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INTERFACE CONDITIONS FOR A SINGULAR REACTION-DIFFUSION SYSTEM

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Abstract. For a chemical reaction/diffusion system, a very fast reaction $A + B \to C$ implies non-coexistence of A, B with resulting interfaces. We try to understand how these interfaces evolve in time. In particular, we seek a characterizing system of equations and conditions for the sharp interface limit: when this fast reaction is taken as infinitely fast.

1. **Introduction.** From the viewpoint of chemical engineering, our objective here is to understand the diffusion controlled reaction $2A + B \to$'product' when the reaction path consists of a coupled pair of irreversible simple reactions

$$A + B \xrightarrow{\lambda} C, \qquad A + C \xrightarrow{\mu} P$$

i.e., A combines simply with B (at rate λ) to form an intermediate compound C which will then combine with A (at rate μ) to form P. The resulting system of partial differential equations in a 'film' (cf., e.g., [2, 6]) was treated as a model problem in [3] with quite different concerns and with quite different boundary conditions relating to a context of bubble reactors. We are here concerned with treatment of the case $\lambda \gg \mu$ so our consideration of the system involves two asymptotic limits — as $\lambda \to \infty$ and as $t \to \infty$. Our principal objective here is to examine the behavior of this problem in the fast reaction limit $\lambda = \infty$ — although we do not entirely forget here the other presumptive (long time) limit, a steady state at $t = \infty$.

Standard chemical kinetics give a system of partial differential equations for the concentrations u, v, w of A, B, C respectively:

$$\begin{aligned} u_t &= u_{ss} - \lambda uv - \mu uw \\ v_t &= v_{ss} - \lambda uv \\ w_t &= w_{ss} + \lambda uv - \mu uw \end{aligned} \tag{1}$$

[We may write $U^\lambda = \left[u^\lambda, v^\lambda, w^\lambda\right]$ (similarly q^λ for the expression λuv) to emphasize the dependence on the Thiele modulus λ, but normally will suppress this superscript.] In considering this problem in the limit, it is impossible to set $\lambda = \infty$ directly in (1) to characterize the limit solution. A characterization was obtained [8, 4] for the steady state problem (see (8), (9) in the next section) and our primary objective here, hypothesizing the existence of such a limit, is to obtain a

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corresponding characterization for the time dependent problem, noting that the interface conditions are the major missing ingredient in this.

[Assuming the same diffusion coefficient for all components (but see Remark 3), there is no loss of generality in choosing time/space units so this is 1 and the spatial interval is $0 \le s \le 1$ in (1); by also scaling u, v, w, we also can — and henceforth do — take $\mu = 1$.]

Here we study this system with the boundary conditions

$$\left\{\begin{array}{l} u = \alpha \\ v_s = 0 \\ w_s = 0 \end{array}\right. \quad \text{at } s = 0, \qquad \left\{\begin{array}{l} u_s = 0 \\ v = \beta \\ w_s = 0 \end{array}\right. \quad \text{at } s = 1 \tag{2}$$

— i.e., there is no material flux at the boundaries except that reservoirs of A, B, respectively, at the two sides of the region maintain the respective concentrations at the given levels $\alpha, \beta > 0$. We adjoin to this the initial conditions

$$u(0,\cdot) = u_0(\cdot), \qquad v(0,\cdot) = v_0(\cdot), \qquad w(0,\cdot) = w_0(\cdot) \tag{3}$$

so the complete system is (1), (2), (3). From the interpretation as chemistry it is clear that the concentrations u, v, w should remain non-negative and, as expected, the mathematics follows this.

Theorem 1.1. *Let $\lambda, \alpha, \beta > 0$ be given and assume u_0, v_0, w_0 on $(0,1)$ are continuous and non-negative. Then the system (1), (2), (3) has a unique global solution $U = U^\lambda$ in*

$$\mathcal{U} = \left\{ (u,v,w) \in C([0,\infty) \to [H^1(0,1)]^3) : \begin{array}{l} 0 \le u \le \alpha' \\ 0 \le v \le \beta' \\ 0 \le w \le \gamma_0 + \lambda\alpha'\beta' t \end{array} \right\}$$

where $\alpha' = \max\{\alpha, u_0(\cdot)\}$, $\beta' = \max\{\beta, v_0(\cdot)\}$, $\gamma_0 = \max\{w_0(\cdot)\}$.

Proof. The techniques are reasonably standard so we need only sketch the proof here.

For $\bar{U} = (\bar{u}, \bar{v}, \bar{w}) \in \mathcal{U}$, define $\mathcal{F}(\bar{U})$ as the solution $U = (u, v, w)$ of the linear decoupled system

$$\begin{array}{rl} u_t &= u_{ss} - [\lambda\bar{v} + \bar{w}]\, u \\ v_t &= v_{ss} - \lambda\bar{u}\, v \\ w_t &= w_{ss} - \bar{u}\, w + \lambda\bar{u}\bar{v} \end{array} \tag{4}$$

with (2) and (3). We show that this $\mathcal{F}$ maps $\mathcal{U}$ to itself and then that it is, in a suitable sense, a contraction and so has a unique fixpoint in $\mathcal{U}$.

