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FAST-REACTION ASYMPTOTICS FOR A TIME-DEPENDENT REACTION-DIFFUSION SYSTEM WITH A GROWING NONLINEAR SOURCE TERM

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Abstract. In this paper, we prove rigorously the fast-reaction asymptotics $\lambda \to \infty$ for a reaction-diffusion system having a nonlinear production term with very rapid reaction rate λ. We derive the limit PDE system and prove the uniqueness of its solutions. The key tools of our analysis include (uniform w.r.t. λ) L^1-estimates for both q and w and a *balanced formulation*, where combinations of the original components which balance the fast reaction are used. The results reported here answer some open questions raised by T.I. Seidman in the paper [16].

1. Introduction

If an irreversible chemical reaction $A + B \rightharpoonup C$ is very much faster than diffusive mass transport, one expects to see a separation into regions where A is present with B almost non-existent and vice versa, with a narrow separating interfacial reaction zone where diffusion brings the components together. This is often then modeled by a free boundary problem. We are here concerned to justify that model by considering the existence of a limit for the 'true' situation with fast but finite reaction rate.

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As a specific model problem we are considering the chemical reaction $2A + B \rightharpoonup$ products following the reaction path

$$A + B \overset{\lambda}{\rightharpoonup} C, \qquad A + C \overset{\mu}{\rightharpoonup} \text{ products} \tag{1}$$

involving an intermediate compound C. Here λ, μ denote rate constants for the reactions; the reaction rate for the fast reaction is given by $\lambda \gg 1$ with time scaled so $\mu = 1$ for the slower reaction. We will then be interested in the asymptotics for the reaction-diffusion problem as $\lambda \to \infty$. [If there is indeed convergence to a limit, one might hope that this limit solution can be computed more easily than a solution with large finite λ and will provide a good approximation for that.]

The situation indicated in (1) is rather basic – it enters as a distinct component in a variety of complex chemical scenarios where different slow and fast characteristic reaction times interplay with a moderate characteristic transport time. We refer the reader to standard monographs like [6] and [8] for concrete examples of (1), but also to the celebrated paper by Nernst [14] where similar problems were addressed.

A few questions arise naturally here (see also [16]):

(Q1) What happens with the reaction-diffusion system as $\lambda \to \infty$?
(Q2) Can we show some convergence of the solution vector to a limit?
(Q3) Is there a well-defined characterization of the limit system?
(Q4) How can we approximate numerically in an efficient way the solutions of the limit system?
(Q5) What happens with the system as $t \to \infty$? Is this at all related to the asymptotics as $\lambda \to \infty$?

The target of this paper is to address the questions (Q1)–(Q4).

[For the model reaction-diffusion system (2)–(4) one might additionally consider (a) analysis of the initial transient, (b) singular perturbation analysis of the fine structure of the interface for large finite λ and its regularity (especially through topology changes) for the limit solutions, (c) the dynamics of this free boundary, (d) stability of the steady state solutions, (e) Bodenstein (QSSA) approximation, and other related questions — including (Q5), which remains open at present. We note that we will here be assuming equal diffusion constants as necessary for the present approach, but one would obviously be interested in all of these questions in the more general setting.]

Note that many things are already known for the stationary version of our reaction-diffusion system; for this we refer the reader to [17, 12, 16]. However, none of the techniques developed there or in other papers where the fast reaction asymptotics is dealt with analytically (as, e.g. [2, 5, 7, 13]) are applicable to the non-stationary case presented here: conceptually new estimates are needed in order to be able to pass to the limit $\lambda \to \infty$. The results we report here answer some of the open questions raised by the first author in [16]. In particular, we will use a compactness argument to show subsequential convergence and then a uniqueness argument to show that one actually has convergence as $\lambda \to \infty$. Of course, for the compactness argument we will need some estimates uniform in λ and our first efforts will go toward these.

The paper is organized as follows: Section 2 formulates the specific model system we are analyzing and notes our hypotheses. In Section 3 the uniform estimates needed to pass to the limit $\lambda \to \infty$ are obtained and in Section 4 a compactness argument is applied to show subsequential convergence. In Section 5.1 we then derive a new system related to the limit solutions and prove the uniqueness of its (weak) solutions in Section 5 and so convergence as $\lambda \to \infty$. Finally, in Section 6 we compare the computational use of the original system (with λ very large) and some related systems in illustrating the behavior of the concentration profiles and of the developing interface separating the species A and B. An Appendix (Section 7) gives the proof of the abstract compactness theorem used in Section 4.

2. Formulation

We denote by $u(x,t)$, $v(x,t)$, $w(x,t)$ the molar concentrations of the chemical species A, B, C, respectively, at position $x \in \Omega$ and time $0 \le t \le T$ where Ω is a bounded spatial region in $\mathbb{R}^d$ and $T > 0$ is fixed but arbitrary; thus we have $(x,t) \in Q = Q_T = [0,T] \times \Omega$. Our model is then the following system of reaction-diffusion equations on Q_T:

$$
\left\{
\begin{array}{rcll}
u_t &=& \Delta u & -\lambda uv - uw \;\text{ in } Q_T \\
u &=& \alpha & \text{on } [0,T] \times \Gamma_A \\
u_\nu &=& 0 & \text{on } [0,T] \times [\partial\Omega \setminus \Gamma_A] \\
u &=& u_0 & \text{at } \{t=0\} \times \bar{\Omega},
\end{array}
\right. \tag{2}
$$

$$
\left\{
\begin{array}{rcll}
v_t &=& \Delta v & -\lambda uv \;\text{ in } Q_T \\
v &=& \beta & \text{on } [0,T] \times \Gamma_B \\
v_\nu &=& 0 & \text{on } [0,T] \times [\partial\Omega \setminus \Gamma_B] \\
v &=& v_0 & \text{at } \{t=0\} \times \bar{\Omega},
\end{array}
\right. \tag{3}
$$

$$
\left\{
\begin{array}{rcll}
w_t &=& \Delta w & +\lambda uv - uw \;\text{ in } Q_T \\
w_\nu &=& 0 & \text{on } [0,T] \times \partial\Omega \\
w &=& w_0 & \text{at } \{t=0\} \times \bar{\Omega}.
\end{array}
\right. \tag{4}
$$

where Γ_A, Γ_B are disjoint relatively closed, nonempty subsets of $\partial\Omega$.

