} \frac{i}{\tilde\phi(i)} K_T,$$

where ϕ and $\tilde\phi$ follows from Eqs. (58) and (59), we conclude that

$$\lim_{n\to+\infty} \mathbf{E} \sup_{t\in[0,T]} \|c^n(t) - c(t)\| = 0\,,$$

which ends the proof. □

5 Stationary measure

In this section, we prove Theorem 2.4. We start by some algebraic manipulations of the non-equilibirum potential given in Eq. (10). We recall that z_s is defined in Eq. (8). One has, for any $c \in \mathcal{E}_\rho^n$, and $z \leq z_s$,

$$\begin{aligned} -\frac{\rho}{n}\ln \Pi^n(c) &= \sum_{i=1}^{n} \left\{ -c_i \ln\left(\frac{n}{\rho} Q_i z^i\right) + \frac{\rho}{n} \ln \frac{n}{\rho} c_i! + Q_i z^i \right\} + \frac{\rho}{n} \ln B_n^z \\ &= \sum_{i=1}^{n} \left\{ c_i \left(\ln \frac{c_i}{Q_i z^i} - 1 \right) + Q_i z^i \right\} + R_n(c) + \frac{\rho}{n} \ln B_n^z \\ &= \mathcal{H}(c|c^z) - \sum_{i=n+1}^{\infty} Q_i z^i + R_n(c) + \frac{\rho}{n} \ln B_n^z \end{aligned} \tag{62}$$

(with convention $0 \ln 0 = 0$), where we recall that $\mathcal{H}$ is the relative entropy of the DBD equations, given in Eq. (10), and the term R_n is given by

$$R_n(c) = \frac{\rho}{n} \sum_{i=1}^{n} \left\{ \ln \frac{n}{\rho} c_i! - \frac{n}{\rho} c_i \ln \frac{n}{\rho} c_i + \frac{n}{\rho} c_i \right\}.$$

The proof of Theorem 2.4 is based on continuity properties of $\mathcal{H}$ and the convergence to 0 of each remaining term in Eq. (62), along appropriate sequences. We divide the proof in three lemmas. Let us start with a lemma about continuity properties of the functional $\mathcal{H}$, mainly from [3].

Lemma 5.1. *Assume $0 < z_s < +\infty$.*

1. *If $0 < z < z_s$ and $\liminf_{i\to+\infty} Q_i^{1/i} > 0$, then $\mathcal{H}(\cdot|c^z)$ is finite and sequentially strongly continuous on X.*

2. *If $z = z_s$ and $\lim Q_i^{1/i}$ exists, then $\mathcal{H}(\cdot|c^{z_s})$ is finite and sequentially $weak-*$ continuous on X.*

Proof. Point 1. Note that we may rewrite

$$\mathcal{H}(c|c^z) = G(c) - \rho\ln z - \sum_{i=1}^{+\infty} ic_i \ln Q_i^{1/i} + \sum_{i=1}^{+\infty} Q_i z^i\,,$$

where $G(c) = \sum_{i=1}^{+\infty} c_i(\ln c_i - 1)$. By [3, Lemma 4.2], G is finite and sequentially $weak-*$ continuous on X, hence also strongly continuous on X. As $z < z_s$, $\sum_{i=1}^{+\infty} Q_i z^i < \infty$. Next, $\{\ln(Q_i^{1/i})\}$ is bounded as $0 < \liminf Q_i^{1/i} \leq z_s^{-1} = \limsup Q_i^{1/i} < +\infty$. Thus, $c \mapsto \sum_{i=1}^{+\infty} ic_i \ln Q_i^{1/i}$ is finite and strongly continuous on X, and so is $\mathcal{H}$. The Point 2 is a consequence of [3, Proposition 4.5]. □

Now we state an intermediate Lemma which proves that the sum R_n in the non-equilibrium potential goes to 0.