Arguing from the weak Maximum Principle, one shows first the non-negativity and then the pointwise upper inequalities. For example, after having first shown $0 \le u \le \alpha'$, $0 \le v \le \beta'$ and $w \ge 0$, we set $\omega(t) = \gamma_0 + \lambda\alpha'\beta' t$ and then take $z = [w - \omega]_+ = \max\{0, w - \omega\}$ as test function in the weak form of the third equation of (4). Note, e.g., that $z_t = w_t - \lambda\alpha'\beta'$ where $z \ne 0$ so $z(w-\omega)_t = z z_t$ and similarly $z_s w_s = [z_s]^2$ (cf., e.g., [9]). Thus we see that

$$(\tfrac{1}{2}\|z\|^2)_t + \|z_s\|^2 = z w_s \,|_0^1 + \int_0^1 [\lambda(uv - \alpha'\beta') - uw] z \le 0$$

whence, integrating, we see that $z \equiv 0$ so $w \le \omega$. With these bounds, taking u, v, w as test functions and applying the Gronwall Inequality one bounds $\|U(t,\cdot)\|$ pointwise in t. Similarly, one estimates differences: noting that the r.h.s. of (4) satisfies a Lipschitz condition (uniform for bounded λ, t), one takes the differences as test functions and the Gronwall Inequality now gives an estimate which corresponds to

contraction in an exponentially weighted sup norm — whence the iteration sequence converges in this norm to the fixpoint of $\mathcal{F}$, which is then the desired solution of (1). [We have shown this convergence only for a bounded interval, but that interval is arbitrary so one obtains a global solution as desired.] □

As noted above, we are interested in the situation $\lambda \gg 1$. For this paper, we hypothesize the existence of a limit solution as $\lambda \to \infty$ and seek to understand it. Following a standard procedure, we can divide each equation of (1) by the large parameter λ and formally make the replacement $1/\lambda \leftarrow\!\shortmid 0$. Unfortunately, what remains is only the condition

$$uv \equiv 0 \qquad \text{when } \lambda = \infty. \tag{5}$$

This is not very informative in itself and is certainly inadequate to characterize the limit solution.

Remark 1. We do note that (5) is physically reasonable: A and B cannot really co-exist when $\lambda \gg 1$, as they then react with each other (almost) immediately. Mathematically, if we were to have $u(\bar{t}, \bar{s}) = a > 0$ and also $v(\bar{t}, \bar{s}) = b > 0$, then we could make the singularly scaled change of variables $\tau = \lambda(t - \bar{t})$, $\sigma = \sqrt{\lambda}(s - \bar{s})$ and divide by λ. Formally making the replacement $0 \leftarrow\!\shortmid 1/\lambda$ in the resulting system — i.e., dropping the terms from the slower reaction and from diffusion as negligible on this scale, we then obtain the ODEs

$$u' = v' = -uv \qquad u(0) = a, v(0) = b$$

with $'$ now denoting $d/d\tau$. Assuming $a > b > 0$, we note that $u - v \equiv 0$ with $u, v > 0$ so $v' \le (a - b)v$ and $v(t) \le e^{-(a-b)\tau}$, converging exponentially in τ to 0 with $u \to (a - b)$ within a transient of duration $\mathcal{O}(1/\lambda)$. If $a = b > 0$, we would have $u = v = 1/[\tau + a^{-1}] \to 0$.

Without assuming for (3) that the condition (5) holds at $\bar{t} = 0$, we must expect a transient layer of duration $\mathcal{O}(1/\lambda)$ — the short time scale — within which the data will adjust to satisfy that condition. On the other hand, this scaling argument shows that such a situation can never develop at a later time so we will have (5) for $t > 0$. □

Henceforth we begin our analysis *after* the initial transient of Remark 1, and so will require that our initial data satisfies the consistency conditions:

$$\begin{gathered} u_0\, v_0 \equiv 0 \qquad u_0(0) = \alpha > 0 \qquad v_0(1) = \beta > 0 \\ \text{with} \qquad u_0,\ v_0, w_0 \text{ continuous and non-negative.} \end{gathered} \tag{6}$$

Such data will then consist of an alternation of regions of 'type A' ($u > 0$, $v \equiv 0$) and 'type B' ($u \equiv 0$, $v > 0$). While we have not actually shown that this is really a consequence (shortly after any initial transient) of the observations above, we will view (6) as including the assumption that that we are here partitioning $[0, 1]$ into a finite number of such regions. Obviously, in view of the boundary conditions, we have type A at $s = 0$ and type B at $s = 1$; physically, the interior regions are isolated pockets of the respective components remaining after the initial transient.

We then refer to the points where two such regions abut as *interfaces*. For finite $\lambda \gg 1$ these are more properly narrow 'reaction zones'. Physically, A and B, respectively, will be transported (within each of the abutting regions) to this zone where they react very rapidly. Even when the reaction is viewed as instantaneous ($\lambda = \infty$), we note that the reaction $A + B \to C$ actually occurs at a rate essentially determined by the rate of diffusive transport to the interfaces — indeed, it is this

which leads us to anticipate the existence of a limit solution in which the reaction zones have become sharp interfaces. The primary emphasis of this paper is to initiate an understanding of the behavior of these interfaces. We restrict our attention to one space dimension and point interfaces moving in time.

In the next section we review briefly what is already known about this problem — partly from computation and partly from theoretical analysis of the steady state problem — and our resulting expectations. The third section presents a reformulation of the system, noting some results, and the fourth section concludes with some comments and some further conjectures.

2. **What is known already? What do we expect?** Much of what we know already about this problem comes from the analysis of the steady state problem in [8, 4]; the computational treatment in [7, 5] of the time dependent problem we consider here was then largely directed to seeing that the analogous behavior also occurred in this context. We begin here by summarizing some of the results of [8, 4].