It will be occasionally be convenient to write $u^\lambda, v^\lambda, w^\lambda$ to indicate the dependence on λ of the solutions u, v, w; we will write $q := q^\lambda$ for the production term λuv. On $\partial\Omega$, f_ν denotes outward flux $\nabla f \cdot \nu$, where ν is the exterior normal vector to $\partial\Omega$.

We will view the data as extended to all of Q_T so u_0, v_0 and w_0 denote functions on Q_T satisfying the homogeneous partial differential equations:

$$u_{0t} = \Delta u_0, \qquad u_{0t} = \Delta u_0, \qquad u_{0t} = \Delta u_0 \tag{5}$$

on Q_T with initial and boundary conditions exactly as for the equations (2), (3), (4). We will later find it convenient to have introduced the solution θ of the elliptic problem

$$\begin{gathered} \Delta\theta = 0 \text{ on } \Omega \\ \theta|_{\Gamma_A} = 0, \quad \theta|_{\Gamma_B} = 1, \quad \theta_\nu = 0 \text{ else on } \partial\Omega. \end{gathered} \tag{6}$$

Note that θ and the data u_0, v_0, w_0 are independent of λ.

Due to our choice of boundary conditions, we have a situation where one has potentially unlimited external supplies of A, B as well as consideration of the component C which is being created by the fast reaction. Computational simulation and analysis in the steady state case show that the production term q grows (in sup-norm) as $\mathcal{O}(\lambda^{1/3})$ when $\lambda \to \infty$ yet, for our purposes, we will need estimates for q and w which are independent of λ.

2.1. **Technical assumptions.**

We assume throughout that

$$\begin{array}{lll} 0 < \alpha \le a, \quad 0 < \beta \le b & \text{on } [0,T] \times \partial\Omega, & \\ 0 \le u_0 \le a, \quad 0 \le v_0 \le b & \text{on } \Omega \text{ at } t = 0 & \text{with } u_0 v_0 \equiv 0, \\ 0 \le w_0 = \text{bounded} & \text{on } \Omega \text{ at } t = 0. & \end{array} \tag{7}$$

We note that under mild regularity conditions on the geometry it follows that:

$$\begin{gathered} 0 \le u \le a, \qquad 0 \le v \le b, \qquad 0 \le w, \\ 0 \le u_0,\ v_0,\ w_0 \in L^\infty(Q_T), \end{gathered} \tag{8}$$

[e.g., taking $u_- = \min\{u, 0\}$ as test function in the weak form of (2)]. Similarly we have

$$0 \le \theta \le 1 \tag{9}$$

and, again as a mild regularity condition on the geometry, we further assume that (6) gives

$$\nabla\theta \in L^\infty(\Omega) \qquad (\text{so } \theta_\nu \in L^\infty(\partial\Omega)). \tag{10}$$

It is standard that the operator $-A = \Delta$, for each of the types of boundary conditions in (2)–(4), is closed, selfadjoint and nonpositive, hence generates an analytic semigroup $S(\cdot)$ on $L^2(\Omega)$. We now assume (compare [1], [9]) that, for each of these sets of boundary conditions,

$$-A = \Delta \text{ generates a } C_0 \text{ semigroup } S(\cdot) \text{ on } L^1(\Omega) \tag{11}$$

which coincides with $S_2(\cdot)$ on $L^2(\Omega)$.

Lemma 2.1. *For each case of* (11)*, $S(t)$ is compact for $t > 0$.*

Proof. To see this, observe that (by interpolation, duality and selfadjointness) one has $A^{-s} : L^1(\Omega) \subset [C(\Omega]^* \to L^2(\Omega)$ for some $s > 0$, whence one has $S(t) = [A^s S_2(t)]\, A^{-s}$ with $A^s S_2(t)$ bounded. Compactness of $S(t)$ on $L^1(\Omega)$ then follows, e.g., from the known relation of $\mathcal{D}(A^s)$ to $H^{2s}(\Omega)$ (compare [10], [11]) and the compact embedding of $H^{2s}(\Omega)$ into $L^2(\Omega)$ and so into $L^1(\Omega)$. □

Finally, we impose two additional technical conditions needed only for the uniqueness argument in Section 5. We will assume a bit more regularity for the data

$$\alpha, \beta \in L^\infty([0,T] \to H^s(\partial\Omega)) \text{ for some } s > 1/2$$

and will also assume that

the Neumann trace map ∂_ν is compact from $H^1(\Omega)$ to $[H^s(\partial\Omega)]^*$.

What we actually need is a consequence of these:

$$\begin{aligned} &\textit{For } \varepsilon > 0 \textit{ there is a constant } C_\varepsilon \textit{ such that} \\ &\left| \int_{\partial\Omega} \alpha(t,\cdot)\, f_\nu \right| \le \varepsilon \|f\| + C_\varepsilon \|\nabla f\| \qquad \textit{for } f \in H^1(\Omega), t \in [0,T] \end{aligned} \tag{12}$$

and similarly for β. Next we assume the dimension $d = 1, 2, 3$ so, by the Sobolev Embedding Theorem and [10, 11], we have

$$\begin{aligned} &\text{For } \sigma > d/4 \text{ one has} \\ &\|f\|_{L^\infty(\Omega)} \quad \le \quad M\|f\|_{H^{2\sigma}(\Omega)} \quad \le \quad M'\|(-\Delta)^\sigma f\| \end{aligned} \tag{13}$$

for f with $(-\Delta)^\sigma f \in L^2(\Omega)$ with $\sigma < 1$.

