THE ENTROPY METHOD FOR REACTION-DIFFUSION SYSTEMS WITHOUT DETAILED BALANCE: FIRST ORDER CHEMICAL REACTION NETWORKS

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Abstract. In this paper, the applicability of the entropy method for the trend towards equilibrium for reaction-diffusion systems arising from first order chemical reaction networks is studied. In particular, we present a suitable entropy structure for weakly reversible reaction networks without detail balance condition.

We show by deriving an entropy-entropy dissipation estimate that for any weakly reversible network each solution trajectory converges exponentially fast to the unique positive equilibrium with computable rates. This convergence is shown to be true even in cases when the diffusion coefficients of all but one species are zero.

For non-weakly reversible networks consisting of source, transmission and target components, it is shown that species belonging to a source or transmission component decay to zero exponentially fast while species belonging to a target component converge to the corresponding positive equilibria, which are determined by the dynamics of the target component and the mass injected from other components. The results of this work, in some sense, complete the picture of trend to equilibrium for first order chemical reaction networks.

1. Introduction and Main results

This paper investigates the applicability of the entropy method and proves the convergence to equilibrium for reaction-diffusion systems, which do not satisfy a detailed balance condition.

The mathematical theory of (spatially homogeneous) chemical reaction networks goes back to the pioneer works of e.g. Horn, Jackson, Feinberg and the Volperts, see [Fei79, Fei87, FH, Hor72, Hor74, HJ72, Vol, VVV] and the references therein. The aim is to study the dynamical system behaviour of reaction networks *independently of the values of the reaction rates*. It is conjectured since the early of 1970s that in a complex balanced system, the trajectories of the corresponding dynamical system always converge to a positive equilibrium. This conjecture was given the name Global Attractor Conjecture by Craciun et al. [CDSS]. The conjecture in its full generality is – up to our knowledge – still unsolved so far, despite many attempts have been made by mathematicians to attack this problem.

From the many previous works concerning the large time behaviour of chemical reaction networks, the majority of the existing results considers the spatially homogeneous ODE setting. The PDE setting in terms of reaction-diffusion systems is less studied. Also detailed quantitative statements like, e.g. rates of convergence to equilibrium, constitute frequently open questions even in the ODE setting.

Our general aim is to prove quantitative results on the large-time behaviour of chemical reaction networks modelled by reaction-diffusion systems. In the present work, we study reaction-diffusion systems arising from first order chemical reaction networks and show that all solution trajectories converge exponentially to corresponding equilibria with explicitly computable rates.

Our approach applies the so called entropy method. Going back to ideas of Boltzmann and Grad, the fundamental idea of the entropy method is to quantify the monotone decay of a suitable entropy (e.g. a convex Lyapunov) functional in terms of a *functional inequality* connecting the time-derivative of the entropy, the so called entropy dissipation functional, back to the entropy functional itself, i.e. to derive a so called *entropy entropy-dissipation (EED)* inequality. Such an EED inequality can only hold provided that all conservation laws are taken into account. After having established an EED inequality and applying it to global solutions of a dissipative evolutionary problem, a direct Gronwall argument implies convergence to equilibrium in relative entropy with rates and constants, which can be made explicit.

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By being based on functional inequalities (rather than on direct estimates on the solutions), a major advantage of the entropy method is its robustness with respect to model variations and generalisations. Moreover, the entropy method is per se a fully nonlinear approach.

The fundamental idea of the entropy method originates from the pioneer works of kinetic theory and from names like Boltzmann and Grad in order to investigate the trend to equilibrium of e.g. models of gas kinetics.

A systematic effort in developing the entropy method for dissipative evolution equations started not until much later, see e.g. the seminal works [Tos, TV, CJMTU, AMTU, DV01] and the references therein for scalar (nonlinear) diffusion or Fokker-Planck equations, and in particular the paper of Desvillettes and Villani concerning the trend to equilibrium for the spatial inhomogeneous Boltzmann equation [DV05]. The derivation of EED inequalities for scalar evolution equations is typically based on the Barky-Emery strategy (see e.g. [CJMTU, AMTU]), which seems to fail (or be too involved) to apply to systems.

The great challenge of the entropy method for systems is, therefore, to be able to derive an entropy entropy-dissipation inequality, which summarises (in the sense of measuring with a convex entropy functional) the entire dissipative behaviour of solutions to a (possibly nonlinear) dynamical system to which the EED inequality shall be applied to. Preliminary results based on a (non-explicit) compactness-contradiction argument in 2D were obtained e.g. in [Grö, GGH, GH] in the context semiconductor drift-diffusion models.

The first proof of an EED inequality with explicitly computable constants and rates for specific nonlinear reaction-diffusion systems was shown in [DF06] and followed by e.g. [DF07, DF08, DF15, FLT, MHM]. The application of these EED inequalities to global solutions of the corresponding reaction-diffusion systems proves (together with Csiszár-Kullback-Pinsker type inequalities) the explicit convergence to equilibrium for these reaction-diffusion systems.

We emphasise that all these previous results on entropy methods for systems assumed a *detailed balance condition* and, thus, features the free energy functional as a natural convex Lyapunov functional.

A main novelty of the paper lies in demonstrating how the entropy method can be generalised to first order reaction networks without detailed balance equilibria. In particular we shall consider firstly *weakly reversible networks* and secondly even more general *composite systems consisting of source, transmission and target components* (see below for the precise definitions).

We feel that it is important to point out that while there are certainly many classical approaches by which linear reaction-diffusion systems can be successfully dealt with, our task at hand is to clarify the entropic structure and the applicability of the entropy method for linear reaction networks as a first step before being able to turn to nonlinear problems in the future. See [DFT] for such a generalisation of the method to nonlinear reaction-diffusion systems satisfying the so-called complex balance condition (see Definition 1.3 below).

The goal of this present work is to prove the explicit convergence to equilibrium for the complex balanced and more general reaction-diffusion systems corresponding to first order reaction networks. To be more precise, we study first order reaction networks of the form

$$S_i \underset{a_{ji}}{\overset{a_{ij}}{\rightleftarrows}} S_j \qquad i \neq j = 1, 2, \ldots, N, \tag{$\mathcal{N}$}$$

FIGURE 1. A first-order chemical reaction network

where $S_i, i = 1, 2, \ldots, N$, are different chemical substances (or species) and $a_{ij}, a_{ji} \geq 0$ are reaction rate constants. In particular, a_{ij} denotes the reaction rates from the species S_j to S_i.

First order reaction networks appear in many classical models, see e.g. [Smo, Rot]. More recently, first order catalytic reactions are used to model transcription and translation of genes in [TVO]. The evolution of the surface morphology during epitaxial growth involves the nucleation and growth of atomic islands, and these processes may be described by first order adsorption and desorption reactions coupled with diffusion along the surface. A first order reaction network can also be used to describe the reversible transitions between various conformational states of proteins (see e.g. [MGetal]). RNA also exists in several conformations, and the transitions between various folding states follow first order kinetics (see [BRetal]).

In the present paper, we investigate the entropy method and the trend to equilibrium of reaction-diffusion systems modelling first order reaction networks with mass action kinetics. More precisely, we shall consider the reaction network $\mathcal{N}$ in the context of reaction-diffusion equations and assume that for all $i = 1, 2, \ldots, N$ the substances S_i are described by spatial-temporal concentrations $u_i(x,t)$ at position $x \in \Omega$ and time $t \geq 0$. Here, Ω shall denote a bounded domain $\Omega \subset \mathbb{R}^n$ with sufficiently smooth boundary $\partial\Omega$ (that is $\partial\Omega \in C^{2+\alpha}$ to avoid all difficulties with boundary regularity, although the below methods should equally work under weaker assumptions) and the outer unit normal $\nu(x)$ for all $x \in \partial\Omega$. Due to the rescaling $x \to |\Omega|^{1/n}x$, we can moreover consider (without loss of generality) domains with normalised volume, i.e.

$$|\Omega| = 1.$$

In addition, we assume that each substance S_i diffuses with a diffusion rate $d_i \geq 0$ for all $i = 1, 2, \ldots, N$. Finally, we shall assume mass action law kinetics as model for the reaction rates, which leads to the following linear reaction-diffusion system:

$$\begin{cases} X_t = D\Delta X + AX, & x \in \Omega, \quad t > 0, \\ \partial_\nu X = 0, & x \in \partial\Omega, \quad t > 0, \\ X(x,0) = X_0(x) \geq 0, & x \in \Omega, \end{cases} \tag{1.1}$$

where $X(x,t) = [u_1(x,t), u_2(x,t), \ldots, u_N(x,t)]^T$ denotes the vector of concentrations subject to non-negative initial conditions $X_0(x) = [u_{1,0}(x) \geq 0, u_{2,0}(x) \geq 0, \ldots, u_{N,0}(x) \geq 0]^T$, $D = \mathrm{diag}(d_1, d_2, \ldots, d_N)$ denotes the diagonal diffusion matrix and the reaction matrix $A = (a_{ij}) \in \mathbb{R}^{N\times N}$ satisfies the following conditions:

$$\begin{cases} a_{ij} \geq 0, & \text{for all } i \neq j, \quad i,j = 1,2,\ldots,N, \\ a_{jj} = -\sum_{i=1, i\neq j}^N a_{ij}, & \text{for all } j = 1,2,\ldots,N. \end{cases} \tag{1.2}$$

The conditions (1.2) on the reaction matrix A imply in particular that the vector $(1,1,\ldots,1)^T$ constitutes a left-eigenvector corresponding to the eigenvalue zero. Together with homogeneous Neumann boundary conditions this implies that solutions to (1.1) admit the following *conservation of total mass*:

$$\sum_{i=1}^N \int_\Omega u_i(x,t)dx = \sum_{i=1}^N \int_\Omega u_{i,0}(x)dx =: M > 0, \qquad \text{for all } t > 0, \tag{1.3}$$

where $M > 0$ is the *initial total mass*, which we shall assume positive.

If $X(x,t) \equiv X(t)$, then system (1.1) reduces to the corresponding space-homogeneous ODE model. Independently of PDE- or ODE-setting, we recall the following definitions of equilibria from e.g. [HJ72, Fei79, VVV].

Definition 1.1 (Homogeneous Equilibrium)**.**
A state $X_\infty = (u_{1,\infty}, u_{2,\infty}, \ldots, u_{N,\infty})$ is called a homogeneous equilibrium or shortly equilibrium of the first order reaction network $\mathcal{N}$ if $AX_\infty = 0$.

Definition 1.2 (Detailed Balance Equilibrium)**.**
A positive equilibrium state $X_\infty = (u_{1,\infty}, u_{2,\infty}, \ldots, u_{N,\infty}) > 0$ is called a detailed balance equilibrium for the reaction network $\mathcal{N}$ if a positive reaction rate constant $a_{ij} > 0$ for $i \neq j$ implies also a positive reversed reaction rate constant $a_{ji} > 0$ and that the forward and backward reaction rates balance at equilibrium, i.e.

$$a_{ji}u_{i,\infty} = a_{ij}u_{j,\infty}$$

The reaction network $\mathcal{N}$ is called to satisfy the detailed balance condition if it admits a detailed balance equilibrium.

Definition 1.3 (Complex Balance Equilibrium)**.**
A positive equilibrium state $X_\infty = (u_{1,\infty}, u_{2,\infty}, \ldots, u_{N,\infty}) > 0$ is called a complex balance equilibrium for the reaction network $\mathcal{N}$ if for all $k = 1, 2, \ldots, N$, the total in-flow into the substance S_k balances in equilibrium the total out-flow from S_k to all other substances S_i, i.e.

$$\sum_{\{1\leq i\leq N:\, a_{ki}>0\}} a_{ki}u_{i,\infty} = \Bigg(\sum_{\{1\leq j\leq N:\, a_{jk}>0\}} a_{jk}\Bigg)u_{k,\infty}.$$

The reaction network $\mathcal{N}$ is called complex balanced if it admits a complex balance equilibrium. Moreover for complex balanced chemical reaction networks, all equilibria are complex balanced, see e.g. [Hor72].

Example 1.1 (Detailed balance equilibria are complex balance equilibria)**.**
It is easy to see that detailed balance equilibria are also complex balance equilibria while the reverse does not hold in general, even for reversible networks. For example, consider the reaction network in Figure 2, where all reaction rates constants $a_{ij} > 0$ are assumed positive and the network is thus fully reversible.

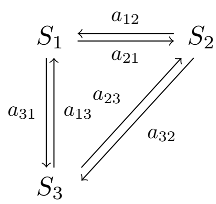


FIGURE 2. A reversible network

The corresponding reaction-diffusion system with homogeneous Neumann boundary conditions

$$\begin{cases} \partial_t u_1 - d_1\Delta u_1 = -(a_{21}+a_{31})u_1 + a_{12}u_2 + a_{13}u_3, \\ \partial_t u_2 - d_2\Delta u_2 = a_{21}u_1 - (a_{12}+a_{32})u_2 + a_{23}u_3, \\ \partial_t u_3 - d_3\Delta u_3 = a_{31}u_1 + a_{32}u_2 - (a_{13}+a_{23})u_3, \\ \partial_\nu u_1 = \partial_\nu u_2 = \partial_\nu u_3 = 0. \end{cases} \tag{1.4}$$

exhibits the constant equilibrium $X_\infty = (u_{1,\infty}, u_{2,\infty}, u_{3,\infty})$ satisfying $AX_\infty = 0$, i.e.

$$\begin{cases} a_{12}u_{2,\infty} + a_{13}u_{3,\infty} = (a_{21}+a_{31})u_{1,\infty}, \\ a_{21}u_{1,\infty} + a_{23}u_{3,\infty} = (a_{12}+a_{32})u_{2,\infty}, \\ a_{31}u_{1,\infty} + a_{32}u_{2,\infty} = (a_{13}+a_{23})u_{3,\infty}, \end{cases} \tag{1.5}$$

which has a unique nontrivial solution once the mass conservation (1.3) is taken into account.