Theorem 2.1. [8] *Fix $\alpha, \beta > 0$. For any λ, the steady state system*

$$u'' = \lambda uv + uw \qquad v'' = \lambda uv \qquad w'' = -\lambda uv + uw \tag{7}$$

with (2) has a solution U^λ. These solutions are bounded uniformly in λ and one has convergence as $\lambda \to \infty$ to a limit solution $U^\infty = (\bar u, \bar v, \bar w)$: for some $\bar s \in (0,1)$ one has

$$\begin{aligned} \bar u'' = \bar u \bar w = \bar w'', \ \bar v \equiv 0 \qquad & \text{on } (0, \bar s) \\ \bar u \equiv 0, \ \bar v(s) = \beta \frac{s - \bar s}{1 - \bar s}, \ \bar w \equiv \text{const.} \qquad & \text{on } (\bar s, 1). \end{aligned} \tag{8}$$

Further, this limit satisfies the balance conditions

$$-\bar u'(\bar s-) = \bar v'(\bar s+) = \bar w'(\bar s-) = -\frac{1}{2}\bar u'(0) = \kappa \tag{9}$$

with $\kappa = \beta/(1-\bar s)$; we have $q^\lambda = \lambda uv \to \kappa \delta_{\bar s}$ in $H^{-1}(0,1)$ as $\lambda \to \infty$. The system (8) and interfacial conditions (9) uniquely determine the limit solution.

A major technical difficulty for Theorem 2.1 is the bound on w for the existence argument. Since we have imposed a no-flux boundary condition on w at both ends without limiting the influxes of A, B, this corresponds physically to the achievement of a balance between the rate at which the compound C is consumed by the slower reaction and the rate at which it is produced by the fast reaction — which, in turn, corresponds to the rate at which A, B are consumed by that, matching the incoming flux of B at the end $s = 1$ where its concentration is maintained (and also half the incoming flux of A at the end $s = 0$). While anticipating analogous behavior in the time dependent context, we must note that one cannot expect such a balance at the initial time so it would have to develop in the system evolution, to be achieved dynamically. [Indeed, if we were to omit the slower reaction, setting $\mu = 0$ in (1), then such a balance becomes impossible and, with the no-flux boundary condition, the concentration w of the compound C would grow without bound. Of course, a similar concern could also arise here if we were to include in our analysis the concentration of the product of the combined reaction $2A + B \to P$.] Certainly the estimate implicit in the definition of $\mathcal{U}$ in Theorem 1.1 is inadequate to show the development of such a balance as the pointwise bound there on w blows up for large λ, t. For the present, however, we defer this challenge to focus on the structure and behavior of the interfaces.

Theorem 2.2. [4] *Setting $\varepsilon := \lambda^{-1/3}$, one has a (formal) singular perturbation expansion giving convergence of $\varepsilon q^\lambda(\bar{s}+\varepsilon\sigma)$ to $\bar{q}(|\sigma|)$ as $\lambda \to \infty, \varepsilon \to 0$ with $\bar{q}$ a scaled version of the Airy function $Ai(\sigma)$. Further, one has an explicit estimate $u(\hat{s}) = \mathcal{O}(\varepsilon)$ where $\hat{s}$ is taken so $u(\hat{s}) = v(\hat{s})$; using the Implicit Function Theorem set in $\mathcal{Y} = W^{-1,1}(0,1)$, the dual space of the Lipschitzian functions $C^{0,1}[0,1]$, one then obtains $\mathcal{O}(\varepsilon)$ estimates for $\|q^\lambda - \kappa\delta_{\bar{s}}\|_{\mathcal{Y}}$ and, uniformly on $[0,1]$, for $|U^\lambda - U^\infty|$.*

Corollary 1. *For large enough λ the steady state solution of Theorem 2.1 is unique.*

Proof. This is an immediate consequence of the IFT argument of Theorem 2.2. □

The analysis underlying Theorem 2.2 is very much based on the known structure of the 'reduced' (limit) solution as obtained in Theorem 2.1, precisely what we are trying to accomplish here for the time dependent setting. In particular, any singular perturbation expansion at the interface requires knowledge that at steady state the interface is a single (uniquely determined) point $\bar{s}$. The major contribution of Theorem 2.2 is the determination that the correct asymptotics in such an expansion is to be given in terms of $\lambda^{-1/3} =: \varepsilon$. For the time dependent setting we no longer can assume a unique interface: our discussion following (6) shows that we must expect, certainly at least initially, a multiplicity $\{\bar{s}_j\}$ of interfacial points separating alternate type A and type B regions. Somehow these must reduce to the single interface of the steady state limit solution as $t \to \infty$ if we are to have convergence to a steady state. We would then expect that for large finite λ one would have a set of narrow reaction zones, each of width $\mathcal{O}(\varepsilon)$ and height $\mathcal{O}(1/\varepsilon)$, with $q^\lambda = \lambda uv$ a sum of terms with the same fine structure $\varepsilon q^\lambda(\bar{s}_j + \varepsilon\sigma) \approx \bar{q}(|\sigma|)$ as at steady state.

For confirmation of these expectations we will first turn to computation: from the computations of [7, 5] we provide two figures showing the evolution in t of the interface structure (where $u = v = 0$) for an elementary example. [Actual computation was done for (1) with $\lambda = 10^9$.] Note that Figures 1 and 2 show the same computation except for the time scales.

