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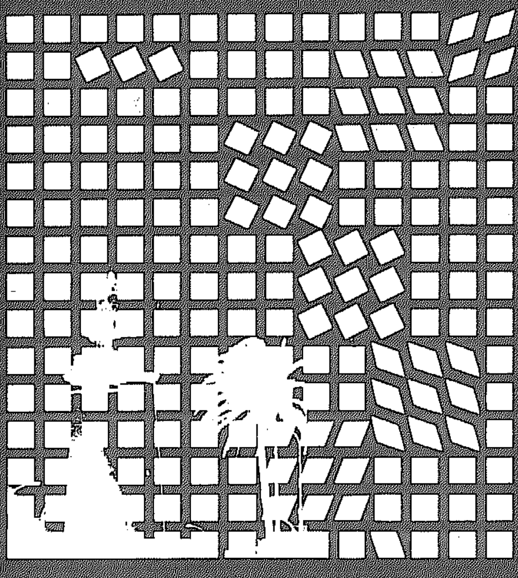

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MATHEMATICAL ANALYSIS OF THE CONVERSION OF A POROUS SOLID BY A DISTRIBUTED GAS REACTION

by

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1. Introduction. In this communication we present some of the results of a forthcoming article by the authors [11]. We consider the conversion (or combustion) of a porous solid, known as the *pellet*, as it reacts with a gas diffusing through its pores. Problems of this nature are of current interest in Chemical Engineering and Metallurgy. ([13],[21]). We assume that the reaction (which involves only one species of gas and one of the solid) is simple, irreversible and isothermal.

The gas, whose concentration is v(x,t) is replenished from the ambient region. The porous solid occupies the domain Ω (an open regular and bounded set of $\mathbb{R}^n$ with n=3 or n=2) and its concentration is u(x,t). Here t denotes the time and x a macroscopic position vector. The reaction is taken to be of the form

$$R(u,v)=f(u)g(v)$$

where f and g are real continuous monotone nondecreasing functions such that f(0)=g(0)=0. After nondimensionalization and under some suitable simplifications mass balances yield (see e.g.[13],[21])

$$P_\varepsilon \begin{cases} u_t = -f(u)g(v) & \text{in } \Omega\times(0,T)\equiv Q_T \\ \varepsilon v_t - \Delta v = -\lambda f(u)g(v) & \text{in } Q_T \\ \alpha \dfrac{\partial v}{\partial n} = 1-v & \text{on } \partial\Omega\times(0,T) \\ u(x,0) = u_0(x) & \text{on } \Omega \\ v(x,0) = v_0(x) & \text{on } \Omega \end{cases}$$

Here ε is the porosity (usually of the order 10^{-1} or 10^{-2}) and λ is the Thiele modulus ($1\le\lambda\le10^2$). The parameter α is assumed to be

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nonnegative ($\alpha\equiv 0$ corresponds to Dirichlet conditions). Some specially important choice of f and g are

$$f(u)=u^m \quad , \; g(v)=v^q \quad , \; m,q>0 \; , \tag{1}$$

where the case $m=2/3$ corresponds to the *Sohn-Szekely grain-pellet model* ([21]). For obvious reasons we assume $u_0\geq 0$ and $v_0\geq 0$ on Ω which, by the maximum principle, implies $u(x,t)\geq 0$ and $v(x,t)\geq 0$ on $\overline{\Omega}x[0,T]$. In section 2 we shall collect some results on the existence and uniqueness of solutions. Section 3 will be devoted to the "pseudo-steady-state problem" ($\varepsilon=0$) and, finally, in Section 4 we shall consider the solid conversion and gas penetration phenomena.