According to Definition 1.3, it is clear that system (1.5) constitutes a complex balance equilibrium for all reaction rate constants $a_{ij} > 0$. For X_∞ to be a detailed balance equilibrium, however, it is additionally necessary that

$$\begin{cases} a_{12}u_{2,\infty} = a_{21}u_{1,\infty}, \\ a_{23}u_{3,\infty} = a_{32}u_{2,\infty}, \\ a_{31}u_{1,\infty} = a_{13}u_{3,\infty}, \end{cases} \tag{1.6}$$

which obviously implies (1.5). Yet the equations (1.6) can only have a solution if

$$\frac{a_{12}\cdot a_{23}\cdot a_{31}}{a_{21}\cdot a_{32}\cdot a_{13}} = 1, \tag{1.7}$$

holds; in other words if the product of the reaction rate constants multiplied in the clockwise sense of the above reaction network graph equals the product of the reaction rate constants multiplied in the counterclockwise sense. The condition (1.7) is thus necessary and sufficient for system (1.4) to admit a detailed balance equilibrium.

Remark 1.1 (General definition of detailed and complex balance).
The concepts of detailed balance and complex balance are also defined for general higher order chemical reaction networks, see e.g. [Hor72]. For simplicity, we stated here the definition corresponding to the first order network $\mathcal{N}$. In general, one can roughly say that a state X_∞ is called a complex balanced equilibrium if at equilibrium the total in-flow to each specie S_i is equal to the total out-flow from S_i.

Remark 1.2 (Detailed balance and reversibility).
It follows from Definition (1.2) that if $\mathcal{N}$ satisfies the detailed balance condition, then it is also reversible in the sense that for any reaction $S_i \to S_j$ also the reverse reaction $S_j \to S_i$ takes place.

The set of complex balanced systems is much larger than the one of detailed balance systems. Horn already gave necessary and sufficient conditions for a network to satisfy the complex balance condition in [Hor72]. For convenience of the reader, we present in the following the associated definitions of directed graphs as representations of reaction networks. The image of the associated graphs will also help following some of our main estimates.

A directed graph G corresponding to a given reaction network $\mathcal{N}$ is defined by considering the substances $S_i, i = 1,2,\ldots,N$, as the N nodes of G, which are connected for all $i \neq j = 1,2,\ldots,N$ by an edge with starting node S_i and finishing node S_j if and only if the reaction $S_i \xrightarrow{a_{ji}} S_j$ occurs with a positive reaction rate constant $a_{ji} > 0$.

Definition 1.4 (Linkage classes partition of a first order reaction network, Connected networks)**.**
A linkage class $\mathcal{L}$ of a first order network $\mathcal{N}$ is a maximal set of connected substances, i.e. $S_i, S_j \in \mathcal{L}$ implies that S_i and S_j are connected (in the sense that there exist $S_i \equiv S_{r_1}, S_{r_2} \ldots, S_{r_{k-1}}, S_{r_k} \equiv S_j$ such that for each $1 \le \ell \le k-1$, either the reaction $S_{r_\ell} \to S_{r_{\ell+1}}$ or $S_{r_{\ell+1}} \to S_{r_\ell}$ happens) but $S_i \in \mathcal{L}$ and $S_j \notin \mathcal{L}$ implies that S_i and S_j are not connected.

If a reaction network consists only of one linkage class, we shall call such a network connected.

Definition 1.5 (Weak reversibility of a first order reaction network)**.**
A first order reaction network $\mathcal{N}$ is called weakly reversible if for any reaction $S_i \to S_j$ with $i \neq j$, there exists a chain of reactions $S_j \equiv S_{j_1} \to S_{j_2} \to \ldots \to S_{j_r} \equiv S_i$ where $S_{j_1}, S_{j_2}, \ldots, S_{j_r}$ are other chemical substances of $\mathcal{N}$.

If a reaction network $\mathcal{N}$ is weakly reversible, then we also call the corresponding directed graph G weakly reversible.

Definition 1.6 (Strongly connected components of a directed graph)**.**
A subgraph $H \subset G$ of a directed graph G is called a strongly connected component if for any two nodes S_i, S_j in H, we can find a path from S_i to S_j of the form $S_i \to S_{i_1} \to \ldots \to S_{i_r} \to S_j$ with all $S_{i_1}, S_{i_2}, \ldots, S_{i_r}$ belonging to H.

We call a first order reaction network $\mathcal{N}$ strongly connected when its corresponding graph G is strongly connected.

Remark 1.3 (Partition of weakly reversible first order reaction networks $\mathcal{N}$ into disjoint strongly connected components/subnetworks)**.**
Firstly, it follows directly from Definition 1.4 that any first order reaction network $\mathcal{N}$ can be uniquely partitioned into a pairwise disjoint union of linkage classes and each linkage class $\mathcal{L}$ constitutes a connected subnetwork $\mathcal{N}_{\mathcal{L}}$. In particular, for a weakly reversible first order reaction network $\mathcal{N}$, each linkage class $\mathcal{L}$ forms a connected weakly reversible subnetwork $\mathcal{N}_{\mathcal{L}}$ and it is straightforward to show that the directed graph corresponding to $\mathcal{N}_{\mathcal{L}}$ is strongly connected according to Definition 1.6. (Consider that for all reactions being part of the connection between $S_i, S_j \in \mathcal{N}_{\mathcal{L}}$, the weak reversibility implies the existence of a returning chain of reactions. Thus, there exist chains of reactions connecting S_i to S_j and vice versa.) black Secondly, any directed graph G can be partitioned into a pairwise disjoint union of strongly connected components, all of which are weakly reversible according to Definition 1.5. Note that these strongly connected components can still be connected via "non-weakly-blackreversible" reactions (see e.g. Figure 3). Therefore, for general directed graphs, multiple strongly connected components may constitute one linkage class. However, if the directed graph G is additionally weakly reversible, then each strongly connected component has to constitute exactly one linkage class since otherwise we have already seen that weakly reversible subnetworks $\mathcal{N}_{\mathcal{L}}$ corresponding to one linkage class $\mathcal{L}$ are strongly connected.

Thus, for weakly reversible first order reaction networks $\mathcal{N}$, the partition of linkage classes is identical to the partition of strongly connected components of the corresponding directed graphs.

Therefore, with a marginal abuse of notation, we will use the terminology "strongly connected component" or "strongly connected subnetwork" both for such a connected weakly reversible first order reaction subnetwork $\mathcal{N}_{\mathcal{L}}$ and its corresponding strongly connected subgraph/component.

Remark 1.4 (Linkage classes of first order reaction networks can be treated independently)**.**
For first order reaction networks, each node represents exactly one substance. Thus, any linkage class of a first order reaction network can be treated independently from the others. In particular, all the strongly connected components of a weakly reversible first order reaction network can be treated independently since these subnetworks form different linkage classes.

For higher order reaction networks, where the nodes of the corresponding graphs are so-called complexes consisting of multiple substances, this is not necessarily true since one substance might need to be represented by different nodes.

Because of Remarks 1.3 and 1.4, we will consider in Section 2 weakly reversible first order networks partitioned into strongly connected first order reaction subnetworks $\mathcal{N}_{\mathcal{L}}$, and each strongly connected component $\mathcal{N}_{\mathcal{L}}$ can (w.l.o.g) be treated independently. In Section 3, we will consider (w.l.o.g) connected reaction networks $\mathcal{N}$ consisting of one linkage class, yet we shall not assume weak reversibility. Hence the corresponding directed graphs are not strongly connected and may consists of multiple strongly connected components, but the underlying undirected graphs are connected (see e.g. Figure 3).

Lemma 1.1 (Strongly connected networks, irreducible reaction matrices and complex balance equilibria)**.**
For any first order reaction network $\mathcal{N}$ the following statements are equivalent:

- *The first order reaction network $\mathcal{N}$ is strongly connected.*
- *The corresponding reaction matrix A of $\mathcal{N}$ is irreducible.*
- *The first order reaction network $\mathcal{N}$ is complex balanced and for any positive mass $M > 0$ (as set by the conservation law (1.3)), and there exists of a unique, positive complex balance equilibrium $X_\infty = (u_{1,\infty}, u_{2,\infty}, \ldots, u_{N,\infty}) > 0$ of system (1.1), which satisfies*

$$\begin{cases} AX_\infty = 0, \\ \sum_{i=1}^N u_{i,\infty} = M > 0. \end{cases} \tag{1.8}$$

Proof. The equivalence of strong connectivity for first order networks and irreducibility of the reaction matrix A follows e.g. from [Sen81, Definition 2.1, page 46] and [Min88, Theorem 3.2, page 78]. Next, the Perron-Frobenius theorem implies for any irreducible reaction matrix A and any positive mass $\sum_{i=1}^N u_{i,\infty} = M > 0$ the existence of a unique positive equilibrium, see e.g. [Sen81, Per07] and Lemma 2.2 below. This equilibrium satisfies $AX_\infty = 0$ and is thus a complex balance equilibrium according to Definition 1.3. Hence, the strongly connected first order reaction network $\mathcal{N}$ is complex balanced (independently of the value of M). Finally, Lemma 2.2 below implies that strongly connected first order reaction networks possessing unique positive equilibrium (for fixed $M > 0$) have irreducible reaction matrices A. □

Remark 1.5 (Complex balanced higher order systems are necessarily weakly reversible)**.**
For higher order reaction network, it holds only true that systems with complex balance equilibrium are necessarily weakly reversible. Thus, weakly reversible systems constitute the more general class of reaction networks.

Remark 1.6. *The equilibrium X_∞ in (1.8) is spatially homogeneous. Thus, it coincides with the equilibrium for the corresponding spatially homogeneous ODE system $X_t = AX$ of the reaction network given in Figure 1. In [And] or [SiMa], the authors proved that $X(t) \longrightarrow X_\infty$ as $t \longrightarrow +\infty$. However, the method used in this paper cannot be directly applied to prove the convergence to equilibrium for PDE system (1.1).*

The first main result of this paper concerns the convergence to equilibrium for weakly reversible reaction networks of the form displayed in Figure 1. Our method of proof applies the entropy method to prove explicit exponential convergence of solutions of system (1.1) to the unique equilibrium.

As mentioned above, all previous results of explicit EED inequalities (see e.g. [DF06, DF07, DF08, DF15, FLT, MHM]) considered reaction-diffusion systems satisfying a detailed balance condition.

In the current paper, we shall show that the following quadratic relative entropy between any two solutions $X = (u_1, \ldots, u_N)$ and $Y = (v_1, \ldots, v_N)$

$$\mathcal{E}(X|Y)(t) = \sum_{i=1}^N \int_\Omega \frac{|u_i|^2}{v_i} dx \tag{1.9}$$

is an entropy functional, see Lemma 2.3 below, which is the first key result of this paper.

In particular, we can consider the special case $Y = X_\infty$ for such an entropy functional. By using the linearity of first order systems, it is then straightforward to check (by using (1.2) and $AX_\infty = 0$) that the quadratic relative entropy towards an equilibrium state X_∞, i.e.

$$\mathcal{E}(X - X_\infty|X_\infty) = \sum_{i=1}^N \int_\Omega \frac{|u_i - u_{i,\infty}|^2}{u_{i,\infty}} dx \tag{1.10}$$

is equally an entropy functional, which decays monotone in time according to the following explicit form of the entropy dissipation functional $\frac{d}{dt}\mathcal{E}(X - X_\infty|X_\infty) = -\mathcal{D}(X - X_\infty|X_\infty)$:

$$\begin{aligned} \mathcal{D}(X - X_\infty|X_\infty) &= 2\sum_{i=1}^N d_i \int_\Omega \frac{|\nabla(u_i - u_{i,\infty})|^2}{u_{i,\infty}} dx \\ &\quad + \sum_{i,j=1;i<j}^N (a_{ji}u_{i,\infty} + a_{ij}u_{j,\infty}) \int_\Omega \left(\frac{u_i - u_{i,\infty}}{u_{i,\infty}} - \frac{u_j - u_{j,\infty}}{u_{j,\infty}}\right)^2 dx \geq 0 \\ &= 2\sum_{i=1}^N d_i \int_\Omega \frac{|\nabla u_i|^2}{u_{i,\infty}} dx + \sum_{i,j=1;i<j}^N (a_{ji}u_{i,\infty} + a_{ij}u_{j,\infty}) \int_\Omega \left(\frac{u_i}{u_{i,\infty}} - \frac{u_j}{u_{j,\infty}}\right)^2 dx \\ &= \mathcal{D}(X|X_\infty) = -\frac{d}{dt}\mathcal{E}(X|X_\infty) \geq 0. \end{aligned} \tag{1.11}$$

The dissipative structure of the quadratic relative entropy towards equilibrium (1.10) is a special cases of generalised relative entropies discussed e.g. in [Per07, Chapter 6]. The entropy functional (1.9), i.e. the observation of the dissipativeness of the relative entropy between any two solutions, is however related to a general property of linear Markow processes, which was recently shown in [FJ16].

With the help of the explicit form of entropy dissipation (1.11), we are able to show (in Lemma 2.4 below) an entropy-entropy dissipation inequality of the form

$$\mathcal{D}(X - X_\infty | X_\infty) \geq \lambda\, \mathcal{E}(X - X_\infty | X_\infty), \tag{1.12}$$

where $\lambda > 0$ is an explicitly computable constant. Once the EED inequality (1.12) is proven, the statement of the first main theorem follows from a standard Gronwall argument, see Section 2 below:

Theorem 1.2 (Exponential equilibration of weakly reversible first order reaction networks)**.** *Given a weakly reversible first order reaction network partitioned into linkage classes. Consider (w.l.o.g.) any corresponding strongly connected subnetwork $\mathcal{N}_{\mathcal{L}}$. Assume for $\mathcal{N}_{\mathcal{L}}$ that the diffusion coefficients d_i are positive for all $i = 1, 2, \ldots, N$, and the initial mass M is positive.*

Then, the unique global solution to initial-boundary problem (1.1) converges exponentially to the unique positive equilibrium $X_\infty = (u_{1,\infty}, u_{2,\infty}, \ldots, u_{N,\infty})$, i.e.

$$\sum_{i=1}^N \int_\Omega \frac{|u_i(t) - u_{i,\infty}|^2}{u_{i,\infty}} dx \leq e^{-\lambda t} \sum_{i=1}^N \int_\Omega \frac{|u_{i,0} - u_{i,\infty}|^2}{u_{i,\infty}} dx,$$

where the constant $\lambda > 0$ depends explicitly on the reaction matrix A, the domain Ω, the diffusion matrix D and the initial mass M.