Lemma 5.2. *Let $\{c^n\}$ a sequence belonging to $\mathcal{E}_\rho^n$ for each $n \geq 1$. We have*

$$\lim_{n\to+\infty} \sum_{i=1}^{n} \frac{\rho}{n}\left(\ln\frac{n}{\rho}c_i^n! - \frac{n}{\rho}c_i^n \ln\frac{n}{\rho}c_i^n + \frac{n}{\rho}c_i^n\right) = 0.$$

Proof. By Stirling's formula, there exists $K > 0$ such that for all $N \geq 2$

$$0 \leq \ln N! - N\ln N + N \leq K\ln N\,.$$

Hence, for all i such that $\frac{n}{\rho}c_i^n \geq 2$

$$0 \leq \frac{\rho}{n}\left(\ln\frac{n}{\rho}c_i^n! - \frac{n}{\rho}c_i^n \ln\frac{n}{\rho}c_i^n + \frac{n}{\rho}c_i^n\right) \leq K\frac{\rho}{n}\ln\frac{n}{\rho}c_i^n \tag{63}$$

We define, for all $i \geq 1$,

$$u_i^n = \begin{cases} \frac{\rho}{n}\left(\ln\frac{n}{\rho}c_i^n! - \frac{\rho}{n}c_i^n \ln\frac{n}{\rho}c_i^n + \frac{n}{\rho}c_i^n\right), & \text{if } \frac{n}{\rho}c_i^n \geq 2,\\ \frac{\rho}{n}, & \text{if } \frac{n}{\rho}c_i^n = 1,\\ 0, & \text{else.}\end{cases} \tag{64}$$

Since for all i, $c_i^n \leq \rho$, we have by Eqs. (63) and (64), that for all i, $u_i^n \to 0$ as $n \to +\infty$. Moreover, again by Eqs. (63) and (64), we can check that $u_i^n \leq Kc_i^n$ for all $i \geq 1$. Thus, using the mass conservation $\sum_{i=1}^n ic_i^n = \rho$, we deduce that, for all $N \geq 1$, and $n \geq N$,

$$R_n = \sum_{i=1}^{n} u_i^n \leq \sum_{i=1}^{N} u_i^n + K\sum_{i=N}^{n} c_i^n \leq \sum_{i=1}^{N} u_i^n + \frac{K}{N}\rho.$$

Taking the limit in $n \to +\infty$ and then $N \to +\infty$ ends the proof. □

In the last lemma, we control the convergence of the normalizing constant B_n^z. We recall that $z(\rho)$ is defined in Eq. (9).

Lemma 5.3. *Assume $0 < z_s < +\infty$.*

1. *If* $\rho \leq \rho_s$*, and* $\liminf_{i\to+\infty} Q_i^{1/i} > 0$*, we have, for* $z = z(\rho)$*,*

$$\lim_{n\to+\infty} \frac{\rho}{n} \ln B_n^{z(\rho)} = 0.$$

2. *If* $\rho > \rho_s$ *and* $\lim Q_i^{1/i}$ *exists, we have, for* $z = z_s$*,*

$$\lim_{n\to+\infty} \frac{\rho}{n} \ln B_n^{z_s} = 0.$$

Proof. For any $z > 0$, we have by Eq. (6) that

$$B_n^z \leq \sum_{C\in\mathbb{N}^n} \prod_{i=1}^n \frac{(\frac{n}{\rho} Q_i z^i)^{C_i}}{(C_i)!} e^{-\frac{n}{\rho} Q_i z^i} = 1\,,$$

hence $\frac{\rho}{n} \ln B_n^z \leq 0$ which entails

$$\limsup_{n\to+\infty} \frac{\rho}{n} \ln B_n^z \leq 0.$$

For $z \leq z_s$, and for any $x^n \in \mathcal{E}_\rho^n$, as $\Pi(x^n) \leq 1$, we deduce from Eq. (62) that

$$\frac{\rho}{n} \ln B_n^z \geq -H(x^n|c^z) - R_n(x^n) + \sum_{i=n+1}^{\infty} Q_i z^i\,. \tag{65}$$