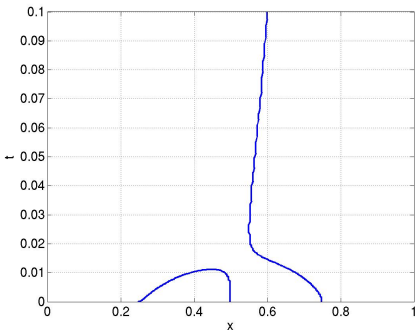

Fig.1: intermediate time scale

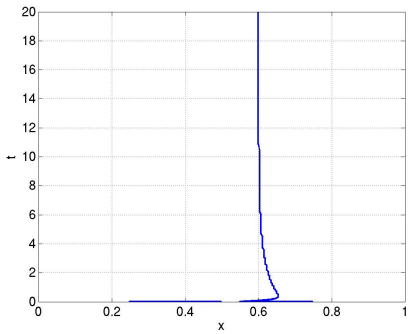

Fig.2: long time scale

In these figures we are considering a typical example beginning (after any short $\mathcal{O}(1/\lambda)$ time scale initial transient) with data consisting of four alternating regions of types A, B, A, B so we have a pair of isolated pockets between the two regions fed by the external sources at $s = 0, 1$. For each of these pockets the respective concentration is positive, vanishing at the ends of the subinterval with the fluxes there determining the effective rate of the fast reaction. Physically, the component

retained in each pocket will be consumed. If, as we expect, this happens in finite time — i.e., on what we might refer to as the *intermediate time scale* — then that interval vanishes and the number of interfacial points is thus reduced by 2. For the A, B, A, B example shown above one is starting initially with 3 interfaces so this reduction results in a single interfacial point separating the permanently maintained end regions; if there is to be any convergence to steady state, this single point must move — on the *long time scale* — to its position in the steady state solution of Theorem 2.1.

These figures confirm the expectations described above for the interfacial evolution. At the bottom of Fig. 1 we see the initial interfaces for A, B, A, B regions. As we move up ($t \nearrow$) the two internal regions (isolated pockets of B, A concentration) are both shrinking with their common interface roughly constant at $s = .5$. For the particular initial conditions, it is the left pocket which disappears first (roughly, at $t \approx .01$), so the remaining right pocket is then connected with the externally supported type A region on the left. The single remaining interface continues to move toward the left until about $t \approx .025$, then moves toward the right. Taking the picture up now from Fig. 2, we see the interface continuing rightward until about $t \approx .05$ and then again moving left, increasingly slowly, toward its position $\bar{s} \approx .6$ for the steady state solution. It is this interface motion on which we are focusing our attention.

The computations of [7, 5] also appear to confirm convergence of U^λ as $\lambda \to \infty$ for the time dependent problem: the computed results for $\lambda = 10^6$ and for $\lambda = 10^9$ remain essentially the same — except that the fine structure scaling for the computed q^λ, defining the width of the reaction zone, behaves as we had expected from the steady state analysis. [Note that this is computationally intensive: one must discretize here so as to resolve rapid changes over spatial scales of order $\varepsilon = .01$ or .001, e.g., in computing q^λ.] We further note that the computed solutions are essentially also independent of t for only moderately sized t — with U_t negligible so we are already matching the steady state characterization (8) quite closely by, e.g., $t = 20$.

3. **A related system.** It is clear that the terms $q^\lambda = \lambda uv$ are causing much of the difficulty when λ becomes very large. Assuming the behavior for the time dependent problem corresponds to that known for the steady state version, we would have $\mathcal{O}(\lambda^{1/3})$ growth of $\max\{q^\lambda\}$ as $\lambda \to \infty$, although in a space of distributions q^λ remains bounded and converges to a δ-function. However, we do not, as yet, have any reasonable characterization here for the limiting interface structure for the time dependent evolution; even for the steady state problem, the characterization in [8] of $\bar{s}$ is rather indirect, strongly dependent on the balance conditions (9) which are unavailable here.

In looking at evolution of the interfaces, we note that these are now the zero-set of the difference $y = u - v$ and we note that the occurrences of q^λ cancel out in considering this. We may also set $z = w + v$ and again the occurrences of q^λ cancel out. Initial data for these are immediately available from (3) and we translate the boundary conditions (2) to obtain

$$\begin{array}{lllll} & \text{on } (0,1) & \text{at } 0 & \text{at } 1 & \\ y_t & = y_{ss} - \phi & y = \alpha - v(t,0) & y = -\beta + u(t,1) & (10) \\ z_t & = z_{ss} - \phi & z_s = 0 & z_s = -y_s & \end{array}$$

with $\phi = uw$. This is true, but seems rather useless as it is not self-contained: we know neither the nonlinearity $\phi = uw$ which appears here nor the boundary data $v(t,0)$ and $u(t,1)$.

Somewhat paradoxically, perhaps, it is precisely in the limit case $\lambda = \infty$ (where we cannot even write (1)) that (5) becomes available, enabling us to handle this difficulty. We first note that, since $u = \alpha > 0$ at $s = 0$, (5) gives $v(t,0) \equiv 0$ and, similarly, $u(t,1) \equiv 0$ so the boundary data is now self-containedly available. We next note that, with (5), $w = z - v$ gives $\phi = uw = uz - uv = uz$ — still not quite self-contained. However, Theorem 1.1 gives $u, v, w \geq 0$ so this should also hold in the limit. This, together with (5), enables us to recover u, v from $y = u - v$: pointwise, one must have

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

and we then have $\phi = uz = y_+ z$. [Equivalently, since also $v, w \geq 0$ so $z \geq 0$, we could write instead $\phi = y_+ z_+$.]

Given (5), then, the system above is now self-contained:

$$\begin{array}{lccc} & \text{on } (0,1) & \text{at } 0 & \text{at } 1 \\ y_t & = y_{ss} - z y_+ & y = \alpha & y = -\beta \\ z_t & = z_{ss} - y_+ z & z_s = 0 & z_s = -y_s \end{array} \tag{11}$$

[Using $\phi = y_+ z_+$ would not give a system universally equivalent to (11) as stated above, but one might reasonably hope that they would give the same results for the relevant class of initial data consistent with (6).] We emphasize here that the system (11) is related to (1), (2) only at the limit $\lambda = \infty$, since we have no useful self-contained determinations of ϕ or of the boundary data when λ is finite.