Remark 1.7 (Lyapunov functionals for ODE systems)**.** *For ODE systems, Lyapunov functionals have been mainly considered in the analysis of nonlinear ODE systems. Moreover, for nonlinear ODE systems, L^1-type Lyapunov functionals are most commonly used in the study of the large-time-behaviour. For reaction-diffusion systems, however, L^1-functionals are not useful for the entropy method and proving explicit convergence to equilibrium, since they do not measure the spatial diffusion in an exploitable way.*

We also remark, that while logarithmic relative entropy functionals of the form

$$V_{X_\infty}(X)(t) = \sum_{i=1}^N \left(u_i(\ln u_i - \ln u_{i,\infty} - 1) + u_{i,\infty}\right) \tag{1.13}$$

were known to constitute a monotone decaying Lyapunov functional for complex balanced ODE reaction networks (see e.g. [HJ72, Gop, SiMa]), up to our knowledge and somewhat surprisingly, no explicit expression of the entropy dissipation $-dV/dt$ in complex balanced systems has been derived so far.

We also refer the reader to e.g. [MiSi] for the stability of some mass action law reaction-diffusion systems, where the author used techniques of ω-limit sets along with the monotonicity of L^1-type Lyapunov functional.

Our results in this paper are significantly stronger in the sense that we show, by using the entropy method, the exponential convergence to equilibrium with computable rates.

In addition and in comparison to ω-limit techniques, the entropy method has also the major advantage of relying on functional inequalities rather than on specific estimates of solutions to a given system. Having such functional entropy entropy-dissipation inequalities once and for all established makes the entropy method robust with respect to model variations and generalisations.

As example, it is the intrinsic robustness of the entropy method, which makes it possibly to also apply to non weakly reversible reaction networks, see Theorems 1.4 and 1.5 below.

The assumption on the positivity of all diffusion coefficients in Theorem 1.2 is not necessary as such. As already shown in e.g. [DF07, FLT], the combined effect of diffusion of a specie and its weakly reversible reaction with other (possibly non-diffusive) species will lead to a indirect "diffusion-effect" on the latter specie. This indirect diffusion-effect can also be measured in terms of functional inequalities. Hence the exponential convergence to equilibrium still holds for systems with partial degenerate diffusion.

Note that the indirect "diffusion transfer" and the convergence results of this paper resembles to some degree the framework of hypocoercivity for evolution equations like linear kinetic Fokker-Planck equations, see e.g. [Vil09, DMS, AAS]. However, while hypocoercivity typically requires the use of suitably constructed Lyapunov functionals, the indirect "diffusion-effect" can be entirely express in functional inequalities linking the relative entropy and the associated entropy dissipation functional. The entropy method present in this paper proves convergence to equilibrium essentially regardless of full- or degenerate diffusion matrices.

The exponential convergence for weakly reversible systems (1.1) with degenerate diffusion is stated in the following Theorem 1.3 to be proved in Section 2 below:

Theorem 1.3 (Equilibration of linear networks with degenerate diffusion)**.**
Given a weakly reversible first order reaction network partitioned into linkage classes. Consider (w.l.o.g.) any corresponding strongly connected subnetwork $\mathcal{N}_{\mathcal{L}}$. Assume that the initial mass M is positive for $\mathcal{N}_{\mathcal{L}}$. Moreover, assume that at least one diffusion coefficient d_i is positive for some $i = 1, 2, \ldots, N$.

Then, the solution to (1.1) converges exponentially fast to the unique positive equilibrium $X_\infty = (u_{1,\infty}, u_{2,\infty}, \ldots, u_{N,\infty})$:

$$\sum_{i=1}^{N}\int_\Omega \frac{|u_i(t) - u_{i,\infty}|^2}{u_{i,\infty}}dx \le e^{-\lambda' t}\sum_{i=1}^{N}\int_\Omega \frac{|u_{i,0} - u_{i,\infty}|^2}{u_{i,\infty}}dx$$

with a computable rate $\lambda' > 0$, which depends explicitly on A, Ω, D and M.

Remark 1.8 (Same results of linear ODE reaction networks)**.**
We remark that our approach can of course be adapted to equally apply to linear ODE reaction networks by eliminating the terms and calculations concerning spatial diffusion. Thus, all the results of this paper hold equally for such linear ODE systems.

As the second main result of this manuscript, we shall derive an entropy approach and prove convergence to equilibrium for reaction networks as in Figure 1, for which *the weak reversibility assumption does not hold*. For first order reaction networks, this implies that the system is not complex balanced, or in other words, that equilibria are not necessarily positive.

Due to the lack of positivity of equilibria, it follows immediately that the relative entropy used for weakly reversible systems is not directly applicable. In the following we proposed a modified entropy approach. At first, it is necessary to understand the structure of non weakly reversible reaction networks.

We state here the necessary terminology and the main ideas. Since for any non weakly reversible linkage class, the associated directed graph G is connected (which means that the underlying undirected version of G is a connected graph) but not strongly connected, G consists of $r \ge 2$ strongly connected components, which we denote by $C_1, C_2, \ldots, C_r$. Then, we can construct a *directed acyclic graph* G^C, i.e. G^C is a directed graph with no directed cycles as follows:

a) G^C has as nodes the r strongly connected components $C_1, C_2, \ldots, C_r$,
b) for two nodes C_i and C_j of G^C, if there exists a reaction $C_i \ni S_k \xrightarrow{a_{\ell k}} S_\ell \in C_j$ with $a_{\ell k} > 0$, then there exists also the edge $C_i \to C_j$ on G^C.

Due to the structure of G^C, its nodes, or equivalently the strongly connected components of G, can be labeled as one of the following three types:

- A strongly connected component C_i is called a *source component* if there is no in-flow to C_i, i.e. there does not exist an edge $S_k \to S_j$ where $S_k \notin C_i$ and $S_j \in C_i$.
- A strongly connected component C_i is called a *target component* if there is no out-flow from C_i, i.e. there does not exist and edge $S_k \to S_j$ where $S_k \in C_i$ and $S_j \notin C_i$.
- If C_i is neither a source component nor a target component, then we call C_i a *transmission component.*

Example 1.2. *Consider the reaction network in Figure 3. The depicted network has* 4 *strongly connected*

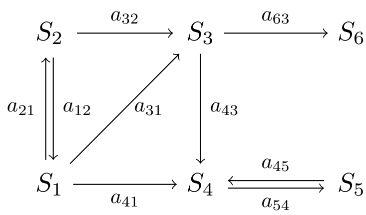


FIGURE 3. A non-weakly reversible reaction network consisting of four strongly connected components

components $C_1 = \{S_1, S_2\}$, $C_2 = \{S_3\}$, $C_3 = \{S_4, S_5\}$, $C_4 = \{S_6\}$, where C_1 is a source component, C_2 is a transmission component and C_3, C_4 are target components.

By definition, each of the three types of strongly connected components is subject to a different dynamic, which can be written as follows: Let C_i be a strongly connected component and denote by X_i the concentrations within C_i. Moreover, denote by A_i the reaction matrix formed by all reactions within the component C_i. Then, we have

- for a source component C_i:
$$\partial_t X_i - D_i \Delta X_i = A_i X_i + \mathcal{F}_i^{out},$$
where $\mathcal{F}_i^{out}$ summarises the out-flow from the source component C_i.
- for a target component C_i:
$$\partial_t X_i - D_i \Delta X_i = \mathcal{F}_i^{in} + A_i X_i,$$
where $\mathcal{F}_i^{in}$ summarises the in-flow into the target component C_i.
- for a transmission component C_i:
$$\partial_t X_i - D_i \Delta X_i = \mathcal{F}_i^{in} + A_i X_i + \mathcal{F}_i^{out},$$
where $\mathcal{F}_i^{in}$, $\mathcal{F}_i^{out}$ are the in/out-flow of the transmission component C_i.

In the dynamics of transmission and target components, the in-flow $\mathcal{F}_i^{in}$ depends only on species which do not belong to C_i, so that $\mathcal{F}_i^{in}$ can be treated as an external source for the system for C_i. However, it may happen that $\mathcal{F}_i^{in}$ contains inflow from species whose behaviour is not a-priori known.

For acyclic graphs G^C, however, it is possible to avoid these difficulties, since the *topological order* of acyclic graphs allows to re-order the r strongly connected components $C_1, C_2, \ldots, C_r$ in such a way that for every edge $C_i \to C_j$ of G^C it holds that $i < j$. This permits to study the dynamics of all components C_i sequentially according to the topological order and, when at times considering a transmission component (or later a target component) C_i, the required in-flow $\mathcal{F}_i^{in}$ contains only species whose behaviour is already known.

Due to the structure of the network, it is expected that species belonging to source or transmission components are subsequently losing mass such that the concentrations decay to zero in the large-time behaviour as time goes to infinity. In contrast, the species belonging to a target component converge to an equilibrium state, which is determined by the reactions within this component and by the mass "injected" from other components.

Since the source and transmission components do not converge to positive equilibria, the relative entropy method used for weakly reversible networks is directly not applicable. Instead, for each component C_i we will modify the entropy method by introducing an *artificial equilibrium state with normalised mass*, which balances the reaction within C_i. The artificial equilibrium will allow us to consider a quadratic functional, which is similar to the relative entropy in weakly reversible networks and which can be proved to decay exponentially to zero. This result is stated in the following Theorem:

Theorem 1.4 (Exponential decay to zero of source and transmission components).
Given an arbitrary first order reaction network partitioned into linkage classes and consider (w.l.o.g.) any corresponding connected subnetwork $\mathcal{N}_\mathcal{L}$. Assume for $\mathcal{N}_\mathcal{L}$ that all diffusion coefficients d_i are positive.

Then, for each C_i being a source or a transmission component of $\mathcal{N}_\mathcal{L}$, there exist constants $K_i > 0$ and $\lambda_i > 0$ depending explicitly on A_i and Ω such that, for any specie $S_\ell \in C_i$, the concentration u_ℓ of S_ℓ decays exponentially to zero, i.e.
$$\|u_\ell(t,\cdot)\|^2_{L^2(\Omega)} \le K_i e^{-\lambda_i t}, \qquad \text{for all } \ t > 0.$$

For a target component C_i, due to the in-flow $\mathcal{F}_i^{in}$, the total mass of C_i is not conserved but increasing. Hence, C_i does not possesses an equilibrium as weakly reversible networks, which is explicitly given in terms of the reaction rates and the conserved initial total mass.

However, since each target component is strongly connected and thus a weakly reversible reaction network with mass influx, there still exists a unique, positive equilibrium of C_i denoted by $X_{i,\infty}$, which balances the reactions within C_i and has a total mass, which is the sum of the total initial mass of C_i and the total "injected mass" from the other components via the in-flow $\mathcal{F}_i^{in}$ (see Lemma 3.3). We emphasis that in general the injected mass is not given explicitly but depends on the time evolution of all the influencing species higher up with respect to the topological order of the graph G^C.

Since the equilibrium $X_{i,\infty}$ is positive, we can use again a relative entropy functional to prove the convergence of the species belonging to a target component to their corresponding equilibrium states.

Theorem 1.5 (Exponential convergence for target components).
Given an arbitrary first order reaction network partitioned into linkage classes and consider (w.l.o.g.) any corresponding connected subnetwork $\mathcal{N}_\mathcal{L}$. Assume for $\mathcal{N}_\mathcal{L}$ that all diffusion coefficients d_i are positive.

Then, for all target components $C_i = \{S_{i_1}, S_{i_2}, \ldots, S_{i_{N_i}}\}$ of $\mathcal{N}_\mathcal{L}$, where N_i is the number of species belonging to C_i, there exists a unique positive equilibrium state $X_{i,\infty} = (u_{i_1,\infty}, \ldots, u_{i_{N_i},\infty})$ and the concentrations u_{i_ℓ} of S_{i_ℓ} converges exponentially to the corresponding equilibrium value

$$\|u_{i_\ell}(t) - u_{i_\ell,\infty}\|^2_{L^2(\Omega)} \le K_i e^{-\lambda_i t}, \qquad \text{for all } t > 0,$$

with the constants $K_i > 0$ and $\lambda_i > 0$ depending explicitly on A_i, Ω and D_i and on the equilibrium state $X_{i,\infty}$.

Remark 1.9. *Note that by Lemma 3.3, the equilibrium state $X_{i,\infty}$ depends explicitly on the mass injected into the target component C_i, but that injected mass itself depends non-explicitly on the initial data and on the history of the reaction-diffusion network.*

Remark 1.10. *We remark that in the same way as Theorem 1.3 generalises Theorem 1.2 to allow for degenerate diffusion matrices, it is equally possible to generalise Theorems 1.4 and 1.5 in the sense that it is sufficient to assume that for each target component there is at least one diffusion coefficient is positive. In particular, the proof of Theorem 1.4 holds independently from the entries of a non-negative diffusion matrices D_i.*

Outline: The rest of the paper is organised as follows: In Section 2, we present the entropy method for weakly reversible networks and prove exponential convergence to the positive equilibrium. Non weakly reversible networks will be investigated in the Section 3. By using the structure of the underlying graphs, we are able completely resolve the large-time behaviour of all species belonging to such first order networks.

We also remark that all constants in this manuscript are explicit in the sense that they are derived in constructive ways. However, since these constants are not optimal, we will denote them by using generic letters like K_i or λ_i, etc. The issue of optimal rates and constants for the convergence is subtle, and can be investigated in future works.