Summarizing, we assume (7) (so (8)) and (9)—(13).

3. Estimates uniform in λ

If (7) holds, it was already noted in [16, Theorem 1.1] that the problem (2)–(4) has a unique global solution for each λ with $0 \le u \le a$, $0 \le v \le b$ and $0 \le w$ as in (8). In this section, we give the following new results, independent of λ :

(i) an $L^1(Q_T)$ estimate for $q^\lambda = \lambda uv$,

(ii) an $L^1(\Omega)$ estimate for w^λ, also independent of $t \in [0,T]$.

In the following we consistently use K to indicate a positive constant independent of λ. These λ-independent estimates (i) and (ii) will be crucial tools in the compactness argument of the next section.

3.1. **Estimate for q^λ.** Using θ, given by (6), as test function in the weak form of (2) gives

$$\begin{aligned}\left(\int_\Omega \theta u\right)_t + \int_\Omega \theta q \;&\le\; \int_\Omega \theta(\Delta u) = -\int_\Omega \nabla\theta\cdot\nabla u \\ &= \int_\Omega (\Delta\theta)u - \int_{\partial\Omega} (\theta_\nu)\, u \qquad \le Ka,\end{aligned}$$

noting that the boundary term from the first use of the Divergence Theorem vanishes by our choice of the boundary conditions in (6); the constant K here depends only on $\nabla\theta$ [as indicated in (10)] independently of λ. Integrating this over $[0,T]$ and using the fact that $\theta u \ge 0$, we obtain

$$\int_{Q_T} \theta q \;\le\; \int_\Omega \theta u_0 + \int_0^T Ka \;\le\; K(1+T)a. \tag{14}$$

Similarly, using $(1-\theta)$ as test function in the weak form of the equation (3) for v, we obtain, since $(1-\theta)v \ge 0$,

$$\int_{Q_T} (1-\theta)q \;\le \int_\Omega \theta v_0 + \int_0^T Kb \;\le\; K(1+T)b. \tag{15}$$

Adding (14) to (15), we obtain

$$\|q^\lambda\|_{L^1(Q_T)} \;=\; \int_{Q_T} q^\lambda \;\le\; K, \tag{16}$$

Note that K here depends on u_0, v_0, and the constants of (14), (15) with increase linear in T, but is independent of λ so $\{q^\lambda\}$ is uniformly bounded in $L^1([0,T]\to L^1(\Omega))$ as $\lambda\to\infty$.

Remark 1. We never used here the fact that q has the particular form $q^\lambda = \lambda u^\lambda v^\lambda$: all we are really using is that $q \ge 0$ and the term $-q$ in never drives u or v negative.

We note that a somewhat related situation when L^1-bounds on production terms growing linearly in T enter the game is reported, for instance, in [7]. Note that in the scenario described in [7], the precise structure of the reaction-diffusion system (with homogeneous Neumann boundary conditions) is specific to reversible chemical reactions and allows for the construction of entropy and dissipation functionals completely describing the evolution of the system. Essentially, the entropy inequality for finite λ is there preserved during the limit process $\lambda\to\infty$; see also [3].

3.2. **Estimate for w^λ.** Here we prove that $w(t)$ is bounded in $L^1(\Omega)$ independently of λ and uniformly on $[0,T]$ — which just means that we are bounding the total amount of C in Ω, independently of the chemical reaction rate for production of C from A, B. We will later obtain a stronger result, but this estimate is now an immediate consequence of (16).

Integrating (4) over Ω, we get $\left(\int_\Omega w\right)_t \leq \int_\Omega q$ since $\int(-\Delta w) = 0$ and $-uw \leq 0$. On integrating this over $[0,t]$ one then obtains

$$\int_\Omega w^\lambda(t) \;\leq \int_\Omega w_0 + \int_0^t \int_\Omega q^\lambda \;\leq\; K. \tag{17}$$

Here the bound K of (17) depends only on w_0 and the constant of (16) and so is again independent of λ; this K also grows linearly in T.

4. Compactness and subsequential convergence

We begin with an abstract compactness result which we will later use with $X = L^1(\Omega)$ and $p = 1$. Here, however, X is an arbitrary Banach space and $\mathcal{X}_p$ denotes $L^p([0,T] \to X)$ for $1 \leq p \leq \infty$.

Theorem 4.1. *Let $S(\cdot)$ be a C_0 semigroup on X with infinitesimal generator $-A$; assume $S(t)$ is compact for each $t > 0$. Then the solution map $L : g \mapsto u$ of the differential equation*

$$u_t + Au = g \quad \text{on } [0,T], \qquad u(0) = 0, \tag{18}$$

given by

$$u(t) := \int_0^t S(t-s)\, g(s)\, ds \qquad \text{for } t \in [0,T],\; g \in \mathcal{X}_1, \tag{19}$$

is a well-defined compact operator: $\mathcal{X}_1 \to \mathcal{X}_p$ for arbitrary $1 \leq p < \infty$.

Proof. Since it is independent of our other considerations, the proof of Theorem 4.1 will be deferred to the Appendix (Section 7). □

Our principal result on subsequential convergence is the following:

Theorem 4.2. *For each sequence $\lambda_k \to \infty$ there is a subsequence $\lambda_{k(j)}$ and functions $\bar{u}, \bar{v}, \bar{w}$ and measure $\bar{q}$ (possibly depending on the choice of subsequence), for which*

$$u_j = u^{\lambda_{k(j)}} \to \bar{u}, \qquad v_j = v^{\lambda_{k(j)}} \to \bar{v}, \qquad w_j = w^{\lambda_{k(j)}} \to \bar{w} \tag{20}$$

(in each case both strong convergence in $\mathcal{X}_1 = L^1(Q_T)$ and also pointwise a.e. convergence on Q_T) with $q_j = q^{\lambda_{k(j)}} \xrightarrow{*} \bar{q}$ (weak-$*$ convergence in the dual space $[C(Q_T)]^*$).