Suppose first that $\rho \leq \rho_s$. Then $z(\rho) \leq z_s$, and we can find $x^n \in \mathcal{E}_\rho^n$ such that $x^n \to c^{z(\rho)}$ strongly (in norm) in X. Indeed, consider $x_i^n = \frac{\rho}{n}\lfloor \frac{n}{\rho} c_i^{z(\rho)} \rfloor$ for $i \leq n-1$ and $x_n^n = \frac{1}{n}\left(\rho - \sum_{i=1}^{n-1} i x_i^n\right)$. Clearly, x^n converges componentwise (thus $weak-*$) to $c^{z(\rho)}$. Moreover, $\|x^n\| = \rho = \|c^{z(\rho)}\|$ thus x^n also converges strongly (in norm) to c^z, see for instance [3, Lemma 3.3]. By Lemma 5.1 we have $H(x^n|c^{z(\rho)}) \to H(c^{z(\rho)}|c^{z(\rho)}) = 0$. As $c^{z(\rho)} \in X$, $\left(Q_i\,(z(\rho))^i\right)$ is summable and $\sum_{i=n+1}^{\infty} Q_i\,(z(\rho))^i \to 0$ as $n \to \infty$. And by Lemma 5.2, as $x^n \in \mathcal{E}_\rho^n$ for each n, $R_n(x^n) \to 0$ as $n \to \infty$. Thus, we deduce from Eq. (65) that $\liminf_{n\to\infty} \frac{\rho}{n} \ln B_n^{z(\rho)} \geq 0$.

Now take $\rho > \rho_s$. Consider $x_i^n = \frac{\rho}{n}\lfloor \frac{n}{\rho} c_i^{z_s} \rfloor$ for $i \leq n-1$ and $x_n^n = \frac{1}{n}\left(\rho - \sum_{i=1}^{n-1} i x_i^n\right)$. Then, $x^n \in \mathcal{E}_\rho^n$ and $weak-*$ converges towards c^{z_s}. Again, by Lemma 5.1 and Lemma 5.2, we deduce from Eq. (65) that $\liminf_{n\to\infty} \frac{\rho}{n} \ln B_n^{z_s} \geq 0$, which concludes the proof. □

We now conclude by the proof of theorem 2.4.

Proof of theorem 2.4. Suppose first $0 < \rho \leq \rho_s$. Choosing $z = z(\rho) \leq z_s$, in Eq. (62), we deduce from Lemma 5.1, Lemma 5.2 and Lemma 5.3 that

$$\lim_{n\to+\infty} -\frac{\rho}{n} \ln \Pi^n(c^n) = \mathcal{H}(c|c^{z(\rho)})\,.$$

Similarly, for $\rho > \rho_s$, choosing $z = z_s$ in Eq. (62), gives, with Lemma 5.1, Lemma 5.2 and Lemma 5.3 that

$$\lim_{n\to+\infty} -\frac{\rho}{n} \ln \Pi^n(c^n) = \mathcal{H}(c|c^{z_s})\,.$$

□

6 Discussion

In this section, we discuss our main results with respect to the literature. Both Theorems 2.2 and 2.3 are a kind of law of large numbers. However, their proof differs from the standard proof of the finite dimensional setting for continuous time Markov chain that converges to a solution of an ordinary differential equation, see the work by Kurtz in [11, Theorem 2.11] and [12, Theorem 2.2]. Indeed, under either hypothesis (H2) or (H3), the right-hand side of the limiting DBD system (1) may not be Lipschitz and the Kurtz strategy cannot be applied. In the proof of Theorem 2.3, we used monotonicity and convex properties to circumvent the lack of Lipschitz property. These arguments are essential in the proof of uniqueness of solution of the DBD equations, see [15, 14]. In the proof of Theorem 2.2, we used careful moment estimates and appropriate topological arguments. Again, the moment estimates were inspired from known results for the DBD equations [3, 15].

To be complete, let us mention that there are, up to our knowledge, two previous results for the law of large numbers on te SBD process. The first one is given by Jeon in [10], who proves a compactness result in $l^2(\mathbb{R})$, under linearly bounded coefficients (rather than in X). The focus of the work by Jeon was on more general coagulation-fragmentation models though, and on gelling solutions (that may arise in finite time for some coagulation-fragmentation models). The second work is by Sun in [18], who proves a strong law of large numbers (in the spirit of Kurtz theorem) using bounded kinetics rates. In such case, the right-hand side of the DBD system (1) is clearly Lipschitz on X. Then Sun was able to prove a functional central limit theorem, in a Hilbert subspace of $l^2(\mathbb{R})$. Our result in Theorem 2.2 needs that a_i to be $O(i)$, consistently with existence theorems for DBD equations (1), see [3]. We achieved the proof thanks to a new super-linear moment in Proposition 3.3. Such moments are well-known in general coagulation-fragmentation equations and seems to be derived for the first time in the stochastic context. Then, in Theorem 2.3, we state a pathwise convergence with assumption on the kinetic rates that are related to the uniqueness of the solution to DBD. Hence, we fit the stochastic theory of the Becker-Döring model to the most general results of existence and uniqueness available for the deterministic problem.