Notation: We shall use the shortcut $\overline{f} = \int_\Omega f(x)\,dx$, whenever $|\Omega| = 1$, and $\|\cdot\|$ for the usual norm in $L^2(\Omega)$, i.e.

$$\|f\|^2 = \int_\Omega |f(x)|^2 dx.$$

2. Strongly connected first order networks

In this section, we consider strongly connected first order reaction networks $\mathcal{N}$, for which the associated directed graph is strongly connected. This is w.l.o.g. by Remarks 1.3 and 1.4, since any weakly reversible first order reaction network can be partitioned into disjoint strongly connected components/subnetworks, which can be treated independently.

Moreover, we recall that $\Omega \subset \mathbb{R}^n$ is a bounded domain with smooth boundary $\partial\Omega$ (say $\partial\Omega \in C^{2+\alpha}$) and normalised volume $|\Omega| = 1$ (w.l.o.g. by rescaling). Finally, we recall the system (1.1)

$$\begin{cases} \partial_t X - D\Delta X = AX, & x \in \Omega, \quad t > 0, \\ \partial_\nu X = 0, & x \in \partial\Omega, \quad t > 0, \\ X(x,0) = X_0(x), & x \in \Omega, \end{cases} \tag{2.1}$$

where $X = [u_1, u_2, \ldots, u_N]^T$ denotes the vector of concentrations, the vector $X_0 = [u_{1,0}, u_{2,0}, \ldots, u_{N,0}]^T$ denotes the initial data, the diffusion matrix $D = \text{diag}(d_1, d_2, \ldots, d_N)$ and the reaction matrix $A = (a_{ij}) \in \mathbb{R}^{N\times N}$ satisfies

$$\begin{cases} a_{ij} \ge 0, & \text{for all } i \ne j, \quad i,j = 1,2,\ldots,N, \\ a_{jj} = -\sum_{i=1, i\ne j}^N a_{ij}, & \text{for all } j = 1,2,\ldots,N. \end{cases} \tag{2.2}$$

Moreover, since $\mathcal{N}_\mathcal{L}$ is strongly connected, we know that the reaction matrix A is irreducible, see Lemma 1.1. For the linear system (2.1), the existence of a global unique solution follows by standard arguments, see e.g. [Smo, Rot]:

Theorem 2.1 (Global well-posedness of linear reaction-diffusion networks).
For all given initial data $X_0 \in (L^2(\Omega))^N$, there exists a unique solution $X \in C([0,T]; (L^2(\Omega))^N) \cap$

$L^2(0,T;(H^1(\Omega))^N)$ for all $T>0$. Moreover, if $X_0 \geq 0$ then $X(t) \geq 0$ for all $t>0$. Finally, the solutions to (2.1) conserve the total mass (1.3) for all $t>0$:

$$\sum_{i=1}^N \int_\Omega u_i(x,t)dx = \sum_{i=1}^N \int_\Omega u_{i,0}(x)dx =: M > 0, \tag{2.3}$$

where the initial mass M is assumed positive.

Lemma 1.1 stated the equivalence to weak reversibility first order reaction networks and irreducibility of the reaction matrices A, which follows e.g. from [Sen81, Definition 2.1, page 46] and [Min88, Theorem 3.2, page 78]. Moreover, Lemma 1.1 stated the existence of a unique positive complex balance equilibrium to (2.1) for any given positive initial mass $M>0$. Concerning the proof of this part of Lemma 1.1, it remains to show the following

Lemma 2.2 (Unique positive equilibria for strongly connected networks with fixed mass M)**.** *The first order reaction network $\mathcal{N}$ is strongly connected if and only if the system (2.1) admits a unique positive equilibrium for any fixed positive mass $M>0$.*

Proof. Sufficiency: Assume that $\mathcal{N}$ is strongly connected. Thanks to the first equivalency in Lemma 1.1, the reaction matrix A is irreducible. Moreover, for large enough $\alpha>0$, we have that $A+\alpha E$ is nonnegative in the sense that all of its elements are nonnegative. We can then apply an extension of the Perron-Frobenius theorem, see e.g. [Sen81, Theorem 2.6, page 46] or [Per07, Chapter 6.3.1], to obtain the existence of a unique positive equilibrium, i.e. a positive right zero-eigenvector $X_\infty = (u_{1,\infty}, u_{2,\infty}, \ldots, u_{N,\infty}) > 0$ satisfying $AX_\infty = 0$ such that $\sum_{i=1}^N u_{i,\infty} = M > 0$.

Necessity: Now assume that (2.1) has a unique positive equilibrium X_∞. Since $AX_\infty = 0$ and X_∞ is uniquely determined by the mass conservation, we obtain that $\dim(\ker A) = 1$.

By using a contradiction argument, we assume that $\mathcal{N}$ is not strongly connected, then the reaction matrix A is reducible, i.e.

$$A = P^T \begin{pmatrix} B & 0 \\ C & D \end{pmatrix} P$$

for some permutation matrix P, in which D is irreducible. Choose d to be an eigenvector of D corresponding to zero eigenvalue. Then

$$AP^T \begin{pmatrix} 0 \\ d \end{pmatrix} = P^T \begin{pmatrix} B & 0 \\ C & D \end{pmatrix} \begin{pmatrix} 0 \\ d \end{pmatrix} = P^T \begin{pmatrix} 0 \\ Dd \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$$

which means that $P^T \begin{pmatrix} 0 \\ d \end{pmatrix}$ is an eigenvector of A corresponding to zero eigenvalue. Since X_∞ is strictly positive, $P^T \begin{pmatrix} 0 \\ d \end{pmatrix}$ and X_∞ are linear independent, which leads to a contradiction with $\dim(\ker A) = 1$. □

In the following, we will use the entropy method to study the trend to equilibrium. More precisely, for two trajectories $X = (u_1, u_2, \ldots, u_N)$ and $Y = (v_1, v_2, \ldots, v_N)$ to (2.1), where $Y(t)$ has non-zero components for all times $t>0$, we consider the following quadratic relative entropy functional

$$\mathcal{E}(X|Y)(t) = \sum_{i=1}^N \int_\Omega \frac{|u_i|^2}{v_i} dx. \tag{2.4}$$

The following key Lemma 2.3 provides an explicit expression of the entropy dissipation associated to (2.4):

Lemma 2.3 (Relative entropy dissipation functional)**.** *Assume that $v_i(t) \neq 0$ for all $i = 1, 2, \ldots, N$ and $t>0$. Then, we have*

$$\mathcal{D}(X|Y) = -\frac{d}{dt}\mathcal{E}(X|Y) = 2\sum_{i=1}^N d_i \int_\Omega v_i \left|\nabla\left(\frac{u_i}{v_i}\right)\right|^2 dx + \sum_{i,j=1;i<j}^N \int_\Omega (a_{ij}v_j + a_{ji}v_i)\left(\frac{u_i}{v_i} - \frac{u_j}{v_j}\right)^2 dx.$$

Proof. For convenience we recall that

$$\partial_t u_i - d_i \Delta u_i = \sum_{j=1}^N a_{ij} u_j \qquad \text{and} \qquad \partial_t v_i - d_i \Delta v_i = \sum_{j=1}^N a_{ij} v_j,$$

for all $i = 1, \ldots, N$. Hence, we compute

$$\begin{aligned}
\frac{d}{dt}\mathcal{E}(X|Y) &= \sum_{i=1}^{N} \int_\Omega \left[2\frac{u_i}{v_i}\partial_t u_i - \frac{u_i^2}{v_i^2}\partial_t v_i \right] dx \\
&= \sum_{i=1}^{N} \int_\Omega \left[2\frac{u_i}{v_i}\left(d_i \Delta u_i + \sum_{j=1}^{N} a_{ij} u_j \right) - \frac{u_i^2}{v_i^2}\left(d_i \Delta v_i + \sum_{j=1}^{N} a_{ij} v_j \right) \right] dx \\
&= \sum_{i=1}^{N} \int_\Omega \left(2d_i \frac{u_i}{v_i}\Delta u_i - d_i \frac{u_i^2}{v_i^2}\Delta v_i \right) dx + \sum_{i=1}^{N} \int_\Omega \left(2\frac{u_i}{v_i}\sum_{j=1}^{N} a_{ij}u_j - \frac{u_i^2}{v_i^2}\sum_{j=1}^{N} a_{ij}v_j \right) dx \\
&=: \sum_{i=1}^{N} \int_\Omega J_D^{(i)} dx + \int_\Omega \sum_{i=1}^{N} J_R^{(i)} dx \\
&=: \mathcal{I}_D + \mathcal{I}_R.
\end{aligned} \tag{2.5}$$

Using integration by parts, we have

$$\begin{aligned}
\int_\Omega J_D^{(i)} dx &= \int_\Omega \left(2d_i \frac{u_i}{v_i}\Delta u_i - d_i \frac{u_i^2}{v_i^2}\Delta v_i \right) dx \\
&= -2d_i \int_\Omega \left(\nabla\left(\frac{u_i}{v_i}\right)\nabla u_i - \frac{u_i}{v_i}\nabla\left(\frac{u_i}{v_i}\right)\nabla v_i \right) dx \\
&= -2d_i \int_\Omega v_i \left| \nabla\left(\frac{u_i}{v_i}\right) \right|^2 dx.
\end{aligned} \tag{2.6}$$

Thus,

$$\mathcal{I}_D = -2\sum_{i=1}^{N} d_i \int_\Omega v_i \left| \nabla\left(\frac{u_i}{v_i}\right) \right|^2 dx. \tag{2.7}$$

For the reaction terms $\mathcal{I}_R$, we use $a_{ii} = -\sum_{j=1,j\neq i}^{N} a_{ji}$ to calculate

$$\begin{aligned}
J_R^{(i)} &= 2\frac{u_i}{v_i}\sum_{j=1}^{N} a_{ij}u_j - \frac{u_i^2}{v_i^2}\sum_{j=1}^{N} a_{ij}v_j \\
&= 2\frac{u_i}{v_i}\left(\sum_{j=1,j\neq i}^{N} a_{ij}u_j + a_{ii}u_i \right) - \frac{u_i^2}{v_i^2}\left(\sum_{j=1,j\neq i}^{N} a_{ij}v_j + a_{ii}v_i \right) \\
&= 2\frac{u_i}{v_i}\left(\sum_{j=1,j\neq i}^{N} a_{ij}u_j - u_i \sum_{j=1,j\neq i}^{N} a_{ji} \right) - \frac{u_i^2}{v_i^2}\left(\sum_{j=1,j\neq i}^{N} a_{ij}v_j - v_i \sum_{j=1,j\neq i}^{N} a_{ji} \right) \\
&= \sum_{j=1,j\neq i}^{N} \left(2\frac{u_i}{v_i}(a_{ij}u_j - a_{ji}u_i) - \frac{u_i^2}{v_i^2}(a_{ij}v_j - a_{ji}v_i) \right).
\end{aligned} \tag{2.8}$$

Therefore,

$$\mathcal{I}_R = \sum_{i=1}^N \int_\Omega J_R^{(i)} dx = \int_\Omega \sum_{i=1}^N \sum_{j=1, j\neq i}^N \left(2\frac{u_i}{v_i}(a_{ij}u_j - a_{ji}u_i) - \frac{u_i^2}{v_i^2}(a_{ij}v_j - a_{ji}v_i) \right) dx \tag{2.9}$$

$$= \sum_{i,j=1; i<j}^N \int_\Omega \Bigg[2\frac{u_i}{v_i}(a_{ij}u_j - a_{ji}u_i) - \frac{u_i^2}{v_i^2}(a_{ij}v_j - a_{ji}v_i) + 2\frac{u_j}{v_j}(a_{ji}u_i - a_{ij}u_j) - \frac{u_j^2}{v_j^2}(a_{ji}v_i - a_{ij}v_j) \Bigg] dx$$

$$= \sum_{i,j=1; i<j}^N \int_\Omega \left[2(a_{ij}u_j - a_{ji}u_i)\left(\frac{u_i}{v_i} - \frac{u_j}{v_j}\right) - (a_{ij}v_j - a_{ji}v_i)\left(\frac{u_i^2}{v_i^2} - \frac{u_j^2}{v_j^2}\right)\right] dx$$

$$= \sum_{i,j=1; i<j}^N \int_\Omega \left(\frac{u_i}{v_i} - \frac{u_j}{v_j}\right) \left[2(a_{ij}u_j - a_{ji}u_i) - (a_{ij}v_j - a_{ji}v_i)\left(\frac{u_i}{v_i} + \frac{u_j}{v_j}\right)\right] dx$$

$$= - \sum_{i,j=1; i<j}^N \int_\Omega (a_{ij}v_j + a_{ji}v_i) \left(\frac{u_i}{v_i} - \frac{u_j}{v_j}\right)^2 dx. \tag{2.10}$$

By combining (2.5), (2.7) and (2.10), we obtain the result stated in the Lemma. □

In order to simplify the following calculations, we introduce the difference to the equilibrium

$$W := (w_1, w_2, \dots, w_N) = (u_1 - u_{1,\infty}, u_2 - u_{2,\infty}, \dots, u_N - u_{N,\infty}) = X - X_\infty,$$

and remark that thanks to the linearity of the system, the difference W is the solution to (2.1) subject to the shifted initial data

$$W(x,0) = X(x,0) - X_\infty, \qquad \text{for all } x \in \Omega.$$

Note that the total initial mass corresponding to W is zero, i.e.

$$M_W := \sum_{i=1}^N \int_\Omega w_{i,0}\, dx = \sum_{i=1}^N \int_\Omega (u_{i,0}(x) - u_{i,\infty})\, dx = 0,$$

and that W conserves the zero mass

$$\sum_{i=1}^N \int_\Omega w_i(t,x)\, dx = 0, \qquad \text{for all } t > 0.$$

By using the relative entropy dissipation functional derived in Lemma 2.3, we have

$$\mathcal{D}(W|X_\infty) = 2\sum_{i=1}^N \int_\Omega d_i \frac{|\nabla w_i|^2}{u_{i,\infty}} dx + \sum_{i,j=1; i<j}^N (a_{ij}u_{j,\infty} + a_{ji}u_{i,\infty}) \int_\Omega \Big(\frac{w_i}{u_{i,\infty}} - \frac{w_j}{u_{j,\infty}}\Big)^2 dx.$$

The following Lemma about entropy-entropy dissipation estimate is the main key to prove the convergence to equilibrium for (2.1).