While we have seen that the limit solution of (1), (2), (3) provides a solution of (11), we now show that, at least for the steady state of Theorem 2.1, the converse also holds.

Theorem 3.1. *There exists a unique steady state solution (y, z) of the system (11) with $z \geq 0$.*

Proof. We know from Theorem 2.1 that there is a steady state limit solution ($\lambda = \infty$) satisfying of (8), (9) and taking $y = u - v, z = w + v$, as above, from this gives a steady state solution (y, z) of the system (11).

Conversely, if (y, z) is a steady state solution of (11), we may partition $[0, 1]$ into subintervals (regions $[\bar{s}_j, \bar{s}_{j+1}]$) of type A, B. On each interior subinterval we have $y(\bar{s}_j) = 0 = y(\bar{s}_{j+1})$ and either $-y'' + zy = 0$ (with $z \geq 0$) or $y'' = 0$, depending on the type of the subinterval. Either way, one gets $y \equiv 0$ on each of these interior subintervals so $\phi = y_+ z \equiv 0$. On the union of these together with the 'permanent' type B region on the right — i.e., on $[\bar{s}_1, 1]$ — one thus has $y'' = 0$ and $z'' = 0$ with $y(\bar{s}_1) = 0, y(1) = -\beta$. [Note that this gives $y < 0$ there — which ensures that there never were any interior pockets for this solution and we may simply write $\bar{s}$ rather than $\bar{s}_1$.] On the permanent type A region $[0, \bar{s}_1]$ on the left one has $-y'' = \phi = -z''$ with $y(0) = \alpha, y(\bar{s}_1) = 0$ so $y > 0$ there. Setting $u = y_+, v = -y_-$, and then $w = z - v$, we see that $u \equiv 0, v = -y$ on $[\bar{s}_1, 1]$ so $u'(1) = 0$ and $w'(1) = [z - v]'(1) = -y'(1) - v'(1) = 0$ so $w'' = z'' = 0$ gives $w' \equiv 0$ there. We also note that $-u'(\bar{s}-) = -y'(\bar{s}) = v(\bar{s}+) = v'(1) = \kappa$. On $[0, \bar{s}_1]$ we have $v \equiv 0$ so $v'(0) = 0$ and $w'(0) = [z - v]'(0) = 0$ and we also note that $w'(\bar{s}-) = z'(\bar{s}) = v'(\bar{s}+) = \kappa$. Finally, $w'' \equiv u''$ there so $u'(\bar{s}) - u'(0) = w'(\bar{s}) - w'(0) = \kappa$.

We have thus shown that a steady state solution (y,z) of (11) corresponds to (u,v,w) satisfying the characterization in Theorem 2.1 of the steady state limit solution. Since we know this is unique, we have the uniqueness of (y,z). □

Since the boundary conditions for (11) involve a somewhat unfamiliar coupling, some comment about this system seems in order. Consider first the pure initial value problem

$$\begin{array}{cc} y_t = y_{ss} & z_t = z_{ss} \\ y(t,0)=0,\ y(t,1)=0 & z_s(t,0)=0,\ [z-y]_s(t,1)=0 \\ y(0,s)=y_0(s) & z(0,s)=z_0(s) \end{array}$$

It is easy to see that this generates an analytic semigroup $S(\cdot)$ on, e.g., the space $\mathcal{X} = \{(y,z) : y,z \in C^{0,1}[0,1]$, with BC$\}$. [To see this, note that the y-equation decouples here and is standard, giving solutions analytic in t on the open right half plane (as well as analytic in $s \in (0,1)$ there, but certainly in $C^{0,1}$). It follows that $y_s(t,1)$ is similarly analytic in t and, if this is treated as an inhomogeneity for the z-equation, we see that the solution of that standard initial/boundary value problem is also analytic. Since causality is clear, we thus have an analytic semigroup on $\mathcal{X}$ for the coupled system.] Setting $Y=(y,z)$ and $\Phi(Y) = \text{diag}\{y_+ z, y_+ z\}$, the system (11) is equivalent to the integral equation (mild solution)

$$Y(t) = S(t)\,Y_0 - \int_0^t S(t-s)\,\Phi(Y(s))\,ds. \tag{12}$$

By the analyticity of the semigroup, we then have (cf., e.g., [1]) the spectrum determined growth condition: $\|S(t)\| \le Me^{\sigma t}$ where σ is the least upper bound of real parts of the spectrum of the infinitesimal generator. We note here that the infinitesimal generator G of $S(\cdot)$ is

$$G = \begin{pmatrix} \partial^2/\partial s^2 & 0 \\ 0 & \partial^2/\partial s^2 \end{pmatrix} \quad \text{with domain } \mathcal{D}(G) = \left\{(y,z) \in \left(C^{2,1}[0,1]\right)^2 : \text{BC}\right\}.$$