Limits of non-equilibrium potential are known to be related to relative entropy in general complex balanced stochastic chemical reaction networks, see for instance [2, 1]. We have thus extended theses results for an *infinite* chemical reaction network, that is *detailed balance*. Importantly, we have made the connection with the long-time behavior of the deterministic system. A challenge that remains is to investigate the interplay between the two limits $n \to \infty$ and $t \to \infty$, *simultaneously*.

A Criterion for weak compactness of density measures

A function between two topological spaces is said to be proper if the preimage of any compact set is compact. We said a family $\mathcal{F}$ of Borel measure on a complete separable metric space E is uniformly bounded if, $\sup_{\nu\in\mathcal{F}} \nu(E) < +\infty$, and uniformly tight if, for any $\varepsilon > 0$, there exists a compact K_ε of E such that $\sup_{\nu\in\mathcal{F}} \nu(E - K_\varepsilon) < \varepsilon$. We recall that the weak convergence of measure is the convergence of integrals against bounded continuous on E. For convenience reader, we write below a version of the Prohorov's theorem (see [4, Theorem 8.6.2]).

Theorem A.1 (Prohorov)**.** *Let $\mathcal{F}$ a family of Borel measure on a complete separable metric space. The following conditions are equivalent*

1. *$\mathcal{F}$ is relatively sequentially weakly compact.*
2. *$\mathcal{F}$ is uniformly bounded and tight.*

The aim of this section, is to state an alternative criterion of weak compactness, based on a refined version of the De La Vallé Poussin's theorem, see [7, Proposition I.1.1] and [14, Theorem 2.8]. We introduce a set of functions that have remarkable properties when conjugate to the structure of Becker-Döring equations and provide important estimates, see for instance [15].

Definition A.2. *We denote by $\mathcal{U}$ the set of non-negative convex functions ϕ, continuously differentiable with piecewise continuous second derivative, such that $\phi(x) = \frac{x^2}{2}$ for $x \in [0,1]$, ϕ' is concave, $\phi'(x) \leq x$ for $x > 0$, and*

$$\lim_{x\to+\infty} \phi(x)/x = +\infty.$$

One can obtain the following useful properties for the functions in $\mathcal{U}$:

Proposition A.1. *Let $\phi \in \mathcal{U}$. Then, ϕ is increasing, non-negative, and there exists $m > 0$ and $K > 0$ such that, for all $i \geq 1$*

$$\begin{aligned}
&(i+1)(\phi(i+1) - \phi(i) - \phi(1)) \leq m(i\phi(1) + \phi(i)),\\
&\phi(i+1) - \phi(i) - \phi(1) \geq 0, \\
&\phi''(i) \leq \phi''(0),\ \phi'(i) \leq i\phi''(i),\ \phi(i) \leq i\phi'(i),\ \text{and } \phi(i)/i^2 \leq K.
\end{aligned} \tag{66}$$

Proof. The first line in Eq. (66) follows from [13, Lemma 3.2]. The second line follows from the convexity inequality $\phi(i+1) - \phi(i) \geq \phi(1) - \phi(0) \geq 0$. The third line also follows directly from convexity properties. ☐

We state our alternative criterion of weak compactness in the following Theorem.