Lemma 2.4 (Entropy-Entropy Dissipation Estimate)**.**
There exists an explicit constant $\lambda > 0$ depending explicitly on the reaction matrix A, the domain Ω, the diffusion matrix D and the initial mass M such that

$$\mathcal{D}(W|X_\infty) \geq \lambda\, \mathcal{E}(W|X_\infty).$$

Proof. We divide the proof in several steps:
Step 1. (Additivity of the relative entropy w.r.t. spatial averages)
Straightforward calculation leads to

$$\mathcal{E}(W|X_\infty) = \sum_{i=1}^N \int_\Omega \frac{|w_i|^2}{u_{i,\infty}} dx = \sum_{i=1}^N \int_\Omega \frac{|w_i - \overline{w_i}|^2}{u_{i,\infty}} dx + \sum_{i=1}^N \frac{|\overline{w_i}|^2}{u_{i,\infty}}$$

$$= \mathcal{E}(W - \overline{W}|X_\infty) + \mathcal{E}(\overline{W}|X_\infty) \tag{2.11}$$

where we denote $\overline{W} = (\overline{w_1}, \overline{w_2}, \dots, \overline{w_N})$ and we recall that $\overline{w_i} = \int_\Omega w_i\, dx$ for $i = 1, \dots, N$ due to $|\Omega| = 1$.

Step 2. (Entropy dissipation due to diffusion)
By using Poincaré's inequality

$$\|\nabla f\|^2 \geq C_P\|f-\overline{f}\|^2, \qquad \text{for all } f\in H^1(\Omega), \tag{2.12}$$

we have

$$\begin{aligned}\frac{1}{2}\mathcal{D}(W|X_\infty) &\geq \sum_{i=1}^N d_i\int_\Omega \frac{|\nabla w_i|^2}{u_{i,\infty}}dx \geq C_P\sum_{i=1}^N d_i\int_\Omega\frac{|w_i-\overline{w_i}|^2}{u_{i,\infty}}dx\\ &\geq C_P\min\{d_1,d_2,\ldots,d_N\}\,\mathcal{E}(W-\overline{W}|X_\infty).\end{aligned}\tag{2.13}$$

Step 3. (Entropy dissipation due to reactions)
From (2.11) and (2.13), it remains to control

$$\mathcal{E}(\overline{W}|X_\infty) = \sum_{i=1}^N\frac{\overline{w_i}^2}{u_{i,\infty}}.$$

By using Jensen's inequality we have, recalling that $|\Omega|=1$,

$$\begin{aligned}\frac{1}{2}\mathcal{D}(W|X_\infty) &\geq \frac{1}{2}\sum_{i,j=1;i<j}^N(a_{ij}u_{j,\infty}+a_{ji}u_{i,\infty})\int_\Omega\left(\frac{w_i}{u_{i,\infty}}-\frac{w_j}{u_{j,\infty}}\right)^2dx\\ &\geq \frac{1}{2}\sum_{i,j=1;i<j}^N(a_{ij}u_{j,\infty}+a_{ji}u_{i,\infty})\left(\frac{\overline{w_i}}{u_{i,\infty}}-\frac{\overline{w_j}}{u_{j,\infty}}\right)^2dx.\end{aligned}\tag{2.14}$$

It then remains to prove that

$$\frac{1}{2}\sum_{i,j=1;i<j}^N(a_{ij}u_{j,\infty}+a_{ji}u_{i,\infty})\left(\frac{\overline{w_i}}{u_{i,\infty}}-\frac{\overline{w_j}}{u_{j,\infty}}\right)^2dx \geq \gamma\sum_{i=1}^N\frac{\overline{w_i}^2}{u_{i,\infty}}\tag{2.15}$$

for some $\gamma>0$. Note that if both reactions $S_i\to S_j$ and $S_j\to S_i$ do not appear in the reaction network, then we have $a_{ij}=a_{ji}=0$ and thus

$$a_{ij}u_{j,\infty}+a_{ji}u_{i,\infty}=0.$$

Hence, the expression

$$\sum_{i,j=1;i<j}^N(a_{ij}u_{j,\infty}+a_{ji}u_{i,\infty})\left(\frac{\overline{w_i}}{u_{i,\infty}}-\frac{\overline{w_j}}{u_{j,\infty}}\right)^2dx$$

may not contain all pairs (i,j) with $i\neq j$. However, the weak reversibility of the network allows to make all pairs (i,j) with $i\neq j$ appear in the following sense: There exists an explicit constant $\xi>0$ such that

$$\sum_{i,j=1;i<j}^N(a_{ij}u_{j,\infty}+a_{ji}u_{i,\infty})\left(\frac{\overline{w_i}}{u_{i,\infty}}-\frac{\overline{w_j}}{u_{j,\infty}}\right)^2dx \geq \xi\sum_{i,j=1;i<j}^N\left(\frac{\overline{w_i}}{u_{i,\infty}}-\frac{\overline{w_j}}{u_{j,\infty}}\right)^2.\tag{2.16}$$

Indeed, assume that $a_{ij}=a_{ji}=0$ for some $i\neq j$. Due to the weak reversibility of the network, there exists a path from S_i to S_j as follows

$$S_i\equiv S_{j_1}\xrightarrow{a_{j_2j_1}}S_{j_2}\xrightarrow{a_{j_3j_2}}\ldots\xrightarrow{a_{j_rj_{r-1}}}S_{j_r}\equiv S_j$$

with $r\geq 3$ and $a_{j_kj_{k-1}}>0$ for all $k=2,3,\ldots,r$. Thus, with

$$0<\sigma=\min_{(a_{ij},a_{ji})\neq(0,0);1\leq i<j\leq N}\{a_{ij}u_{i,\infty}+a_{ji}u_{j,\infty}\}\leq\min_{2\leq k\leq r}\{a_{j_kj_{k-1}}u_{j_{k-1},\infty}+a_{j_{k-1}j_k}u_{j_k,\infty}\}$$

we have

$$\begin{aligned}&\sum_{k=2}^r(a_{j_kj_{k-1}}u_{j_{k-1},\infty}+a_{j_{k-1}j_k}u_{j_k,\infty})\left(\frac{\overline{w_{j_k}}}{u_{j_k,\infty}}-\frac{\overline{w_{j_{k-1}}}}{u_{j_{k-1},\infty}}\right)^2\\ &\qquad\geq\sigma\sum_{k=2}^r\left(\frac{\overline{w_{j_k}}}{u_{j_k,\infty}}-\frac{\overline{w_{j_{k-1}}}}{u_{j_{k-1},\infty}}\right)^2\\ &\qquad\geq\frac{\sigma}{r-1}\left(\frac{\overline{w_{j_1}}}{u_{j_1,\infty}}-\frac{\overline{w_{j_r}}}{u_{j_r,\infty}}\right)^2=\frac{\sigma}{N-1}\left(\frac{\overline{w_i}}{u_{i,\infty}}-\frac{\overline{w_j}}{u_{j,\infty}}\right)^2.\end{aligned}\tag{2.17}$$

Since there are less than $N(N-1)/2$ pairs (i,j) with $a_{ij} = a_{ji} = 0$, we can repeat this procedure to finally get (2.16) with $\xi = 2\sigma/(N(N-1)^2)$. From (2.15) and (2.16), we are left to find a constant $\gamma > 0$ satisfying

$$\sum_{i,j=1;i<j}^{N} \left(\frac{\overline{w_i}}{u_{i,\infty}} - \frac{\overline{w_j}}{u_{j,\infty}}\right)^2 \geq \frac{2\gamma}{\xi}\sum_{i=1}^{N}\frac{\overline{w_i}^2}{u_{i,\infty}} \tag{2.18}$$

with the constraint of the conserved zero total mass

$$\sum_{i=1}^{N}\overline{w_i} = 0. \tag{2.19}$$

Because of (2.19),

$$\sum_{i=1}^{N}\overline{w_i}^2 = -\sum_{i,j=1;i\neq j}^{N}\overline{w_i}\,\overline{w_j} = -2\sum_{i,j=1;i<j}^{N}\overline{w_i}\,\overline{w_j}. \tag{2.20}$$

Therefore, we can estimate for $C = \min_{1\leq i<j\leq N}\frac{1}{u_{i,\infty}u_{j,\infty}}$

$$\begin{aligned}\sum_{i,j=1;i<j}^{N}\left(\frac{\overline{w_i}}{u_{i,\infty}} - \frac{\overline{w_j}}{u_{j,\infty}}\right)^2 &\geq \min_{i<j}\frac{1}{u_{i,\infty}u_{j,\infty}}\sum_{i<j}u_{i,\infty}u_{j,\infty}\left(\frac{\overline{w_i}}{u_{i,\infty}} - \frac{\overline{w_j}}{u_{j,\infty}}\right)^2\\ &\geq -2\min_{i<j}\frac{1}{u_{i,\infty}u_{j,\infty}}\sum_{i<j}\overline{w_i}\,\overline{w_j} = \min_{i<j}\frac{1}{u_{i,\infty}u_{j,\infty}}\sum_{i=1}^{N}\overline{w_i}^2 \geq \min_{i<j}\frac{1}{u_{i,\infty}u_{j,\infty}}\sum_{i=1}^{N}\frac{\overline{w_i}^2}{u_{i,\infty}}.\end{aligned} \tag{2.21}$$

In conclusion, we have proved (2.18) with $\gamma = \frac{\xi}{2}\min_{i<j}\frac{1}{u_{i,\infty}u_{j,\infty}}$, which in combination with (2.16) implies (2.15) and thus completes the proof of this Lemma. □

Theorem 2.5 (Convergence to Equilibrium)**.**
Consider (w.l.o.g) a strongly connected subnetwork $\mathcal{N}$ of a weakly reversible first order reaction network. Assume for $\mathcal{N}$ that the diffusion coefficients d_i are positive for all $i = 1, 2, \ldots, N$, and the initial mass M is positive.

Then, the unique global solution to (2.1) converges to the unique positive equilibrium X_∞ in the following sense:

$$\sum_{i=1}^{N}\int_\Omega \frac{|u_i(t) - u_{i,\infty}|^2}{u_{i,\infty}}dx \leq e^{-\lambda t}\sum_{i=1}^{N}\int_\Omega\frac{|u_{i,0} - u_{i,\infty}|^2}{u_{i,\infty}}dx,$$

where the constant $\lambda > 0$ is computed as in Lemma 2.4.

Proof. From Lemma 2.4 we have

$$\frac{d}{dt}\mathcal{E}(X - X_\infty|X_\infty) = -\mathcal{D}(X - X_\infty|X_\infty) \leq \lambda\,\mathcal{E}(X - X_\infty|X_\infty).$$

By Gronwall's inequality,

$$\mathcal{E}(X(t) - X_\infty|X_\infty) \leq e^{-\lambda t}\mathcal{E}(X_0 - X_\infty|X_\infty),$$

and the proof is complete. □

Proof of Theorem 1.2. Theorem 1.2 is a direct consequence of Theorem 2.5 and the partition of weakly reversible first order reaction network into strongly connected components. □

We now turn to the case of degenerate diffusion, where some of the diffusion coefficients d_i can be zero. In the proof of the Theorem 2.5, we have used non-degenerate diffusion of all species in order to control distance of the concentrations to their spatial averages (see estimate (2.13)). This procedure must thus be adapted in the case of degenerate diffusion.

It was already proven in [DF07, FLT, MHM] that even if some diffusion coefficients vanish, one can still show exponential convergence to equilibrium provided reversible reactions. The technique used in these mentioned references is based on the fact that diffusion of one specie, which is connected through a reversible reaction with another specie, induces a indirect kind of "diffusion effect" to the latter specie.

We will prove that this principle is still valid for weakly reversible reaction networks as considered in this section.

Theorem 2.6 (Convergence to Equilibrium with Degenerate Diffusion)**.**
Consider (w.l.o.g) a strongly connected subnetwork $\mathcal{N}$ of a weakly reversible first order reaction network. Assume for $\mathcal{N}$ that the initial mass M is positive. Moreover, assume that at least one diffusion coefficient d_i is positive for some $i = 1, 2, \ldots, N$.

Then, the solution to (2.1) converges exponentially to equilibrium via the following estimate

$$\sum_{i=1}^{N}\int_{\Omega}\frac{|u_i(t)-u_{i,\infty}|^2}{u_{i,\infty}}dx \le e^{-\lambda' t}\sum_{i=1}^{N}\int_{\Omega}\frac{|u_{i,0}-u_{i,\infty}|^2}{u_{i,\infty}}\,dx,$$

for some explicit rate $\lambda' > 0$ which depends explicitly on A, Ω, D and M..