Proof. We begin with the sequence $q^\lambda = q^{\lambda_k}$. From Subsection 3.1, we know that $\{q^\lambda\}$ is bounded in $L^1(Q_T)$, which embeds in the dual space $[C(Q_T)]^*$. Hence, using Alaoglu's Theorem, we may extract a weak-$*$ convergent subsequence; abusing notation slightly, we continue to denote this by λ_k.

The argument next proceeds (independently and essentially identically) for each of u, v, w. We will use the fact (11) that $-A = \Delta$ generates a C_0 semigroup $S(\cdot)$ on $X = L^1(\Omega)$ for each of the (homogeneous) boundary conditions. We have already, just following (11), noted the compactness of $S(t)$ so Theorem 4.1 can be applied.

For the sequence $u = u^\lambda = u^{\lambda_k}$ we set $\hat{u} := u^{\lambda_k} - u_0$. Then $\hat{u}$ satisfies the homogeneous boundary conditions and so satisfies the differential equation (18) with $g = -(q + uw)$ whence $u = u_0 + Lg$ for this g. The λ-independent estimates of Subsections 3.1 and 3.2 show that $\{g = g_k^\lambda\}$ is bounded in $\mathcal{X}_1 = L^1(Q_T)$ so, by Theorem 4.1, we have total boundedness of $\{Lg^{\lambda_k}\} \subset \mathcal{X}_1$ and we can extract a subsequence strongly convergent to $\bar{u}$. As usual, if we further extract a subsequence converging rapidly enough in norm, there is no loss in generality to ask hat we also have convergence pointwise a.e. on Q_T.

Further extracting subsequences, we may proceed similarly to ask also that $v^{\lambda_k(j)} \to \bar{v}$ and $w^{\lambda_k(j)} \to \bar{w}$ in the same senses. $\square$

4.1. **The limit system.** We next wish to claim that these limit functions of Theorem 4.2 will satisfy the limit system

$$\begin{aligned} \bar{u}_t &= \Delta\bar{u} - \bar{q} - \bar{u}\bar{w} \\ \bar{v}_t &= \Delta\bar{v} - \bar{q} \\ \bar{w}_t &= \Delta\bar{w} + \bar{q} - \bar{u}\bar{w} \end{aligned} \tag{21}$$

with the appropriate initial and boundary conditions, just as in (2), (3), (4), noting that those are independent of λ.

Since $[\partial_t - \Delta]$ is, in each case, a closed operator, we have the desired convergence (in some appropriate space) of each term, except possibly the product term where we must show that $u_j w_j \to \bar{u}\bar{w}$. Since $u_j \to \bar{u}$ and $w_j \to \bar{w}$ pointwise a.e., the same is true for the product and we easily see that this holds in $L^1(Q_T)$-norm. [To see this strong convergence, we note that

$$u_j w_j - \bar{u}\bar{w} = (u_j - \bar{u})\bar{w} + u_j(w_j - \bar{w}).$$

The second term has $L^1(Q_T)$-norm bounded by $a\|w_j - \bar{w}\|_1 \to 0$ as $k \to \infty$. The first term is bounded by the L^1 function $a\bar{w}$ so one has

$$\|(u_j - \bar{u})\bar{w}\|_{L^1(Q)} = \int_{Q_T} |u_j - \bar{u}|\bar{w}$$

then going to 0 by Lebesgue's Dominated Convergence Theorem and pointwise a.e. convergence to 0 of $(u_j - \bar{u})$. Hence $u_j w_j \to \bar{u}\bar{w}$ in $L^1(Q_T)$.] Thus one obtains (21).

5. Uniqueness

While the system (21) can be solved uniquely for $\bar{u}, \bar{v}, \bar{w}$, if one is given $\bar{q}$, we recall that $\bar{q}$ was itself obtained in Theorem 4.2 above by a compactness argument and so may possibly depend on the selection of the subsequence $\lambda_{k(j)}$. Our goal in this section is to show that such subsequential dependence cannot occur, that these limit functions are already uniquely determined by (21) *without* information about $\bar{q}$. This will then show that we actually have convergence in $L^1(Q_T)$ (as contrasted with the subsequential convergence in Theorem 4.2) for $\{u^\lambda, v^\lambda, w^\lambda\}$ as $\lambda \to \infty$.

5.1. **An auxiliary system.** For this uniqueness we turn to a trick introduced in [16]: if we consider the auxiliary function

$$y = \bar{u} - \bar{v}, \tag{22}$$

we have $y_t = \Delta y - \bar{u}\bar{w}$ on taking the difference of the first equations in (21) — and note that the difficult production term $\bar{q}$ has cancelled out and no longer appears. This subsection is devoted to the derivation of a self-contained system which will make this idea usable.

Our first observation is that, much as in Subsection 4.1 above, we have $u_j v_j \to \bar{u}\bar{v}$. On the other hand, since $\{q_j = \lambda_{k(j)} u_j v_j\}$ is bounded it follows that

$$\|u_j v_j\|_{L^1(Q)} = \|q_j\|/\lambda_{k(j)} \quad \to 0$$

so $\|\bar{u}\bar{v}\|_{L^1(Q)} = 0$ and the function $\bar{u}\bar{v}$ must vanish identically. Since $\bar{u}, \bar{v} \geq 0$, a consequence of this identity $\bar{u}\bar{v} \equiv 0$ is that (22) gives

$$y_+ := \max\{y, 0\} \equiv \bar{u}, \qquad y_- := \min\{y, 0\} \equiv -\bar{v}, \tag{23}$$

as $y > 0$ requires $\bar{u} > 0$ so $\bar{v} = 0$ and where $y = y_- < 0$ one has $\bar{u} = 0$ and $y = \bar{v}$.