Theorem A.3. *Let $\{\nu^n\}$ be a sequence of Borel measure on a complete separabale metric space E and g be a non-negative proper continuous function. The sequence of density measure $\{g \cdot \nu^n\}$ is relatively weakly compact, if and only if, $\{g \cdot \nu^n\}$ is uniformly bounded and there exists $\phi \in \mathcal{U}$ such that*

$$\sup_{n\geq 1} \int_E \phi \circ g \, \nu^n < +\infty\,. \tag{67}$$

Proof. Assume that $\{g \cdot \nu^n\}$ is uniformly bounded such that Eq. (67) is satisfied for some $\phi \in \mathcal{U}$. Let $R > 0$ and define the compact $K = g^{-1}[0, R]$, then

$$\int_{E-K} g(x)\nu^n(dx) \leq \sup_{y>R} \frac{y}{\phi(y)} \int_E \phi(g(x))\nu^n(dx)\,.$$

Since $\phi \in \mathcal{U}$, the right hand side goes to 0 has $R \to \infty$, uniformly in n according to Eq. (67). Thus the sequence is uniformly tight. By the Prohorov theorem A.1, the sequence is relatively weakly compact.

Now assume $\{g\cdot\nu^n\}$ is relatively weakly compact, or equivalently, $\{g\cdot\nu^n\}$ is uniformly bounded and tight. We will follow the construction of ϕ proposed in [7, Proposition I.1.1] for uniform integrability. Define for each ν^n and $k \geq 0$, $M_k^n := \nu^n(\{k \leq g < k+1\})$. By construction of M_k^n it follows that

$$\sum_{k\geq 0} kM_k^n \leq \int_E g(x)\nu^n(dx).$$

Since the sequence $\{g\cdot\nu^n\}$ is uniformly bounded, $\sum_{k\geq 0} kM_k^n$ is also uniformly bounded, and we deduce

$$\sup_{n\geq 0}\sum_{k\geq 1}(k+1)M_k^n < +\infty. \tag{68}$$

Let $i\geq 1$. Since g is proper, the set $K_i=\{g\leq i\}$ is compact, and

$$\sum_{k\geq i}(k+1)M_k^n = \sum_{k\geq i}\int_{\{k\leq g<k+1\}}(k+1)\nu^n(dx) \leq 2\int_{E-K_i} g(x)\nu^n(dx). \tag{69}$$

The function g is continuous, hence bounded on the compacts. Thus, for any compact K, there exists i_0 such that $K\subset K_{i_0}$ and thus $E-K_{i_0}\subset E-K$. By uniform tightness, and Eq. (69), for all $m\geq 0$, there exists N_m such that

$$\sup_{n\geq 0}\sum_{k\geq N_m}(k+1)M_k^n < \frac{1}{(m+3)^3}.$$

Moreover the sequence $\{N_m\}$ can be chosen such that $N_0\geq 2$, $N_1\geq N_0$ and $N_{m+1}-N_m\geq N_m-N_{m-1}$ for all $m\geq 2$. We define the sequence 5

$$\alpha_k = \begin{cases} 2\,, & 0\leq k\leq N_0-1\,,\\ m+3\,, & N_m\leq k< N_{m+1}\,. \end{cases}$$

for all $k\geq 0$. Thus, we have

$$\begin{aligned}\sum_{k\geq 1}\alpha_{k+1}(k+1)M_k^n &= \sum_{k=1}^{N_0-1}\alpha_{k+1}(k+1)M_k^n + \sum_{m\geq 0}\sum_{k=N_m}^{N_{m+1}-1}\alpha_{k+1}(k+1)M_k^n\\ &\leq 3\sum_{k=1}^{N_0-1}(k+1)M_k^n + \sum_{m\geq 0}\frac{1}{(m+3)^2},\end{aligned}$$

and thanks to Eq. (68), it yields

$$\sup_{n\geq 0}\sum_{k\geq 1}\alpha_{k+1}(k+1)M_k^n < +\infty. \tag{70}$$

Now, we define the function p on $\mathbb{R}_+$ by

$$p(t) = \begin{cases} t & 0\leq t\leq 1\\ \dfrac{1}{N_0-1}t + \dfrac{N_0-2}{N_0-1} & 1\leq t\leq N_0\\ \dfrac{1}{N_{m+1}-N_m}t + \left(m+2-\dfrac{N_m}{N_{m+1}-N_m}\right) & N_m\leq t\leq N_{m+1},\ \forall m\in\mathbb{N}. \end{cases}$$