Proof. We aim for a similar entropy-entropy dissipation inequality as stated in Lemma (2.4), i.e. we want to find a constant $\lambda' > 0$ such that

$$\mathcal{D}(W|X_\infty) \ge \lambda'\,\mathcal{E}(W|X_\infty) = \lambda'\,[\mathcal{E}(W-\overline{W}|X_\infty) + \mathcal{E}(\overline{W}|X_\infty)]. \tag{2.22}$$

Due to the degenerate diffusion, the diffusion part of $\mathcal{D}(W|X_\infty)$ is insuffcient to control $\mathcal{E}(W-\overline{W}|X_\infty)$ as in (2.13), since some of diffusion coefficients can be zero. This difficulty can be resolved by quantifying the fact that diffusion of one specie is transferred to another species when connected via a weakly reversible reaction path. Without loss of generality, we assume that $d_1 > 0$ and estimate $\mathcal{D}(W|X_\infty)$ by

$$\mathcal{D}(W|X_\infty) \ge d_1\int_{\Omega}\frac{|\nabla w_1|^2}{u_{1,\infty}}\,dx + \sum_{i,j=1;i<j}^{N}(a_{ij}u_{j,\infty} + a_{ji}u_{i,\infty})\int_{\Omega}\left(\frac{w_i}{u_{i,\infty}} - \frac{w_j}{u_{j,\infty}}\right)^2 dx. \tag{2.23}$$

By arguments similar to (2.16) and (2.17), we have

$$\mathcal{D}(W|X_\infty) \ge d_1\int_{\Omega}\frac{|\nabla w_1|^2}{u_{1,\infty}}dx + \xi\sum_{i,j=1;i<j}^{N}\int_{\Omega}\left(\frac{w_i}{u_{i,\infty}} - \frac{w_j}{u_{j,\infty}}\right)^2 dx. \tag{2.24}$$

To control $\mathcal{E}(W-\overline{W}|X_\infty)$, we use the following estimate for all $i = 2, 3, \ldots, N$:

$$\int_{\Omega}\frac{|\nabla w_1|^2}{u_{1,\infty}}\,dx + \int_{\Omega}\left(\frac{w_1}{u_{1,\infty}} - \frac{w_i}{u_{i,\infty}}\right)^2 dx \ge \beta\int_{\Omega}\frac{|w_i-\overline{w_i}|^2}{u_{i,\infty}}\,dx, \tag{2.25}$$

with $\beta = \frac{1}{2u_{1,\infty}}\min\left\{\frac{C_P}{u_{1,\infty}}, 1\right\}$: Indeed, thanks to Poincaré's inequality $\|\nabla f\|^2 \ge C_P\|f-\overline{f}\|^2$, we estimate for various sufficiently small constants C

$$\begin{aligned}
\int_{\Omega}\frac{|\nabla w_1|^2}{u_{1,\infty}}\,dx + \int_{\Omega}\left(\frac{w_1}{u_{1,\infty}} - \frac{w_i}{u_{i,\infty}}\right)^2 dx &\ge \int_{\Omega}\left[C_P\frac{|w_1-\overline{w_1}|^2}{u_{1,\infty}} + \left(\frac{w_1-\overline{w_1}}{u_{1,\infty}} + \frac{\overline{w_1}}{u_{1,\infty}} - \frac{w_i}{u_{i,\infty}}\right)^2\right] dx\\
&\ge \frac{1}{2}\min\left\{\frac{C_P}{u_{1,\infty}}, 1\right\}\int_{\Omega}\left(\frac{\overline{w_1}}{u_{1,\infty}} - \frac{w_i}{u_{i,\infty}}\right)^2 dx\\
&= \frac{1}{2}\min\left\{\frac{C_P}{u_{1,\infty}}, 1\right\}\int_{\Omega}\left(\frac{\overline{w_1}}{u_{1,\infty}} - \frac{\overline{w_i}}{u_{i,\infty}} + \frac{\overline{w_i}}{u_{i,\infty}} - \frac{w_i}{u_{i,\infty}}\right)^2 dx\\
&= \frac{1}{2}\min\left\{\frac{C_P}{u_{1,\infty}}, 1\right\}\int_{\Omega}\left(\frac{\overline{w_1}}{u_{1,\infty}} - \frac{\overline{w_i}}{u_{i,\infty}}\right)^2 dx + \frac{1}{2}\min\left\{\frac{C_P}{u_{1,\infty}}, 1\right\}\int_{\Omega}\left(\frac{\overline{w_i}}{u_{i,\infty}} - \frac{w_i}{u_{i,\infty}}\right)^2 dx\\
&\ge \frac{1}{2u_{1,\infty}}\min\left\{\frac{C_P}{u_{1,\infty}}, 1\right\}\int_{\Omega}\frac{|w_i-\overline{w_i}|^2}{u_{i,\infty}}\,dx.
\end{aligned} \tag{2.26}$$

Now, thanks to (2.24) and (2.25)

$$\begin{aligned}
\mathcal{D}(W|X_\infty) &\ge \min\left\{\frac{d_1}{N}, \frac{\xi}{2}\right\}\beta\sum_{i=1}^{N}\int_{\Omega}\frac{|w_i-\overline{w_i}|^2}{u_{i,\infty}}dx + \frac{\xi}{2}\sum_{i,j=1;i<j}^{N}\int_{\Omega}\left(\frac{w_i}{u_{i,\infty}} - \frac{w_j}{u_{j,\infty}}\right)^2 dx\\
&\ge \min\left\{\frac{d_1}{N}, \frac{\xi}{2}\right\}\beta\,\mathcal{E}(W-\overline{W}|X_\infty) + \frac{\xi}{2}\sum_{i,j=1;i<j}^{N}\int_{\Omega}\left(\frac{\overline{w_i}}{u_{i,\infty}} - \frac{\overline{w_j}}{u_{j,\infty}}\right)^2 dx\\
&\ge \min\left\{\frac{d_1}{N}, \frac{\xi}{2}\right\}\beta\mathcal{E}(W-\overline{W}|X_\infty) + \frac{\gamma}{4}\mathcal{E}(\overline{W}|X_\infty) \qquad \text{(by using (2.18))}\\
&\ge \lambda'\,\mathcal{E}(W|X_\infty)
\end{aligned} \tag{2.27}$$

with $\lambda' = \min\left\{\frac{\beta d_1}{N}, \frac{\xi\beta}{2}, \frac{\gamma}{4}\right\}$. Thus (2.22) is proved and the proof is complete. □

Proof of Theorem 1.3. Theorem 1.3 is a direct consequence of Theorem 2.6 and the partition of weakly reversible first order reaction network into strongly connected components. □

Remark 2.1. *The estimate (2.25) is usually interpreted as follows: the sum of the dissipation due to the diffusion of w_1 and the dissipation caused by the reaction between w_1 and w_i are bounded below by (2.26), which is essentially a diffusion dissipation term of the specie w_i (after having applied Poincaré's inequality). In this sense, a "diffusion effect" has been transferred onto w_i.*

We remark that while the presented proof for the linear case is straightforward, the proof of an analogous estimate to (2.25) in nonlinear cases turns out to be quite tricky. Readers are referred to [DF07] or [FLT, Lemma 3.6] for more details.

3. Non-weakly reversible networks

In this section, we consider (w.l.o.g.) reaction networks $\mathcal{N}$ which are not weakly reversible, yet form one linkage class. Thus, the corresponding directed graph G is connected yet not strongly connected (i.e. the underlying undirected graph of G is connected). We will show that in the large time behaviour, each specie tends exponentially fast either to zero or to a positive equilibrium value depending on its position in the graph representing the network.

For weakly reversible reaction-diffusion networks (corresponding to strongly connected graphs), it was proven in Section 2 that each specie converges exponentially fast to a unique, positive equilibrium value, which is given explicitly in terms of the reaction rates and the conserved initial total mass.

For non weakly reversible reaction networks, however, we will show that while the equilibria are still unique and attained exponentially fast, the equilibrium values are in general no longer explicitly given but depend on the position in the graph in general and on the history of the concentrations of the influencing species in particular.

Moreover, since non weakly reversible reaction networks (2.1) may no longer have positive equilibria, the relative entropy method used in Section 2 is not directly applicable. Nevertheless, we will see that the relative entropy and the ideas of the entropy method still play the essential role our analysis of non-weakly reversible networks.

As the large time behaviour of the species depend on their position within the network, we need to first state some important properties of the graph G. The following Lemmas 3.1 and 3.2 are well known in graph theory. We refer the reader to the book [BJG08] for a reference.

Lemma 3.1 (Strongly connected components form acyclic graphs G^C).
Let G be a directed graph which is connected, that is the underlying undirected graph of G is connected, but not strongly connected such that the graph G contains at least $r \geq 2$ strongly connect components, which we shall denote by $C_1, C_2, \ldots, C_r$. Thus, we can define a directed graph G^C of strongly connected components as follows

- *G^C has as nodes the r strongly connected components $C_1, C_2, \ldots, C_r$,*
- *for two nodes C_i and C_j of G^C, if there exists a reaction $C_i \ni S_k \xrightarrow{a_{\ell k}} S_\ell \in C_j$ with $a_{\ell k} > 0$, then we define a directed edge $C_i \to C_j$ of G^C.*

Then, the directed graph G^C is acyclic, that is G^C does not contain any cycles.

Proof. The proof can be found in e.g. [BJG08, Chapter 1] and shows that if G^C would contain a cycle then this cycle should have been contained in a strongly connected component in the first place. □

Lemma 3.2 (Topological order of acyclic graphs, [BJG08, Chapter 1]).
There exists a reordering of the nodes of G^C in such a way that for all direct edges $C_i \to C_j$ we always have $i < j$.

From now on, we will always consider topologically ordered graphs G^C. For each $i = 1, 2, \ldots, N$, we denote by N_i the number of species belonging to C_i. For notational convenience later on, we shall set $L[0] = 0$ and introduce the cumulative number $L[i]$ of the species contained in all strongly connected components up to C_i, i.e.

$$L[i] = N_1 + N_2 + \ldots + N_i \qquad \text{for all } i = 1, 2, \ldots, r. \tag{3.1}$$

We then reorder the species of the network $\mathcal{N}$ in such a order that the species belong to the component C_i are $S_{L[i-1]+1}, S_{L[i-1]+2}, \ldots, S_{L[i]}$ for all $i = 1, 2, \ldots, N$.

Each component C_i belongs to one of the following three types:

- *Source component*: C_i is a source component if there is no in-flow to C_i, i.e. there does not exist an edge $C_i \not\ni S_k \to S_j \in C_i$,
- *Target component*: C_i is a target component if there is no out-flow from C_i, i.e. there does not exist an edge $C_i \ni S_k \to S_j \notin C_i$,
- *Transmission component*: If C_i is neither a source component nor a target component, then C_i is called a transmission component.

The above classification of strongly connected components greatly improves the notation of the corresponding dynamics, which quantifies the behaviour of the species belonging to the three types of components. In the following, we denote by $X_i = (u_{L[i-1]+1}, u_{L[i-1]+2}, \ldots, u_{L[i]})^T$ the concentration vector of the species belonging to C_i.

The evolution of the species belonging to a component C_i depends on the type of C_i:

(i) For a source component C_i, the system for X_i is of the form

$$\begin{cases} \partial_t X_i - D_i \Delta X_i = A_i X_i - F_i^{out} X_i, & x \in \Omega, \quad t > 0, \\ \partial_\nu X_i = 0, & x \in \partial\Omega, \quad t > 0, \\ X_i(x,0) = X_{i,0}(x), & x \in \Omega, \end{cases} \tag{3.2}$$

where the diffusion matrix D_i is

$$D_i = \mathrm{diag}(d_{L[i-1]+1}, d_{L[i-1]+2}, \ldots, d_{L[i]}) \in \mathbb{R}^{N_i \times N_i}, \tag{3.3}$$

the reaction matrix A_i is

$$A_i = (a_{L[i-1]+k, L[i-1]+\ell})_{1 \le k, \ell \le N_i} \in \mathbb{R}^{N_i \times N_i}, \tag{3.4}$$

and the out flow matrix is defined as

$$F_i^{out} = \mathrm{diag}(f_{L[i-1]+1}, f_{L[i-1]+2}, \ldots, f_{L[i]}) \in \mathbb{R}^{N_i \times N_i} \tag{3.5}$$

with

$$f_{L[i-1]+k} = \sum_{\ell = L[i]+1}^{N} a_{\ell, L[i-1]+k} \qquad \forall k = 1, 2, \ldots, N_i,$$

where the lower summation index $L[i]+1$ follows for the topological order of the graph G^C.

Roughly speaking, $f_{L[i-1]+k}$ is the sum of all the reaction rates from the specie $S_{L[i-1]+k}$ to species outside of C_i. It may happen that $f_{L[i-1]+k} = 0$ for some $k = 1, 2, \ldots, N$, but there exists at least one k_0 such that $f_{L[i-1]+k_0} > 0$ since C_i is a source component.

(ii) If C_i is a transmission component, the system for X_i writes as

$$\begin{cases} \partial_t X_i - D_i \Delta X_i = \mathcal{F}_i^{in} + A_i X_i - F_i^{out} X_i, & x \in \Omega, \quad t > 0, \\ \partial_\nu X_i = 0, & x \in \partial\Omega, \quad t > 0, \\ X_i(x,0) = X_{i,0}(x), & x \in \Omega, \end{cases} \tag{3.6}$$

where the diffusion matrix D_i, the reaction matrix A_i and the out flow matrixF_i^{out} are defined as above in (3.3), (3.4) and (3.5), respectively. The in-flow vector $\mathcal{F}_i^{in}$ is defined by

$$\mathcal{F}_i^{in} = \begin{pmatrix} z_{L[i-1]+1} \\ z_{L[i-1]+2} \\ \cdots \\ z_{L[i]} \end{pmatrix} \quad \text{with} \quad z_{L[i-1]+\ell} = \sum_{k=1}^{L[i-1]} a_{L[i-1]+\ell, k} u_k. \tag{3.7}$$

We remark that by studying all components C_i within the topological order of G^C, the dynamics of the previous components $C_1, C_2, \ldots, C_{i-1}$ is already known at the time we analyse the component C_i. Thus, in system (3.6) the in-flow vector $\mathcal{F}_i^{in}$ can be considered as a given external in-flow.

(iii) If C_i is a target component, we can write

$$\begin{cases} \partial_t X_i - D_i \Delta X_i = \mathcal{F}_i^{in} + A_i X_i, & x \in \Omega, \quad t > 0, \\ \partial_\nu X_i = 0, & x \in \partial\Omega, \quad t > 0, \\ X_i(x,0) = X_{i,0}(x), & x \in \Omega, \end{cases} \tag{3.8}$$

where the reaction matrix A_i and the in-flow $\mathcal{F}_i^{in}$ are defined in the same way as above in (3.4) and (3.7).

By modifying the relative entropy method in Section 2, we obtain the

Proof of Theorem 1.4. Since the ongoing outflow vanishes the mass of all source components and subsequently all transmission components, the corresponding equilibrium values are expected to be zero and the relative entropy method used for weakly reversible networks is not directly applicable here. We instead introduce a concept of "artificial equilibrium states with normalised mass" for these components, which allows to derive a quadratic entropy-like functional, which can be proved to decay exponentially. Due to their different dynamics, we have to distinguish the two cases: C_i is a source component and C_i is a transmission component.