2. Existence, continuous dependence and uniqueness.

Given $(u_0,v_0)\in L^\infty(\Omega)xL^\infty(\Omega)$ satisfying

$$0\leq u_0(x)\leq 1 \quad , \quad 0\leq v_0(x)\leq 1 \quad , \quad \text{a.e.} x\in\Omega, \tag{2}$$

the existence of a weak solution

$$(u,v)\in \left[C([0,T]:L^\infty(\Omega))\cap W^{1,1}(0,T:L^2(\Omega))\right]xH^1(0,T:L^\infty(\Omega))$$

and such that

$$0\leq u(x,t)\leq 1 \quad , \quad 0\leq v(x,t)\leq 1 \quad , \quad \text{a.e.} \quad (x,t)\in\Omega \tag{3}$$

can be proved in several different ways: monotone iterations [11] (see also [15] and [14]) and compactness arguments [12]. It is not difficult to show that if, for instance, f and g are Holder continuous then (u,v) is a classical solution in Q. Nevertheless there are particular examples showing that $u\notin C^2_t$ and $v\notin C^3_x$.

The following result gives a continuous dependence leading, in particular, to the uniqueness of solutions (that improves the previous result of [15] related to the case when f and g are Lipschitz continuous).

Theorem 1. *Let* (u,v) *and* (u*,v*) *be any solutions corresponding to the initial data* (u_0,v_0), (u^*_0,v^*_0) *respectively. Then for any* $t\in[0,T]$ *we have*

$$\varepsilon\int_\Omega |v(x,t)-v^*(x,t)|dx+\lambda\int_\Omega |u(x,t)-u^*(x,t)|dx+$$
$$\frac{1}{\alpha}\int_0^t\int_{\partial\Omega}|v(\sigma,t)-v^*(\sigma,t)|d\tau d\sigma\leq\varepsilon\int_\Omega |v_0(x)-v^*_0(x)|dx+\lambda\int_\Omega |u_0(x)-u^*_0(x)|dx. \tag{4}$$

In particular, if $u_0=u^*_0$ *and* $v_0=v^*_0$ *we conclude that* $u=u^*$ *and* $v=v^*$.

Idea of the proof. Some manipulations lead to

$$(u-u^*)_t+(f(u)-f(u^*))g(v^*)=-f(u)(g(v)-g(v^*)) \tag{5}$$

and

$$\varepsilon(v-v^*)_t-\Delta(v-v^*)+\lambda f(u)(g(v)-g(v^*))=-\lambda g(v^*)(f(u)-f(u^*)). \tag{6}$$

By multiplying (5) by $\text{sign}_0(u-u^*)$ ($=1$ if $u-u^*>0, =0$ if $u-u^*=0$ and -1 otherwise) and by using property

$$\int_0^t\int_\Omega h_t \operatorname{sign}_0 h \, dx d\tau = \int_\Omega |h(t,x)|dx - \int_\Omega |h(0,x)|dx$$

which holds for any $h \in W^{1,1}(0,T:L^1(\Omega))$ (see e.g.[1],[2],[3]), we obtain

$$\int_\Omega |u-u^*|dx + \int_0^t\int_\Omega g(v^*)|f(u)-f(u^*)|dxd\tau \le \int_\Omega |u_0-u_0^*|dx + \int_0^t\int_\Omega f(u)|g(v)-g(v^*)|dxd\tau \quad (7)$$

Analogously, by multiplying (6) by $\operatorname{sign}_0(v-v^*)$ and by using that

$$-\int_\Omega \Delta(v-v^*)\operatorname{sign}_0(v-v^*)dx \ge \frac{1}{\alpha}\int_{\partial\Omega} |v-v^*|ds$$

(which holds by regularization of $\operatorname{sign}_0(.)$ and by applying Green's formula) we get

$$\varepsilon\int_\Omega |v-v^*| dx + \frac{1}{\alpha}\int_0^t\int_{\partial\Omega} |v-v^*| dsd\tau + \lambda\int_0^t\int_\Omega f(u)|g(v)-g(v^*)|dxd\tau \le$$
$$\le \varepsilon\int_\Omega |v_0-v_0^*|dx + \lambda\int_0^t\int_\Omega g(v^*)|f(u)-f(u^*)|dxd\tau. \quad (8)$$