and, for all $y\geq 0$,

$$\phi(y) = \int_0^y p(t)\,dt.$$

Hence, for $x\leq 1$, $\phi(x)=x^2/2\leq x/2$. Let $k\geq 1$. It exists $m\geq 0$ such that $N_m\leq k+1< N_{m+1}$. Hence for all $t\leq k+1$, as p is increasing, $p(t)\leq p(N_{m+1})=m+3=\alpha_{k+1}$. Thus for all $k\geq 1$ we have $\phi(k+1)\leq (k+1)\alpha_{k+1}$. Then, we obtain,

$$\begin{aligned}\int_0^\infty \phi(g(x))\nu^n(dx) &\leq \int_{\{g(x)<1\}}\phi(g(x))\nu^n(dx) + \sum_{k\geq 1}\int_{\{k\leq g(x)<k+1\}}\phi(k+1)\nu^n(dx)\\ &\leq \frac{1}{2}\int_0^\infty g(x)\nu^n(dx) + \sum_{k\geq 1}(k+1)\alpha_{k+1}M_k^n\,.\end{aligned}$$

By hypothesis on $\{g \cdot \nu^n\}$ and the uniform bound (70), we obtain

$$\sup_{n\geq 0} \int_0^\infty \phi(g(x))\nu^n(dx) < +\infty.$$

The fact that ϕ belongs to $\mathcal{U}$ is easily checked by construction. □

B Tightness criterion for jump processes

Let E be a Polish space and d a complete metric that metrizes the topology on E, and $D([0,+\infty), E)$ the space of right continuous with left limit E-valued functions defined on $[0,+\infty)$ equipped with the Skorohod topology. For $\delta > 0$ and $T > 0$, the set Π_δ is the set of all partitions $\{t_i\}$ of $[0,T]$ such that for some N we have $0 = t_0 < t_1 < \cdots < t_{N-1} < T \leq t_N$ with $\min_{i=0,\ldots,N} |t_{i+1} - t_i| > \delta$. For any $x \in D([0,+\infty), E)$, the modulus of continuity is defined by

$$w(x,\delta,T) = \inf_{\{t_i\}\in\Pi_\delta} \max_i \sup_{s,t\in[t_i,t_{i+1}[} d(x_t, x_s),$$

The following lemma is classical for jump processes, but we prove it here for the convenience reader.

Lemma B.1. *Let $\{X^n\}$ be a sequence of pure jump Markov processes on E whose (stochastic) transition rate is given by $(\lambda_t^n)_{t\geq 0}$, for each $n \geq 1$. If there exists a positive sequence $\{\alpha_n\}$ such that*

$$\lim_{n\to+\infty} \alpha_n = 0,$$

and, almost surely,

$$\sup_{t\geq 0} \lambda_t^n \leq \alpha_n^{-1} \text{ and } d(X_t^n, X_{t^-}^n) \leq \alpha_n,$$

then, for all $T > 0$ and $\eta > 0$, there exists $\delta > 0$ such that

$$\lim_{n\to+\infty} \mathbf{P}\{w(X^n, T, \delta) \geq \eta\} \leq \eta.$$

Proof of Lemma B.1. Let $(\mathcal{F}_t^n)_{t\geq 0}$ the natural filtration associated to $(X_t^n)_{t\geq 0}$. We define $(N_t^n)_{t\geq 0}$ the counting process given by the jump times of $(X_t^n)_{t\geq 0}$. Namely,

$$N_t^n = \sum_{k\geq 0} \mathbf{1}_{t\geq \tau_k^n},$$

where the random sequence $\{\tau_k^n\}_{k\geq 0}$ is defined by, for each $n \geq 1$, $\tau_0^n = 0$ and

$$\tau_{k+1}^n = \inf\{t \,|\, t > \tau_k^n,\ X_t^n \neq X_{t^-}^n\}.$$

Hence $(N_t^n)_{t\geq 0}$ is a conditional Poisson process with stochastic intensity $(\lambda_t^n)_{t\geq 0}$ (see [5, Chap 2 Defintion D1]). In particular,

$$P\{N_t^n - N_s^n = k \,|\, \mathcal{F}_s\} = \mathbf{E}\left[\exp\left(-\int_s^t \lambda_\sigma^n d\sigma\right) \frac{\left(\int_s^t \lambda_\sigma^n d\sigma\right)^k}{k!}\right].$$