To deal with the product term $\bar{u}\bar{w}$, the paper [16] introduced another auxiliary function $z = w + v$, noting that this satisfies $z_t = \Delta z - \bar{u}\bar{w}$, again with $\bar{q}$ no longer appearing. This choice of z gives a somewhat awkward coupling in the boundary conditions and it will here be more convenient to choose to work with the combination

$$z = \bar{w} + [\theta\bar{u} + (1-\theta)\bar{v}] \quad = \bar{w} + \bar{v} + \theta y \tag{24}$$

with θ given by (6). Using (23), one has

$$\bar{w} = z - \theta y_+ + (1-\theta)y_-] \qquad \bar{u}\bar{w} = y_+(z - \theta y_+). \tag{25}$$

This will enable us to make the system self-contained.

From the boundary condition in (2) and (7) we have $\bar{u} = \alpha > 0$ on Γ_A so we must have $\bar{v} = 0$ there whence $y = \alpha$; similarly on Γ_B so, using (25) in the equation, y satisfies

$$\begin{aligned} y_t &= \Delta y - y_+(z - \theta y_+) \\ y &= \begin{cases} \alpha & \text{on } [0,T]\times\Gamma_A \\ -\beta & \text{on } [0,T]\times\Gamma_B \end{cases} \\ y_\nu &= 0 \qquad \text{on } [0,T]\times[\partial\Omega\setminus(\Gamma_A\cup\Gamma_B)], \\ y &= u_0 - v_0 \qquad \text{at } \{t=0\}\times\bar{\Omega}, \end{aligned} \tag{26}$$

Noting, for example, that $\theta\Delta y = \Delta(\theta y) - 2\nabla\theta\cdot\nabla y$, we obtain the differential equation for z in self-contained form and now need self-contained conditions to adjoin to that; we begin with $z_\nu = \bar{w}_\nu + \bar{v}_\nu + (\theta y)_\nu = \bar{w}_\nu + \bar{v}_\nu + \theta y_\nu + \theta_\nu y$. On Γ_A we have $\bar{v}_\nu, \bar{w}_\nu = 0$ and $\theta = 0$ with $y = \alpha$ and, similarly, on Γ_B we have $\bar{u}_\nu, \bar{w}_\nu = 0$ and $\theta = 1$ with $y = -\beta$. Putting these together, the choice (24) of z satisfies

$$\begin{aligned} z_t &= \Delta z - 2\nabla\theta\cdot\nabla y - (1+\theta)y_+(z-\theta y_+) \\ z_\nu &= \begin{cases} \theta_\nu\alpha & \text{on } [0,T]\times\Gamma_A \\ -\theta_\nu\beta & \text{on } [0,T]\times\Gamma_B \\ 0 & \text{on } [0,T]\times[\partial\Omega\setminus(\Gamma_A\cup\Gamma_B)] \end{cases} \\ z &= w_0 + [\theta u_0 + (1-\theta)v_0] \qquad \text{at } \{t=0\}\times\bar{\Omega}. \end{aligned} \tag{27}$$

The differential equation in (27) for the auxiliary function of (24) is more complicated than the differential equation of (36) used in [16], but the boundary conditions now involve only fixed terms, without any coupling.

Much as with (5), we can extend the data in (27) to all of Q_T as z_0 satisfying

$$\begin{aligned} z_{0t} &= \Delta z_0 \\ z_{0\nu} &= \begin{cases} \theta_\nu\alpha & \text{on } [0,T]\times\Gamma_A \\ -\theta_\nu\beta & \text{on } [0,T]\times\Gamma_B \\ 0 & \text{on } [0,T]\times[\partial\Omega\setminus(\Gamma_A\cup\Gamma_B)] \end{cases} \\ z_0 &= w_0 + [\theta u_0 + (1-\theta)v_0] \qquad \text{at } \{t=0\}\times\bar{\Omega}. \end{aligned} \tag{28}$$

Since (8) and (10) give pointwise bounds for $z_{0\nu}$ on $[0,T]\times\partial\Omega$ and for z_0 on Ω at $t = 0$, it easily follows (e.g., by a weak maximum principle comparison argument) that $z_0 \in L^\infty(Q_T)$.

5.2. **Additional estimates.** As a preliminary to the uniqueness proof in the next subsection we will obtain an estimate for z, giving stronger estimates for $\bar{w}$ than the $L^1(Q_T)$ estimate in Subsection 3.2.

Begin by using y as test function in the weak form of (26) to get

$$\left(\tfrac{1}{2}\|y\|^2\right)_t + \|\nabla y\|^2 = \int_{\partial\Omega} y y_\nu - \int_\Omega y\bar{u}\bar{w} \quad \le \int_{\Gamma_A} \alpha y_\nu - \int_{\Gamma_B} \beta y_\nu$$

since $y\bar{u}\bar{w} \ge 0$ and y_ν vanishes on $\partial\Omega \setminus [\Gamma_A \cup \Gamma_B]$. Applying (12) to $f = y(t)$ bounds the right hand side above by, e.g., $c + \frac{1}{2}\|\nabla y(t)\|^2$ for some c, so integrating over $(0,t)$ gives a bound on $|\nabla y|$ in $L^2(Q_T)$.