[Clearly, G would not be self-adjoint even if we were to consider it on $L^2(0,1)$ rather than on $\mathcal{X}$.] Since G has compact resolvent, its spectrum consists only of eigenvalues which, with a bit of care, may easily be computed. For an eigenvector, the first component satisfies $y'' = \mu y$ with $y(0) = 0 = y(1)$ so if $y \not\equiv 0$ one has $\mu = -(k\pi)^2$ $(k = 1,2,\ldots)$ with $y = c\sin(k\pi s)$. The second component then satisfies $z'' = \mu z$ with $z'(0) = 0$. With $\mu = -(k\pi)^2$ one has $z = \hat{c}\cos(k\pi s)$ whence $z'(1) = 0$ — which is inconsistent with the given coupling condition $z'(1) = -y'(1) = c(k\pi)\cos(k\pi)$ unless $c = 0, y \equiv 0$. The set of eigenvectors thus has the form: $\{E_k = (0,e_k) : k = 0,1,\ldots\}$ with $e_k = \cos(k\pi s)$ apart from scaling. [Of course, this set $\{E_k\}$ of eigenvectors — each with 0 first component — does not span the state space $\mathcal{X}$: we must adjoin the generalized eigenvectors $\tilde{E}_k = (\tilde{y}_k, \tilde{z}_k)$ satisfying $(\mu_k - G)^m \tilde{E}_k = 0$. Without calculating these, we do note that the first eigenvalue $\mu_0 = 0$ is simple: there is the eigenvector $E_0 = (0,e_0)$ with $e_0 \equiv 1$, but there is no generalized eigenvector $\tilde{E}_0$.] Thus, the spectrum of the generator G is $\{\mu_k = -k^2\pi^2 : k = 0,1,\ldots\}$, giving $\sigma = 0$, so $S(\cdot)$ is uniformly bounded: there is a constant M such that $\|S(t)\| \le M$ for all $t \ge 0$. [Perhaps more relevant for asymptotics is the observation that if we obtain the restricted semigroup $S_0(\cdot)$ by projecting to the invariant complement of E_0 (i.e., span of other eigenspaces), then S_0 decays exponentially like $e^{-\pi^2 t}$ as $t \to \infty$.]

4. **Conclusions, conjectures, and further comments.** At this point we are ready to obtain the desired characterization of the (hypothesized) limit dynamics:

Theorem 4.1. *Let $\{U^\lambda : \lambda > 0\}$ satisfy the system (1), (2), (3) with fixed data satisfying the consistency conditions (6); assume one has convergence as $\lambda \to \infty$ to a limit $U = U^\infty = (u, v, w)$ satisfying (5). Then*

1. *The space-time domain $\mathcal{Q} = [0, \infty) \times [0, 1]$ is subdivided into regions of 'type A' and 'type B': where ($u > 0$, $v \equiv 0$) and ($u \equiv 0$, $v > 0$), respectively. These are separated by interfacial trajectories along which one has $u = v = 0$.*
2. *In the type A regions*

$$u_t = u_{ss} - uw, \qquad v \equiv 0, \qquad w_t = w_{ss} - uw.$$

In the type B regions

$$u \equiv 0 \qquad v_t = v_{ss}, \qquad w_t = w_{ss}.$$

3. *At each interface u, u_s, u_{ss} and v, v_s, v_{ss} are continuous up to the interface. Across the interfaces we have the balance conditions*

$$u = v = 0, \qquad u_s = -v_s \qquad u_{ss} = -v_{ss} \tag{13}$$

for suitable left- and right-hand limits.
4. *Where an interface is given is given locally by $s = \bar{s}(t)$, one has the differential equation for the interface motion*

$$\bar{s}' = \frac{d\bar{s}}{dt} = -\frac{u_{ss}}{u_s}\bigg|_{(t,\bar{s}(t))} = -\frac{v_{ss}}{v_s}\bigg|_{(t,\bar{s}(t))} \tag{14}$$

where the evaluations on the right are actually the "suitable left- and right-hand limits" of 3., above.
5. *As t increases, the number of interfacial points $N(t) = \#\{\bar{s}_j(t)\}$ is odd, non-increasing, and can decrease only by cancellation in pairs.*

These conditions, together with the initial conditions (3) uniquely characterize the limit solution U. [We note in passing that the limit steady state solution given by (7),(8) in Theorem 2.1 is of the form described here.]

Proof. We begin by noting that, as discussed above, $y = u - v, z = w + v$ will satisfy the system (11) with type A regions where $y > 0$, type B regions where $y < 0$ and interfaces given by the common boundary where $y = 0$. Obviously we have *1.,2.* above.

We next consider the regularity of solutions of (11). Note that y, z certainly evolve by (11)≡(12) as continuous functions so ϕ will also be continuous. This gives y, z in $C^2[0, 1]$; bootstrapping, we then have ϕ Lipschitzian and y, z in $C^{2,1}$. Since we have $u = y_+$ and $v = -y_-$, it follows that $y_s = u_s$ to one side of the interface (where $y > 0$ so $y = y_+ = u$) and $y_s = -v_s$ on the other side (where $y < 0$ so $y = y_- = -v$). The regularity above for y gives y_s continuous, hence continuous across the interface, so suitable left- and right-hand limits give the second condition in (13). The third condition is obtained similarly and we have *3.* at each interface.

Since we have the identity $y(t, s) \equiv 0$ along the interface by its definition, we may differentiate with respect to t to obtain, as a new identity, $y_t + y_s[d\bar{s}/dt] \equiv 0$. From (11) we have $y_t = y_{ss} - y_+ z$ but note that $y = 0$ here so $y_t = y_{ss}$ at the interface. So long as $y_s \neq 0$, this together with *3.*, will give the asserted differential equation (14) for the interface motion.