Fix $T > 0$ and $\eta > 0$. Consider the partition $t_i = i\Delta t$ for $i = 0, 1, \ldots, N$ of $[0,T]$ for some Δt (to be chosen later). For all $\delta < \Delta t$, we have $\{t_i\} \in \Pi_\delta$. We define

$$Y_i^n = \sup_{s,t\in[t_i,t_{i+1}[} d(X_t^n, X_s^n).$$

Then, we have

$$\mathbf{P}\{w(X^n,\delta,T)\geq\eta\}\leq\mathbf{P}\{\exists i\; Y_i^n\geq\eta\}\leq N\max_i\mathbf{P}\{Y_i^n\geq\eta\}. \tag{71}$$

We aim to bound $\mathbf{P}\{Y_i^n\geq\eta\}$ for each i. Let $i\in\{0,\ldots,N\}$. By hypothesis, any jump size of $(X_t^n)_{t\geq0}$ is less than α_n. Thus, in a time interval $[t_i,t_{i+1}[$, we need strictly more than $\lfloor\frac{\eta}{\alpha_n}\rfloor$ jumps so that $Y_i^n\geq\eta$. Thus,

$$\begin{aligned}\mathbf{P}\{Y_i^n\geq\eta\}\quad&\leq\mathbf{P}\left\{N_{t_{i+1}}^n-N_{t_i}^n\geq\left\lfloor\frac{\eta}{\alpha_n}\right\rfloor\right\}\\&\leq\mathbf{E}\left[e^{-\int_{t_i}^{t_{i+1}}\lambda_\sigma^n d\sigma}\sum_{k\geq\lfloor\frac{\eta}{\alpha_n}\rfloor}\frac{\left(\int_{t_i}^{t_{i+1}}\lambda_\sigma^n d\sigma\right)^k}{k!}\right].\end{aligned}$$

Since $x\mapsto e^{-x}\sum_{k\geq\lfloor\frac{\eta}{\alpha_n}\rfloor}\frac{x^k}{k!}$ is a non-decreasing function on $\mathbb{R}_+$, we have

$$\mathbf{P}\{Y_i^n\geq\eta\}\leq e^{-\alpha_n^{-1}\Delta t}\sum_{k\geq\lfloor\frac{\eta}{\alpha_n}\rfloor}\frac{\left(\alpha_n^{-1}\Delta t\right)^k}{k!}$$

The right hand side of this above inequality is the probability that a Poisson random variable with intensity $\alpha_n^{-1}\Delta t$ is greater than $\left\lfloor\frac{\eta}{\alpha_n}\right\rfloor$. Thus, by Chernoff's inequality, one may obtain, for all $x\in\mathbb{R}$,

$$\mathbf{P}\{Y_i^n\geq\eta\}\leq\exp\left(-x\left\lfloor\frac{\eta}{\alpha_n}\right\rfloor+\alpha_n^{-1}\Delta t(e^x-1)\right).$$

Choosing the minimizing value $x=\ln(\alpha_n\frac{\lfloor\frac{\eta}{\alpha_n}\rfloor}{\Delta t})$, we get

$$\mathbf{P}\{Y_i^n\geq\eta\}\leq\exp\left(\left\lfloor\frac{\eta}{\alpha_n}\right\rfloor\left(1-\frac{\Delta t}{\alpha_n\lfloor\frac{\eta}{\alpha_n}\rfloor}+\ln\frac{\Delta t}{\alpha_n\lfloor\frac{\eta}{\alpha_n}\rfloor}\right)\right).$$

The term in the above exponential is negative as soon as $\Delta_t\neq\alpha_n\lfloor\frac{\eta}{\alpha_n}\rfloor$. Then

$$\lim_{n\to+\infty}\mathbf{P}\{Y_i^n\geq\eta\}=0$$

and by Eq. (71) we conclude that for $\delta<\Delta t$,

$$\lim_{n\to+\infty}\mathbf{P}\{w(X^n,T,\delta)\geq\eta)=0,$$

which ends the proof. □

Acknowledgements

E. Hingant thanks the support by Universidad de Bío-Bío Poyecto Regular Interno n. 172708, Grupo de Investigación EDPMoA, and FONDECYT Iniciación n. 11170655.

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