The aim of the proof is to show that if C_i is a source or a transmission component then for all $k = 1, \ldots, N_i$,

$$\|u_{L[i-1]+k}(t)\|^2_{L^2(\Omega)} \le K_i e^{-\lambda_i t}, \qquad \text{for all } t \ge 0, \tag{3.9}$$

for explicit constants $K_i > 0$ and $\lambda_i > 0$.

In order to simplify the notation, we shall denote

$$v_k = u_{L[i-1]+k}, \quad \text{and} \quad b_{k,\ell} = a_{L[i-1]+k, L[i-1]+\ell}, \qquad \text{for all } 1 \le k, \ell \le N_i. \tag{3.10}$$

Then, the concentration vector X_i and the reaction matrix A_i can be rewritten as

$$X_i = (v_1, v_2, \ldots, v_{N_i}) \qquad \text{and} \qquad A_i = (b_{k,\ell})_{1 \le k,\ell \le N_i}.$$

Note that the index i for the component C_i is fixed.

Case 1: C_i is a source component.

We recall the corresponding system from (3.2)

$$\begin{cases} \partial_t X_i - D_i \Delta X_i = A_i X_i - F_i^{out} X_i, & x \in \Omega, \quad t > 0, \\ \partial_\nu X_i = 0, & x \in \partial\Omega, \quad t > 0, \\ X_i(x,0) = X_{i,0}(x), & x \in \Omega. \end{cases} \tag{3.11}$$

We now introduce an artificial equilibrium state $X_{i,\infty} = (v_{1,\infty}, v_{2,\infty} \ldots, v_{N_i,\infty})^T$ with normalised mass to (3.11), which is defined as the solution of the system

$$\begin{cases} A_i X_{i,\infty} = 0, \\ v_{1,\infty} + v_{2,\infty} + \ldots + v_{N_i,\infty} = 1. \end{cases} \tag{3.12}$$

It follows from Lemma 2.2 that there exists a unique positive solution $X_{i,\infty}$ to (3.12). Here we notice that $X_{i,\infty}$ balances all reactions within C_i while the total mass contained in $X_{i,\infty}$ is normalised to one.

In the following we will study the evolution of the quadratic entropy-like functional

$$\mathcal{E}(X_i | X_{i,\infty}) = \sum_{k=1}^{N_i} \int_\Omega \frac{|v_k|^2}{v_{k,\infty}} dx. \tag{3.13}$$

By similar calculations as in Lemma 2.3, we obtain the time derivative of this quadratic functional

$$\begin{aligned} \mathcal{D}(X_i | X_{i,\infty}) &= -\frac{d}{dt}\mathcal{E}(X_i|X_{i,\infty}) \\ &= 2\sum_{k=1}^{N_i} d_{L[i-1]+k} \int_\Omega \frac{|\nabla v_k|^2}{v_{k,\infty}} dx \\ &\quad + \sum_{k,\ell=1; k<\ell}^{N_i} (b_{k,\ell} v_{\ell,\infty} + b_{\ell,k} v_{k,\infty}) \int_\Omega \left(\frac{v_k}{v_{k,\infty}} - \frac{v_\ell}{v_{\ell,\infty}} \right)^2 dx \\ &\quad + 2\sum_{k=1}^{N_i} f_{L[i-1]+k} \int_\Omega \frac{|v_k|^2}{v_{k,\infty}} dx. \end{aligned} \tag{3.14}$$

We remark that since C_i is a source component, there exists an index $k_0 \in \{1, 2, \ldots, N_i\}$ such that the out-flow $f_{L[i-1]+k_0} > 0$ is positive. Then, an estimate similar to (2.16) gives for various constants C

$$\begin{aligned}
\mathcal{D}(X_i|X_{i,\infty}) &\geq \xi \sum_{k,\ell=1;k<\ell}^{N_i} \int_\Omega \Big(\frac{v_k}{v_{k,\infty}} - \frac{v_\ell}{v_{\ell,\infty}}\Big)^2 dx + 2 f_{L[i-1]+k_0} \int_\Omega \frac{|v_{k_0}|^2}{v_{k_0,\infty}} dx \\
&\geq \min\{\xi/2, f_{L[i-1]+k_0}/2N_i\} \sum_{\ell=1;\ell\neq k_0}^{N_i} \int_\Omega \Big[\Big(\frac{v_\ell}{v_{\ell,\infty}} - \frac{v_{k_0}}{v_{k_0,\infty}}\Big)^2 + \frac{|v_{k_0}|^2}{v_{k_0,\infty}}\Big] dx \\
&\quad + f_{L[i-1]+k_0} \int_\Omega \frac{|v_{k_0}|^2}{v_{k_0,\infty}} dx \\
&\geq \lambda_i \sum_{\ell=1}^{N_i} \int_\Omega \frac{|v_\ell|^2}{v_{\ell,\infty}} dx = \lambda_i\, \mathcal{E}(X_i|X_{i,\infty})
\end{aligned} \tag{3.15}$$

with $\lambda_i = \min\{\xi/4, f_{L[i-1]+k_0}/4N_i\}$. It follows that

$$\frac{d}{dt}\mathcal{E}(X_i|X_{i,\infty}) = -\mathcal{D}(X_i|X_{i,\infty}) \leq -\lambda_i\, \mathcal{E}(X_i|X_{i,\infty}),$$

and thus

$$\sum_{k=1}^{N_i} \int_\Omega \frac{|v_k(t)|^2}{v_{k,\infty}} dx = \mathcal{E}(X_i(t)|X_{i,\infty}) \leq e^{-\lambda_i t}\mathcal{E}(X_{i,0}|X_{i,\infty}),$$

or equivalently

$$\|u_{L[i-1]+k}(t)\|^2 \leq e^{-\lambda_i t}\mathcal{E}(X_{i,0}|X_{i,\infty}) \max_{1\leq i\leq N_i}\{v_{i,\infty}\} \qquad \text{for all } t > 0, \quad \text{for all } k = 1, 2, \ldots, N_i,$$

which proves (3.9) with $K_i = \mathcal{E}(X_{i,0}|X_{i,\infty}) \max_{1\leq i\leq N_i}\{v_{i,\infty}\}$ in the case C_i is a source component.

Case 2: C_i is a transmission component.

By recalling that the components C_i are topologically ordered, we can assume without loss of generality that u_ℓ, with $\ell = 1, 2, \ldots, L[i-1]$, obeys the following exponential decay

$$\|u_\ell(t)\|^2 \leq K^* e^{-\lambda^* t}, \qquad \ell = 1, 2, \ldots, L[i-1], \qquad \text{for all } t > 0. \tag{3.16}$$

for $0 < \lambda^* = \min\limits_{1\leq k\leq i-1} \lambda_k$ and $K^* = \max\limits_{1\leq k\leq i-1} K_i$. We also recall the system for C_i,

$$\begin{cases}
\partial_t X_i - D_i\Delta X_i = \mathcal{F}_i^{in} + A_i X_i - F_i^{out} X_i, & x \in \Omega, \quad t > 0,\\
\partial_\nu X_i = 0, & x \in \partial\Omega, \quad t > 0,\\
X_i(x,0) = X_{i,0}(x), & x \in \Omega,
\end{cases} \tag{3.17}$$

with $\mathcal{F}_i^{in}$ is defined as (3.7). Denote by $X_{i,\infty} = (v_{1,\infty}, \ldots, v_{N_i,\infty})^T$ the artificial equilibrium state of (3.17), which is the unique positive solution to

$$\begin{cases}
A_i X_{i,\infty} = 0,\\
v_{1,\infty} + v_{2,\infty} + \ldots + v_{N_i,\infty} = 1.
\end{cases} \tag{3.18}$$

Again, we can compute the time derivative of

$$\mathcal{E}(X_i|X_{i,\infty}) = \sum_{k=1}^{N_i} \int_\Omega \frac{|v_k|^2}{v_{k,\infty}} dx \tag{3.19}$$

as

$$\begin{aligned}
\mathcal{D}(X_i|X_{i,\infty}) &= -\frac{d}{dt}\mathcal{E}(X_i, X_{i,\infty}) \\
&= 2\sum_{i=1}^{N_i} d_{L[i-1]+k} \int_\Omega \frac{|\nabla v_k|^2}{v_{k,\infty}} dx + \sum_{k,\ell=1;k<\ell}^{N_i} (b_{k,\ell} v_{\ell,\infty} + b_{\ell,k} v_{k,\infty}) \int_\Omega \Big(\frac{v_k}{v_{k,\infty}} - \frac{v_\ell}{v_{\ell,\infty}}\Big)^2 dx \\
&\quad + 2\sum_{k=1}^{N_i} f_{L[i-1]+k} \int_\Omega \frac{|v_k|^2}{v_{k,\infty}} dx - 2\sum_{k=1}^{N_i} \int_\Omega \Big(\frac{v_k}{v_{k,\infty}} \sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\, u_\ell\Big) dx.
\end{aligned} \tag{3.20}$$

Because C_i is a transmission component, there exists an index $k_0 \in \{1, \ldots, N_i\}$ such that $f_{L[i-1]+k_0} > 0$ is positive. In comparison to (3.14), the dissipation $\mathcal{D}(X_i|X_{i,\infty})$ in (3.20) has the additional term

$$-2\sum_{k=1}^{N_i}\int_\Omega \left(\frac{v_k}{v_{k,\infty}}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\, u_\ell\right)dx$$

to be estimated. Thanks to the decay (3.16) of u_ℓ, we can estimate

$$\begin{aligned}\left|2\sum_{k=1}^{N_i}\int_\Omega \left(\frac{v_k}{v_{k,\infty}}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\, u_\ell\right)dx\right| &\le 2\sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_\Omega \left|\frac{v_k}{v_{k,\infty}}u_\ell\right|dx\\ &\le f_{L[i-1]+k_0}\sum_{k=1}^{N_i}\int_\Omega \frac{|v_k|^2}{v_{k,\infty}}dx + \kappa\sum_{\ell=1}^{L[i-1]}\|u_\ell\|^2\\ &\le f_{L[i-1]+k_0}\sum_{k=1}^{N_i}\int_\Omega \frac{|v_k|^2}{v_{k,\infty}}dx + \kappa K^* e^{-\lambda^* t} \end{aligned} \tag{3.21}$$

with $\kappa = N_i L[i-1]\max_{i<j}\{a_{ij}^2\}/(f_{L[i-1]+k_0}\min_k\{v_{k,\infty}\})$. Then, with the help of (3.21), we estimate

$$\mathcal{D}(X_i|X_{i,\infty}) \ge \sum_{k,\ell=1;k<\ell}^{N_i}(b_{k,\ell}v_{\ell,\infty}+b_{\ell,k}v_{k,\infty})\int_\Omega\left(\frac{v_k}{v_{k,\infty}}-\frac{v_\ell}{v_{\ell,\infty}}\right)^2dx + f_{L[i-1]+k_0}\int_\Omega\frac{|v_{k_0}|^2}{v_{k_0,\infty}}dx - \kappa K^* e^{-\lambda^* t},$$

and similarly to (3.15), we obtain for $\overline{\lambda} = \min\{\xi/4, f_{L[i-1]+k_0}/4N_i\}$,

$$\mathcal{D}(X_i|X_{i,\infty}) \ge \overline{\lambda}\,\mathcal{E}(X_i|X_{i,\infty}) - \kappa K^* e^{-\lambda^* t}. \tag{3.22}$$

From (3.22), we can use the classic Gronwall lemma to have

$$\mathcal{E}(X_i(t)|X_{i,\infty}) \le K_i e^{-\lambda_i t},$$

with $\lambda_i = \min\{\overline{\lambda}, \lambda^*\}$ and $K_i = 2\max\{\mathcal{E}(X_{i,0}|X_{i,\infty}), \kappa K^*\}$, which ends the proof in the case that C_i is a transmission component. □

For a target component, we need to define its corresponding equilibrium state. This equilibrium state balances the reactions within the component and has as total mass the sum of the initial total mass of the target component plus the total "injected mass" from the other components. In general, the injected mass will not be given explicitly but depend on the time evolution of the influences species prior to C_i in terms of the topological order.

Lemma 3.3 (Equilibrium state of target components)**.**
For each target component C_i, if

$$\sum_{k=1}^{N_i}\overline{u}_{L[i-1]+k,0} + \sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_0^{+\infty}\overline{u_\ell}(s)ds > 0 \tag{3.23}$$

holds, then there exists a unique positive equilibrium state $X_{i,\infty} = (v_{1,\infty}, v_{2,\infty}, \ldots, v_{N_i,\infty})$ satisfying

$$\begin{cases} A_i X_{i,\infty} = 0,\\ \sum_{k=1}^{N_i} v_{k,\infty} = \sum_{k=1}^{N_i}\overline{u}_{L[i-1]+k,0} + \sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_0^{+\infty}\overline{u_\ell}(s)ds. \end{cases} \tag{3.24}$$

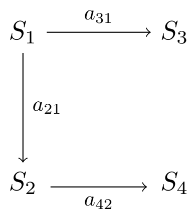
Otherwise, if the sum (3.23) should be zero, then the initial and the total injected mass into the target component C_i is zero and the concentrations of the target component C_i remain zero of all times.

Proof. By (3.16) we have for all $\ell = 1, 2, \ldots, L[i-1]$ that $\|u_\ell(t)\|^2 \le K^* e^{-\lambda^* t}$. Thus, Jensen's inequality yields

$$\int_0^{+\infty}\overline{u_\ell}(s)ds \le \int_0^{+\infty}\|u(s)\|_{L^2(\Omega)}^{1/2}ds \le K^*\int_0^{+\infty}e^{-\frac{\lambda^*}{2}s}ds = \frac{2K^*}{\lambda^*} \tag{3.25}$$

and the right hand side of the second equation in (3.24) is finite. Therefore, the existence of a unique $X_{i,\infty}$ satisfying (3.24) follows from Lemma 2.2. □

Remark 3.1. *The positive sign in assumption (3.23) ensures that either initially or during the ongoing reactions positive mass is present/injected into the component C_i. When this assumption does not hold, then the target component does not possess a positive equilibrium and all of its concentrations remain zero for all times. For example, consider the network*



when the initial data of all species are zero except S_3. In this case, the target component $\{S_4\}$ will not ever receive any mass, and thus remains zero for all $t > 0$.