The assertion *5.* is largely a restatement of *1.* with the further point that the type A, B regions bounded by the interfaces may disappear, but cannot initiate. To see this we need only note that initiation of such a region — say, of type A with $y > 0$ — would imply a prior type B region containing an interval $\mathcal{I}$ where $y < 0$ and develops an interior maximum ($y = 0$). Since y satisfies $y_t - y_{ss} = 0$ with strictly positive 'initial' and 'boundary' data on $\mathcal{I}$, this would violate the maximum principle there. Essentially the same argument shows that a type B region cannot initiate within a type A interval. A somewhat different argument shows that the interfacial set $\{y = 0\}$ cannot 'widen out' to contain an open space-time set. To see this, we consider such a set in conjunction with the adjacent type B region. We then have $y_t - y_{ss} = 0$ on the conjoined set so standard regularity theory for the heat equation gives real analyticity in s there for y — which is inconsistent with having $y \equiv 0$ on one part of that and $y > 0$ on an adjacent part.

To see the final assertion that these conditions uniquely determine the solution, we return to the formulation (11)≡(12). It was already noted that the solutions of that evolve as continuous functions and so remain bounded on any fixed time interval. In the presence of such a bound, the nonlinearity Φ of (12) is uniformly Lipschitzian and the standard argument gives uniqueness for this initial value problem and so uniqueness of $U = U^\infty = (u, v, w)$. □

Remark 2. We note here some conjectures which remain for future work.

1. Obviously, we conjecture convergence as $\lambda \to \infty$ to the limit solution characterized here. This convergence was hypothesized above and we note that what is primarily required to prove this is an improvement of the estimate earlier implicit in the definition of $\mathcal{U}$ in Theorem 1.1 so as, e.g., to bound $\int_0^1 w\, ds$ uniformly in λ on each $0 \le t \le T$. Given such a bound, one has a bound on ϕ with which one can easily obtain compactness from (10). One then gets subsequential convergence to a limit, necessarily satisfying (11), and the uniqueness asserted in Theorem 4.1 would complete the convergence argument.

   Closely related to all this, of course, is the extension to the time dependent problem of the singular perturbation analysis of [4]. We continue to expect asymptotics expressed in terms of $\lambda^{1/3}$ near the interfaces, except possibly at those points where a pocket disappears and the interfacial discontinuity shifts from spatial to temporal.
2. We conjecture that the interior pockets disappear in finite time, thus leaving a single interface; a somewhat stronger conjecture is that one will have $y_s < 0$ on $[0, 1]$ from some time on. This is supported by Fig. 1 and related computation. Certainly there is depletion of the content of such a pocket: if $[\bar{s}_j, \bar{s}_{j+1}]$ were an internal pocket of A we would necessarily have $u = 0, u_s \ge 0$ at $\bar{s}_j$ and $u = 0, u_s \le 0$ at $\bar{s}_{j+1}$ so

$$\begin{aligned}\frac{d}{dt}\int_{\bar{s}_j}^{\bar{s}_{j+1}} u(t,s)\, ds &= \int_{\bar{s}_j}^{\bar{s}_{j+1}} [u_{ss} - uw]\, ds + \bar{s}' u\Big|_{\bar{s}_j}^{\bar{s}_{j+1}} \\ &\le \int_{\bar{s}_j}^{\bar{s}_{j+1}} u_{ss}\, ds = u_s\Big|_{\bar{s}_j}^{\bar{s}_{j+1}} \le 0\end{aligned}$$

   with a similar argument to show depletion of any internal pocket of B.

   Meanwhile, we might look to the y-equation without this reaction to see what might be happening in that case. There we would be considering $y_t = y_{ss}$

with $y(t,0) = \alpha$, $y(t,1) = -\beta$ and (smooth) initial data 'with pockets', i.e., with several sign changes. In this decoupled setting it is clear that we have C^1-convergence as $t \to \infty$ to the steady state solution $y_*(s) = \alpha + (\alpha - \beta)s$ with $(y_*)_s \equiv -(\alpha + \beta) < 0$. The pockets then do, indeed, all disappear in finite time and, since it seems intuitively unlikely that the effect of the slower reaction $A + C \xrightarrow{\mu} P$ would be to slow down the pocket depletions, we may take this also as support for the conjecture. [Indeed, we might hope that this comparison might suggest an approach to a strong enough estimate for the pocket decay to prove the conjecture.]

3. Finally, we conjecture that, at least for initial data consistent with (6), the solution (y, z) of (11) converges as $t \to \infty$ to its (unique) steady state solution. From Theorem 3.1 it then would follow immediately that the limit solution $U = (u, v, w)$ given by Theorem 4.1 would also go to its steady state, given by Theorem 2.1.

   [Given this conjecture, one notes that linearized stability in the neighborhood of the known steady state solution fails if arbitrary perturbations are permitted. However, the appropriate context should permit only those perturbations which preserve the structure (6) — indeed, with a single interface point near the steady state $\bar{s}$. Such perturbations no longer form a linear subspace and this stability analysis remains unsettled.] □

Remark 3. In order to obtain the system (11) it is necessary to have a common diffusion coefficient for the components A, B, C, as we have assumed here, although physically this need not be the case, even as a good approximation. We note at this point that one can seek comparable results without (11) for the more general system

$$\begin{aligned} u_t &= a_1 u_{ss} - \lambda uv - uw \\ v_t &= a_2 v_{ss} - \lambda uv \\ w_t &= a_3 w_{ss} + \lambda uv - uw \end{aligned} \tag{15}$$

in which we no longer assume $a_1 = a_2 = a_3$ (then scaled to 1). [Indeed, the treatment in [8] of the steady state problem was taken in this more general setting.]