Next, we note that $\bar{u}\bar{w} = y_+\hat{z} + [y_+ z_0 - \theta(y_+)^2]$ so $\hat{z} = z - z_0$ satisfies the equation

$$\hat{z}_t = \Delta\hat{z} - (1+\theta)y_+\hat{z} - h_0$$

with $h_0 = 2\nabla\theta\cdot\nabla y - (1+\theta)y_+(z_0 - \theta y_+)$ and with $\hat{z}_\nu \equiv 0$. Note that our assumptions and the estimate above for ∇y ensure a bound for h_0 in $L^2(Q_T)$. Using $\hat{z}$ as test function in the weak form here, we obtain

$$(\tfrac{1}{2}\|\hat{z}\|^2)_t + \|\nabla\hat{z}\|^2 + \int_\Omega (1+\theta)y_+\hat{z}^2 = \int_\Omega \hat{z}h_0 \quad \le \quad \tfrac{1}{2}\|\hat{z}\|^2 + \tfrac{1}{2}\|h_0\|^2$$

and, integrating, Gronwall's Inequality bounds $\hat{z}$ in $L^\infty([0,T] \to L^2(\Omega))$. We now consider this equation rewritten as

$$\hat{z}_t = \Delta\hat{z} - h_1 \qquad \text{with } h_1 := h_0 + (1+\theta)y_+\hat{z}.$$

Note that we have a bound for h_1 in $L^2(Q_T)$ since we now know $\hat{z} \in L^2(Q_T)$. For this we can use the semigroup formula

$$\hat{z}(t) = \int_0^t S(t-s)\,h_1(s)\,ds \tag{29}$$

where $S(\cdot)$ is here the analytic semigroup on $L^2(\Omega)$ generated by Δ with homogeneous Neumann boundary conditions. With $0 < \sigma < 1$ as in (13) we then have

$$\begin{aligned}\|\hat{z}(t)\|_{L^\infty(\Omega)} &\le M'\|(-\Delta)^\sigma\hat{z}(t)\| \\ &\le M'\int_0^t \|(-\Delta)^\sigma S(t-s)\|\,\|h_1(s)\|\,ds\end{aligned}$$

As $S(\cdot)$ is an analytic semigroup, $\|(-\Delta)^\sigma S(t-s)\| \le M(t-s)^{-\sigma}$, which is in $L^p(0,T)$ for $1 < p < 1/\sigma < 4/d$. Thus, $\|\hat{z}(\cdot)\|_{L^\infty(\Omega)}$ is bounded by the convolution of this L^p function and the $L^2(0,T)$ function $\|h_1(\cdot)\|$. This convolution is then in $L^r(0,T)$ where $r = \infty$ if we may take $p \ge 2$ (i.e., for $d = 1$) and, by Young's Inequality, $r = 2p/(2-p)$ when $d = 2,3$ so $p < 2, 4/3$. It follows that also $\bar{w} = \hat{z} + z_0 - [\theta y_+ - (1-\theta)y_-]$ is similarly bounded so there is a scalar function

$$\omega(\cdot) \in L^r(0,T) \qquad \text{giving } 0 \le \bar{w} \le \omega(t). \tag{30}$$

5.3. **Proof of uniqueness.** We are now ready to show uniqueness. From (23) and (25) it follows that the triple of functions $\bar{u}, \bar{v}, \bar{w}$ can be recovered from the pair y, z so a proof of uniqueness for the system (26), (27) — which was constructed so it no longer involves the unknown measure $\bar{q}$ — will consequently give uniqueness of the limit and so convergence as $\lambda \to \infty$ for the system (2), (3), (4).

Theorem 5.1. *For $d = 1, 2, 3$, the solution y, z of the system (26), (27) is unique.*

Proof. Suppose we had two solutions y_1, z_1 and y_2, z_2 (e.g., corresponding to different subsequential limits $\bar{u}_i, \bar{v}_i$, and $\bar{w}_i$ for $i = 1, 2$). We modify our notation to set $y := y_1 - y_2$ and $z := z_1 - z_2$; we also set $u = \bar{u}_1 - \bar{u}_2$ and $w = \bar{w}_1 - \bar{w}_2$. From (26), (27) these differences satisfy

$$
\begin{aligned}
(31)\qquad y_t &= \Delta y + \eta \qquad \text{with} \\
&\ \eta := -\left[\bar{u}_1\bar{w}_1 - \bar{u}_2\bar{w}_2\right] = -\bar{u}_1 w - u\bar{w}_2 \\
(32)\qquad z_t &= \Delta z + \zeta \qquad \text{with} \\
&\ \zeta := -\left[2\nabla\theta \cdot \nabla y + (1+\theta)\eta\right]
\end{aligned}
$$

It is important to observe that we now have homogeneous initial and boundary conditions for (31), (32) since the initial and boundary conditions in (26), (27) involve only fixed functions which then cancel on taking differences.

We proceed to bound η pointwise in terms of y, z. We first observe that $|u| \le |y|$, which is trivial if $\bar{u}_1, \bar{u}_2 > 0$ (so $u = y_1 - y_2 = y$) or if $\bar{u}_1, \bar{u}_2 = 0$ and $\bar{u}_1 > 0$, $\bar{u}_2 = 0$ gives $y_1 = \bar{u}_1$, $y_2 \le 0$ so $0 < u = y_1 \le y$; similarly, $|v| \le |y|$. Since $0 \le \bar{u}_1 \le a$ and $|w| = |z + v + \theta y| \le |z| + 2|y|$, we have $|\bar{u}_2 w| \le a(|z| + 2|y|)$. Next we apply the estimate (30) to $\bar{w}_2$ so $0 \le \bar{w}_2 \le \omega \in L^r(0,T)$ so $|u\bar{w}| \le |y|\omega(t)$. Combining gives

$$
(33)\qquad |\eta(t)| \le \hat{\omega}(t)\,(|y| + |z|) \qquad \text{with } \hat{\omega} = [c + \omega] \in L^r(0,T)
$$

and then, using (11),

$$
(34)\qquad |\zeta(t)| \le c|\nabla y| + 2\hat{\omega}(t)\,(|y| + |z|).
$$

Using y, z as test functions in the weak forms of (26), (27), respectively, and adding the results gives

$$
\tfrac{1}{2}(\|\hat{y}\|^2 + \|\hat{z}\|^2)_t + \|\nabla\hat{z}\|^2 + \|\nabla\hat{z}\|^2 = \int_\Omega (y\eta + z\zeta).
$$