We now begin the

Proof of Theorem 1.5. With the notations introduced in (3.1) and Lemma 3.3, we identify the indexes in the statement of Theorem 1.5 as $i_k = L[i-1]+k$ and the equilibrium state $u_{i_k,\infty} = v_{k,\infty}$ for $k = 1, \ldots, N_i$. The aim now is to prove for all $k = 1, \ldots, N_i$,

$$\|v_k(t) - v_{k,\infty}\|^2_{L^2(\Omega)} \le K_i e^{-\lambda_i t} \quad \text{for all } t \ge 0$$

for some explicit constants $K_i > 0$ and $\lambda_i > 0$.

We recall the system for a target component C_i,

$$\begin{cases} \partial_t X_i - D_i \Delta X_i = \mathcal{F}_i^{in} + A_i X_i, & x \in \Omega, \quad t > 0, \\ \partial_\nu X_i = 0, & x \in \partial\Omega, \quad t > 0, \\ X_i(x,0) = X_{i,0}(x), & x \in \Omega, \end{cases} \tag{3.26}$$

where

$$\mathcal{F}_i^{in} = \begin{pmatrix} z_{L[i-1]+1} \\ z_{L[i-1]+2} \\ \cdots \\ z_{L[i]} \end{pmatrix} \qquad \text{with} \qquad z_{L[i-1]+\ell} = \sum_{k=1}^{L[i-1]} a_{L[i-1]+\ell,k}\, u_k.$$

Note that the total mass of C_i is not conserved but increases in time due to the in-flow vector $\mathcal{F}_i^{in}$. To compute the total mass of C_i at a time $t > 0$, we sum up all the equations of (3.26) then integrating over Ω,

$$\frac{d}{dt} \sum_{k=1}^{N_i} \overline{u}_{L[i-1]+k}(t) = \sum_{k=1}^{N_i} \overline{z}_{L[i-1]+k}(t) = \sum_{k=1}^{N_i} \sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\, \overline{u_\ell}(t)$$

thanks to the homogeneous Neumann boundary condition and the fact that $(1, \ldots, 1)^T$ is a left eigenvector with eigenvalue zero of A_i since A_i is a reaction matrix. Thus, we have

$$\sum_{k=1}^{N_i} \overline{u}_{L[i-1]+k}(t) = \sum_{k=1}^{N_i} \overline{u}_{L[i-1]+k,0} + \sum_{k=1}^{N_i} \sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell} \int_0^t \overline{u_\ell}(s) ds. \tag{3.27}$$

Given that the right hand side of (3.27) should be zero for all times $t > 0$, then $\overline{u}_{L[i-1]+k}(t) = 0$ for all $k = 1, \ldots, N_i$ and for all $t > 0$ and $X_{i,\infty} = 0$ and the statement of the Theorem holds trivially.

Otherwise, if the right hand side of (3.27) is positive for some time $t > 0$, then assumption (3.23) is satisfied an $X_{i,\infty}$ is a positive equilibrium. Recalling the change of notation $v_k = u_{L[i-1]+k}$ in (3.10), we denote by

$$w_k(t) = v_k(t) - v_{k,\infty} = u_{L[i-1]+k}(t) - v_{k,\infty}$$

the distance from $u_{L[i-1]+k}$ to its corresponding equilibrium state for all $k = 1, 2, \ldots, N_i$. It implies that $(w_k)_{k=1,\ldots,N_i}$ solves the system (3.26) subject to the initial data $w_{k,0} = u_{L[i-1]+k,0} - v_{k,\infty}$ for all $k = 1, 2, \ldots, N_i$. We define $W_i = (w_1, w_2, \ldots, w_{N_i})$ and consider the relative entropy-like functional

$$\mathcal{E}(W_i | X_{i,\infty}) = \sum_{k=1}^{N_i} \int_\Omega \frac{|w_k|^2}{v_{k,\infty}} dx = \sum_{k=1}^{N_i} \int_\Omega \frac{|w_k - \overline{w_k}|^2}{v_{k,\infty}} dx + \sum_{k=1}^{N_i} \frac{\overline{w_k}^2}{v_{k,\infty}} =: \mathcal{E}_1 + \mathcal{E}_2. \tag{3.28}$$

By using again arguments of Lemma 2.3, we calculate the entropy dissipation

$$\begin{aligned}\mathcal{D}(W_i|X_{i,\infty}) &= -\frac{d}{dt}\mathcal{E}(W_i|X_{i,\infty})\\ &= 2\sum_{i=1}^{N_i} d_{L[i-1]+k}\int_\Omega \frac{|\nabla w_k|^2}{v_{k,\infty}}dx + \sum_{k,\ell=1;k<\ell}^{N_i}(b_{k,\ell}v_{\ell,\infty}+b_{\ell,k}v_{k,\infty})\int_\Omega\left(\frac{w_k}{v_{k,\infty}}-\frac{w_\ell}{v_{\ell,\infty}}\right)^2dx\\ &\quad - 2\sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_\Omega \frac{w_k}{v_{k,\infty}}u_\ell\,dx \end{aligned} \tag{3.29}$$

For the last term of (3.29), we estimate

$$\begin{aligned}&\left|2\sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_\Omega\frac{w_k}{v_{k,\infty}}u_\ell\,dx\right|\\ &\le 2\sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_\Omega\frac{|w_k-\overline{w_k}|}{v_{k,\infty}}|u_\ell|dx \;+2\sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\frac{|\overline{w_k}|}{v_{k,\infty}}|\overline{u_\ell}|\\ &\le C_P\sum_{k=1}^{N_i} d_{L[i-1]+k}\int_\Omega\frac{|w_k-\overline{w_k}|^2}{v_{k,\infty}}dx + \kappa_1\sum_{\ell=1}^{L[i-1]}\|u_\ell\|^2 + \kappa_2\sum_{k=1}^{N_i}\frac{\overline{w_k}^2}{v_{k,\infty}} + \kappa_3\sum_{\ell=1}^{L[i-1]}\overline{u_\ell}^2\\ &\le \sum_{k=1}^{N_i} d_{L[i-1]+k}\int_\Omega\frac{|\nabla w_k|^2}{v_{k,\infty}}dx + \kappa_2\sum_{k=1}^{N_i}\frac{\overline{w_k}^2}{v_{k,\infty}} + (\kappa_1+\kappa_3)K^*e^{-\lambda^* t}, \end{aligned}\tag{3.30}$$

with

$$\kappa_1 = \frac{N_iL[i-1]\max_{i<j}\{a_{ij}^2\}}{C_P\min_k\{d_{L[i-1]+k}v_{k,\infty}\}},\quad \kappa_2 = \frac{1}{2}\xi\max_k\{v_{k,\infty}\},\quad \kappa_3 = \frac{N_iL[i-1]\max_{i<j}\{a_{ij}^2\}}{\kappa_2 v_{k,\infty}},$$

where κ_2 is chosen in such a way that the last step of the below estimate (3.35) is fulfilled, and we have used $\|u_\ell(t)\|^2 \le K^*e^{-\lambda^* t}$ for all $\ell = 1,\ldots,L[i-1]$ in the last estimate. By inserting (3.30) into (3.29), we obtain

$$\begin{aligned}\mathcal{D}(W_i|X_{i,\infty}) &\ge \sum_{i=1}^{N_i} d_{L[i-1]+k}\int_\Omega\frac{|\nabla w_k|^2}{v_{k,\infty}}dx + \sum_{k,\ell=1;k<\ell}^{N_i}(b_{k,\ell}v_{\ell,\infty}+b_{\ell,k}v_{k,\infty})\left(\frac{\overline{w_k}}{v_{k,\infty}}-\frac{\overline{w_\ell}}{v_{\ell,\infty}}\right)^2\\ &\quad-\kappa_2\sum_{k=1}^{N_i}\frac{\overline{w_k}^2}{v_{k,\infty}} - (\kappa_1+\kappa_3)K^*e^{-\lambda^* t} =: \mathcal{D}_1+\mathcal{D}_2 \end{aligned}\tag{3.31}$$

where $\mathcal{D}_1$ is the term containing the gradients and $\mathcal{D}_2$ is the rest of the right hand side. It follows from Poincaré's inequality that

$$\mathcal{D}_1 \ge \sum_{i=1}^{N_i} d_{L[i-1]+k}\int_\Omega\frac{|\nabla w_k|^2}{v_{k,\infty}}dx \ge C_P\sum_{i=1}^{N_i} d_{L[i-1]+k}\int_\Omega\frac{|w_k-\overline{w_k}|^2}{v_{k,\infty}}dx \ge \kappa_4\mathcal{E}_1 \tag{3.32}$$

with $\kappa_4 = C_P\min_k\{d_{L[i-1]+k}\}$. To control $\mathcal{E}_2$, we use arguments similar to Step 3 in the proof of Lemma 2.4. First, by using (3.27), we have the total mass of $(w_k)_{1\le k\le N_i}$ is computed as,

$$\begin{aligned}\sum_{k=1}^{N_i}\overline{w_k}(t) = \sum_{k=1}^{N_i}\overline{u}_{L[i-1]+k}(t) - \sum_{k=1}^{N_i}v_{k,\infty} &= \sum_{k=1}^{N_i}\overline{u}_{L[i-1]+k,0} + \sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_0^t\overline{u_\ell}(s)ds\\ &\quad-\sum_{k=1}^{N_i}\overline{u}_{L[i-1]+k,0} - \sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_0^{+\infty}\overline{u_\ell}(s)ds\\ &= -\sum_{k=1}^{N_i}\sum_{\ell=1}^{L[i-1]} a_{L[i-1]+k,\ell}\int_t^{+\infty}\overline{u_\ell}(s)ds =: -\delta(t). \end{aligned}\tag{3.33}$$

Hence,

$$-2\sum_{k,\ell=1;k<\ell}^{N_i}\overline{w_k}\,\overline{w_\ell} = -\sum_{k,\ell=1;k\ne\ell}^{N_i}\overline{w_k}\,\overline{w_\ell} = \sum_{k=1}^{N_i}\overline{w_k}^2 - \sum_{k,\ell=1}^{N_i}\overline{w_k}\,\overline{w_\ell} = \sum_{k=1}^{N_i}\overline{w_k}^2 - \delta^2(t). \tag{3.34}$$

By using (2.16) and (3.34), we estimate

$$\begin{aligned}
\mathcal{D}_2 &\geq \xi \sum_{k,\ell=1;k<\ell}^{N_i} \left(\frac{\overline{w_k}}{v_{k,\infty}} - \frac{\overline{w_\ell}}{v_{\ell,\infty}}\right)^2 - \kappa_2 \sum_{k=1}^{N_i} \frac{\overline{w_k}^2}{v_{k,\infty}} - (\kappa_1+\kappa_3)K^* e^{-\lambda^* t}\\
&\geq -2\xi \max_{k<\ell}\{v_{k,\infty}v_{\ell,\infty}\} \sum_{k,\ell=1;k<\ell}^{N_i} \overline{w_k}\,\overline{w_\ell} - \kappa_2 \sum_{k=1}^{N_i} \frac{\overline{w_k}^2}{v_{k,\infty}} - (\kappa_1+\kappa_3)K^* e^{-\lambda^* t}\\
&= \xi \max_{k<\ell}\{v_{k,\infty}v_{\ell,\infty}\} \left(\sum_{k=1}^{N_i} \overline{w_k}^2 - \delta^2\right) - \kappa_2 \sum_{k=1}^{N_i} \frac{\overline{w_k}^2}{v_{k,\infty}} - (\kappa_1+\kappa_3)K^* e^{-\lambda^* t}\\
&\geq \frac{1}{2}\xi \max_k\{v_{k,\infty}\} \sum_{k=1}^{N_i} \frac{\overline{w_k}^2}{v_{k,\infty}} - \xi \max_{k<\ell}\{v_{k,\infty}v_{\ell,\infty}\}\delta^2 - (\kappa_1+\kappa_3)K^* e^{-\lambda^* t} \qquad (3.35)
\end{aligned}$$

for $\varepsilon > 0$ is sufficiently small. It follows from (3.33) and $\overline{u_\ell} \leq \|u_\ell\| \leq \sqrt{K^*}e^{-\lambda^* t/2}$ that

$$\delta^2 \leq N_i L[i-1] \max_{i<j}\{a_{ij}^2\} \sum_{\ell=1}^{L[i-1]} \left(\int_t^{+\infty} \overline{u_\ell}(s)ds\right)^2 \leq \kappa_4 e^{-\lambda^* t}$$

with $\kappa_4 = 4K^* N_i L[i-1]^2 \max_{i<j}\{a_{ij}^2\}(\lambda^*)^{-2}$. Hence, (3.35) implies that

$$\mathcal{D}_2 \geq \frac{1}{2}\xi \max_k\{v_{k,\infty}\} \sum_{k=1}^{N_i} \frac{\overline{w_k}^2}{v_{k,\infty}} - \max\{\kappa_4 \xi \max_{k<\ell}\{v_{k,\infty}v_{\ell,\infty}\}, (\kappa_1+\kappa_3)K^*\} e^{-\lambda^* t} = \kappa_5 \mathcal{E}_2 - \kappa_6 e^{-\lambda^* t}. \qquad (3.36)$$

Combining (3.36) and (3.32) yields

$$\mathcal{D}(W_i|X_{i,\infty}) \geq \min\{\kappa_4, \kappa_5\}\mathcal{E}(W_i|X_{i,\infty}) - \kappa_6 e^{-\lambda^* t}. \qquad (3.37)$$

Therefore, by applying a classic Gronwall lemma,

$$\mathcal{E}(W_i(t)|X_{i,\infty}) \leq K_i e^{-\lambda_i t} \qquad \text{for all } t \geq 0 \qquad (3.38)$$

with $\lambda_i = \min\{\kappa_4, \kappa_5, \lambda^*\}$ and $K_i = 2\max\{\mathcal{E}(X_{i,0}|X_{i,\infty}), \kappa_6\}$. This completes the proof of the Theorem. □

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