As before, we now consider the system in the (hypothesized) limit $\lambda = \infty$ and seek to characterize the limit solution $U^\infty = (u, v, w)$. The argument in Remark 1 justifying (5) immediately extends to this more general setting so the partitioning of $[0, 1]$ by interfaces remains as above. By definition we have $u = 0 = v$ as an identity at the interfaces.

From this point our argument becomes heuristic. Thus, in trying to generalize the interface conditions we will simply assume sufficient regularity for u, v, w within the regions to have these and suitable derivatives continuous up to the interface trajectories and may also omit the fast reaction term $q = \lambda uv$ within a type A region so even as we approach the interface we have $u_t = a_1 u_{ss} - uw$. Essentially as in the proof of Theorem 4.1, we then differentiate the identity $u \equiv 0$ on the interface with respect to t to get $u_t + u_s \bar{s}' = 0$ there. Taking the limit as we approach the interface we have $u_t = a_1 u_{ss} - uw \to a_1 u_{ss}$ and now conclude that

$$\bar{s}' = \frac{d\bar{s}}{dt} = -a_1 \frac{u_{ss}}{u_s}\bigg|_{(t,\bar{s}(t))}.$$

A similar argument in the adjacent type B region also gives

$$\bar{s}' = -a_2 \frac{v_{ss}}{v_s}\bigg|_{(t,\bar{s}(t))}$$

so (14) generalizes suitably to this case. □

Remark 4. We might also consider this problem in higher-dimensional settings as, to some extent, was already done in [8] for the steady state case. Following [8] we would then consider, for example,

$$\begin{aligned} u_t &= \Delta u - \lambda uv - uw \\ v_t &= \Delta v - \lambda uv \\ w_t &= \Delta w + \lambda uv - uw \end{aligned} \tag{16}$$

on a bounded region $\Omega \subset \mathbb{R}^m$. Assuming a pair of separated subsets $\Gamma_A, \Gamma_B \subset \partial\Omega$ we generalize (2) to

$$\begin{aligned} &u = \alpha \text{ on } \Gamma_A \qquad && u_\nu = 0 \text{ on } \partial\Omega \setminus \Gamma_A \\ &v = \beta \text{ on } \Gamma_B \qquad && v_\nu = 0 \text{ on } \partial\Omega \setminus \Gamma_B \\ & && w_\nu = 0 \text{ on } \partial\Omega \end{aligned} \tag{17}$$

where α, β are now positive functions on Γ_A, Γ_B, respectively; we also consider non-negative initial data u_0, v_0, w_0. Use of the weak maximum principle again gives $u, v, w \geq 0$ and an argument quite similar to that of Remark 1 applies equally well here to give (5) for the limit $\lambda \to \infty$. As in the 1-dimensional case, we thus have an approximate partition of Ω into (finitely many) regions with either $u > 0$ and $v \approx 0$ or $u \approx 0$ and $v > 0$ separated by narrow reaction zones where u, v are each small but $q = \lambda uv$ is large. Again we expect that in the limit $\lambda \to \infty$ these reaction zones become sharp interfaces — now curves ($m = 2$) or surfaces ($\dots$) whose regularity is as yet undetermined.

For practical purposes one might attempt computation for the system (16), (17) with a large value $\lambda \gg 1$, but immediately observe that this is a formidable task since one must again be able to resolve the narrow reaction zones: use of a uniform mesh, while possible for $m = 1$, requires prohibitively many points for $m > 1$. Alternative approaches would be the use of adaptive meshing or, once one would determine them, using the differential equations governing interface motion to compute the evolution of a limit solution, but these are almost equivalent in their demands for quite complicated interface tracking. [We note in passing that the partial differential equations for interface motion can be expected to be much more complicated than the ordinary differential equation (14). E.g., one might anticipate here a dependence on the mean curvature of the interface, since that is closely related to the volume/area relation for the diffusive transport.]

It is fortunate, then, that the related system (11) generalizes to handle our consideration of the system (16), (17): again assuming the diffusion coefficients are the same, we can make the same construction $y = u - v$, $z = w + v$ and again have cancellation of the difficult term $q = \lambda uv$ and, in the limit $\lambda = \infty$ for which (5) holds, we get the relations

$$u = y_+ \qquad v = -y_- \qquad w = z - y_- \qquad uv = y_+ z \tag{18}$$

and obtain the coupled but self-contained system of partial differential equations

$$y_t = \Delta y - z y_+ \qquad\qquad z_t = \Delta z - y_+ z. \tag{19}$$

Now define

$$\gamma = \begin{cases} \alpha & \text{on } \Gamma_A \\ -\beta & \text{on } \Gamma_B \end{cases} \qquad \text{on the disjoint union } \Gamma = \Gamma_A \cup \Gamma_B \subset \partial\Omega.$$

From (17) and (5) we have $u > 0$ near Γ_A so $v \equiv 0$ there and $v_\nu = 0$ on Γ_A; similarly, $u \equiv 0$ near Γ_B. It follows that (17) gives

$$\begin{aligned} y &= \gamma \text{ on } \Gamma & \qquad y_\nu &= 0 \quad \text{on } \partial\Omega \setminus \Gamma \\ z_\nu &= 0 \text{ on } \partial\Omega \setminus \Gamma_B & \qquad z_\nu &= -y_\nu \quad \text{on } \Gamma_B \end{aligned} \tag{20}$$

so, as with (11), the system (19), (20) is self-contained. We again have good regularity for solutions of (19) so numerical computation is fairly straightforward. One can then use (18) to obtain u, v, w at $\lambda = \infty$ — and the singular perturbation 'correction', if available, could then be used to obtain still better approximations to $u^\lambda, v^\lambda, w^\lambda$ and q^λ near the interface if desired. □

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