Using (33) and (34), we obtain

$$
\int_\Omega (y\eta + z\zeta) \le \tfrac{1}{2}\|\nabla\hat{z}\|^2 + \phi(t)(\|\hat{y}\|^2 + \|\hat{z}\|^2)
$$

with $\phi(\cdot)$ integrable. Since there is no inhomogeneous term, a standard application of Gronwall's Inequality shows that $\|\hat{y}\|^2 + \|\hat{z}\|^2 \equiv 0$. Thus, $y_1 = y_2$ and $z_1 = z_2$ and these solutions were not distinct. Of course, the measure $\bar{q}$ will then also be uniquely determined. $\square$

6. Comments on simulation

Theorem 4.2 together with Theorem 5.1 show, at least for the physically meaningful dimensions ($d = 1, 2, 3$), that one has strong convergence to well-defined functions, $\bar{u}, \bar{v}, \bar{w}$. The argument presented here through the auxiliary functions $y = \bar{u} - \bar{v}$ and $z = \bar{w} + \theta\bar{u} + (1-\theta)\bar{v}$ does give some information about the regularity of these functions. In particular, the interface (support of the measure $\bar{q}$) is just the zero-set of y and, as noted in [16], this has a regularity consistent with the classical treatment of the free boundary problem. Nevertheless, much remains uncertain about the structure and development of this interface. We also note that the compactness argument used here gives no information as to the rate of convergence, comparable to what is available in [12] for the 1-dimensional problem at steady state. Finally, we have not considered at all the question (Q5) of the Introduction regarding behavior as $t \to \infty$. Apart from the steady state analyses of [17] and [12], most of our information about these open issues comes from numerical simulation as in [18] and [4].

Before we turn to (Q4), consider an example from [18] of the interface evolution in 1-D with $\Gamma_A = \{0\}, \Gamma_B = \{1\}$. This computation was done using $\lambda = 10^9$ in the original system (2)–(4).

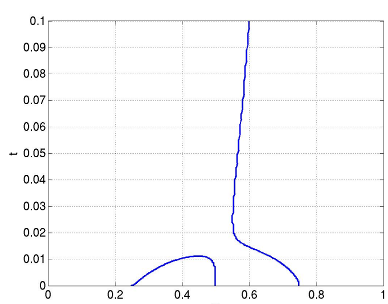

Figure 1. Evolution in t of the interface.

For this example, the initial data was consistent ($uv \equiv 0$), with isolated pockets of A, B so initially three interfaces. We then see the left- and rightmost interfaces moving toward the center as these pockets shrink with the pocket of B consumed first, after which the remaining interfacial point moves more slowly towards its steady state value. This picture is essentially independent of λ, even for fairly moderate values.

If one were to look at q^λ, the simulations for this and other values of λ show profiles (for fixed t) spatially of the same form as given theoretically by the singular perturbation analysis of [12] for steady state: scaling as $\lambda^{1/3}$ in height and as $\lambda^{-1/3}$ in width, so converging in $[C(0,1)]^*$ to a delta function as $\lambda \to \infty$. [This description applies only to isolated interfaces and, of course, cannot apply to the behavior near the moment t_* when the pocket of B vanishes with its left and right boundaries coming together.]

Similar computations in the 2-D setting show that the graph of q^λ taken transversally to a smooth interface curve also shows exactly the same profile and an informal scaling argument, noting that the time derivatives drop out to first order, shows that this is to be expected. [Of course, in the two dimensional setting we may have more complicated topological changes with, e.g., pockets of A or B being consumed or pinching off from a larger region or connecting to one by an isthmus — all of these are exhibited in the examples of [4] — while in 3-D a toroidal pocket might lose its hole or vice-versa, etc.]

Returning to (Q4), we consider how this behavior affects computation. If one applies standard approaches directly to the system (2)–(4), the primary difficulty is the reaction zone: adequately resolving the term q^λ whose local integral is essential in determining the production of C and so the production rate for the product. With the effective width of the q^λ term $\mathcal{O}(\lambda^{1/3})$, this means using a very fine mesh in that region where u, v are each small and proportionally changing rapidly: either one must use a very fine mesh everywhere, which is feasible but expensive in one space dimension and prohibitive for higher dimensions, or one must modify the approach by introducing additional machinery to track the location of this interface.

The free boundary approach to the modeling involves this kind of tracking essentially — with the necessity of resolving q^λ replaced by the necessity of introducing an additional differential equation for the motion of the interface (whether derived by singular perturbation analysis or otherwise, (cf., e.g., [15] or [16]). This, of course, would be intended to approximate the limit solution $\bar{u}, \bar{v}, \bar{w}$ with the understanding that for large λ this is itself a good approximation to the "true" solution $u^\lambda, v^\lambda, w^\lambda$.

As an alternative, we note that we might compute $\bar{u}, \bar{v}, \bar{w}$ through the auxiliary system (26), (27) used here for theoretical purposes in

Section 5 or through the auxiliary system

$$
\begin{aligned}
y_t &= \Delta y - y_+ \hat{z} \\
y &= \begin{cases} \alpha & \text{on } [0,T]\times\Gamma_A \\ -\beta & \text{on } [0,T]\times\Gamma_B \end{cases} \\
y_\nu &= 0 \qquad \text{on } [0,T]\times[\partial\Omega\setminus(\Gamma_A\cup\Gamma_B)], \\
y &= u_0 - v_0 \qquad \text{at } \{t=0\}\times\bar{\Omega},
\end{aligned}
\tag{35}
$$

$$
\begin{aligned}
\hat{z}_t &= \Delta \hat{z} - y_+ \hat{z} \\
\hat{z}_\nu &= \begin{cases} -y_\nu & \text{on } [0,T]\times\Gamma_B \\ 0 & \text{on } [0,T]\times[\partial\Omega\setminus\Gamma_B], \end{cases} \\
\hat{z} &= w_0 + v_0 \qquad \text{at } \{t=0\}\times\bar{\Omega},
\end{aligned}
\tag{36}
$$

for $y = u - v$, $\hat{z} = w + v$ used in [16]. Either of these systems has the considerable advantage of not at all involving $\bar{q}$ — here an unknown measure on Q_T — so the right hand sides are quite well-behaved and no front-tracking is required; we note that these seem to involve only gradient discontinuities coming from the uw term. The system (35), (36) was used computationally in [4] and, especially for higher dimensional problems, is orders of magnitude more efficient than the comparable computation working with (2)—(4) on a uniformly fine mesh. [One does note that the boundary condition coupling in (36) is nonstandard and not all computational packages will handle this. This problem does not arise in connection with (26), (27) (although one has the gradient coupling in (27) and must precompute $\nabla\theta$), but this has not yet been tried computationally.]

7. Appendix: proof of Theorem 4.1

Having defined $\mathcal{X}_p = L^p([0,T] \to X)$ for $1 \le p \le \infty$, we wish to prove here the result:

Theorem 4.1 *Let $S(\cdot)$ be a C_0 semigroup on X with infinitesimal generator $-A$; assume $S(\tau)$ is compact for each $\tau > 0$. Then the solution map $L : g \mapsto u$ of the differential equation $u_t + Au = g$ on $[0,T]$ with $u(0) = 0$, given by*

$$
u(t) := \int_0^t S(t-s)\, g(s)\, ds \qquad \text{for } t \in [0,T],\ g \in \mathcal{X}_1,
\tag{37}
$$

is a well-defined compact operator: $\mathcal{X}_1 \to \mathcal{X}_p$ for arbitrary $1 \le p < \infty$.

Proof. It is sufficient to show that the set $\{Lg : \|g\|_1 \le 1\}$ is totally bounded in $\mathcal{X}_p$, i.e., has a finite cover of $\varepsilon-$balls for each $\varepsilon > 0$. Let $K > 0$ bound $\|S(t)\|$ for $t \in [0,T]$ and choose $\tau := T/M$ with M large enough that $3K\tau^{1/p} < \varepsilon/3$ Note that $\mathcal{K} = \{S(\tau)x : \|x\|_X \le 1\}$ is precompact by assumption so the C_0 condition $S(t)x \to x$ implies uniform convergence on $\mathcal{K}$, i.e., for $\varepsilon > 0$ there is $\delta = \delta(\varepsilon) > 0$ (depending on τ) such that

$$s \leq \delta(\varepsilon) \;\Rightarrow\; |S(s)y - y| \leq \frac{\varepsilon}{3K}|x| \text{ for } y = S(\tau)x. \tag{38}$$

We then choose $N \geq M$ large enough that $\sigma = T/N \leq \delta$.

For $\tau \leq t \leq T$ we will set $n = \lfloor (t-\tau)/\sigma \rfloor$ (so $0 \leq t - n\sigma - \tau < \sigma$) and can then define

$$v(t) = S(\tau) \int_0^{n\sigma} S(n\sigma - s) g(s)\, ds = S(\tau) u(n\sigma) =: v_n$$

for $t \geq \tau$ with $v(t) = 0$ for $0 \leq t < \tau$. As $|u(t)| \leq K$ we have $v_n \in K\mathcal{K}$, which is precompact so we can find a finite set $\{y_j : j = 1, \dots, J\}$ of centers of $(\varepsilon/3)$-balls covering $K\mathcal{K}$. For each $v(\cdot)$ as here we can choose from this set so $|v(t) - y_{j(n)}| < \varepsilon/3$ for $t \in [n\sigma + \tau,\, (n+1)\sigma + \tau$ whence

$$\|v - y\|_\infty < \varepsilon/3 \tag{39}$$

for this piecewise constant function y with $y(t) = y_{j(n)}$. There are just J^N such functions which will be the centers of the desired ε-balls in $\mathcal{X}_p$ covering $\{Lg : \|g\|_1 \leq 1\}$.

It remains only to estimate $u - v$ for which a bit of manipulation gives

$$u(t) - v(t) = \int_{n\sigma}^{t} S(t-s) g(s)\, ds + \left[S(t - n\sigma - \tau) - 1\right] S(\tau) u(n\sigma).$$

We estimate separately these terms e_1 and e_2. First note that (38) gives

$$|e_2(t)| \;\leq\; \frac{\varepsilon}{3K}\, |u(n\sigma)| \;\leq\; \frac{\varepsilon}{3} \qquad \text{so} \quad \|e_2\|_p \;\leq\; \frac{\varepsilon}{3}. \tag{40}$$

We then see that

$$\begin{aligned}
\|e_1\|_p \;&\leq\; K \left[\int_0^T \left(\int_{n\sigma - \tau}^{t} |g(s)|\, ds \right)^p dt \right]^{1/p} \\
&\leq\; K \left[\sum_{m=1}^{M} \int_{(m-1)\tau}^{m\tau} \left(\int_{n\sigma-\tau}^{t} |g(s)|\, ds \right)^p dt \right]^{1/p} \\
&\leq\; K \left[\sum_{m=1}^{M} \int_{(m-1)\tau}^{m\tau} \left(\int_{(m-2)\tau}^{m\tau} |g(s)|\, ds \right)^p dt \right]^{1/p} \\
&=\; K \left[\sum_{m=1}^{M} \tau \left(\int_{(m-3)\tau}^{m\tau} |g(s)|_X\, ds \right)^p dt \right]^{1/p} \\
&\leq\; 3K\tau^{1/p} \int_{-2\tau}^{M\tau} |g(s)|\, ds
\end{aligned}$$

so $\|e_1\|_p \le 3K\tau^{1/p} \le \varepsilon/3$ for $\|g\|_1 \le 1$. Combining gives

$$\|u - y\|_p \le \frac{\varepsilon}{3} + \frac{\varepsilon}{3} + \frac{\varepsilon}{3} = \varepsilon$$

so each $u \in \{Lg : \|g\|_1 \le 1\}$ is within ε (in $\mathcal{X}_p = L^p([0,T] \to X)$) of one of the N^J functions y. Thus this set is totally bounded (precompact) so L is a compact operator to $\mathcal{X}_p$ as asserted. □

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