By multiplying (7) by λ and adding (8) we obtain (4). ∎

Remark 1. If we introduce the Banach space $E=L^1(\Omega)xL^1(\Omega)$ with the norm

$$\|(u,v)\| = \varepsilon\|u\|_{L^1(\Omega)} + \lambda\|v\|_{L^1(\Omega)}$$

and if we denote by $S(t):E \to E$ the semigroup operator defined by

$$S(t)(u_0,v_0)=(u(.,t),v(.,t))$$

with (u,v) solution of P_ε, then Theorem 1 shows that S(t) is a semigroup of contractions in E. The same type of argument (well-known in the framework of porous media equations [1],[6]) allows us to show that the operator $A:D(A)\subset E \to E$ defined by

$$A(u,v)=(f(u)g(v), -\frac{1}{\varepsilon}\Delta v + \frac{\lambda}{\varepsilon} f(u)g(v))$$
$$D(A)=\{(u,v)\in E:A(u,v)\in E\}$$

is accretive in E. In fact this property holds for a larger class of operators of the form

$$A(u,v)=(\mathcal{A}u+f(u)g(v),\ \mathcal{B}v+\mu f(u)g(v))$$

where $\mathcal{A}$ and $\mathcal{B}$ are accretive operators in $L^1(\Omega)$. So, it is possible to apply to problem P_ε the results of the abstract theory of accretive operators (see e.g.[1],[2],[3]).

3. The pseudo-steady-state problem. Since the usual magnitude of ε is very small, specialists in chemical engineering very often replace problem P_ε by the so called pseudo-steady-state formulation.

$$P_0 \begin{cases} \bar{u}_t = -f(\bar{u})g(\bar{v}) & \text{in } Q_\infty \\ -\Delta\bar{v} = -\lambda f(\bar{u})g(\bar{v}) & \text{in } Q_\infty \\ \alpha\frac{\partial\bar{v}}{\partial n} = 1-\bar{v} & \text{on } \partial\Omega x(0,\infty) \\ \bar{u}(x,0) = \bar{u}_0(x) & \text{on } \Omega . \end{cases}$$

where $\bar{v}=\bar{v}(x,t)$ and $\bar{u}_0 \in L^\infty(\Omega)$, $0 \le \bar{u}_0 \le 1$. We remark that no initial condition on $\bar{v}$ is needed because $\bar{v}(x,0)=\zeta(x)$ with $\zeta(x)$ determined as the (unique) solution of

$$\begin{cases} -\Delta\zeta = -\lambda f(\zeta)g(\bar{u}_0) & \text{in } \Omega \\ \alpha\dfrac{\partial\zeta}{\partial n} = 1-\zeta & \text{on } \partial\Omega. \end{cases}$$

It is easy to see that the existence and continuous dependence results of §2 can be extended to problem P_0.

Concerning the **convergence as ε tends to zero** of $(u_\varepsilon, v_\varepsilon)$, solution of P_ε, to $(\bar{u},\bar{v})$ solution of P_0 we obtain in [11] results of a different nature:

(i) Monotone iteration and super-subsolution technique: *Assuming* $u_{0,\varepsilon}(x) \ge \bar{u}_0(x)$ *and* $v_{0,\varepsilon}(x) \le \bar{v}_0(x)$ *then* $u_\varepsilon \searrow \bar{u}$ *and* $v_\varepsilon \to \bar{v}$ $(v_\varepsilon \le \bar{v})$ *uniformly in* Q. *If in addition*

$$-\Delta v_{0,\varepsilon} + \lambda f(1)g(v_{0,\varepsilon}) \le 0 \quad \text{in } \Omega$$

$$v_{0,\varepsilon} - 1 + \alpha\frac{\partial v_{0,\varepsilon}}{\partial n} \le 0 \quad \text{on } \partial\Omega$$

then $v_\varepsilon \nearrow \bar{v}$, *uniformly in* Q.

(ii) L^1-estimates: *Assume* $\|u_{0,\varepsilon}-\bar{u}_0\|_{L^1} \le C_1\varepsilon^{(1+\alpha_1)}$ *and* $\|v_{0,\varepsilon}-\bar{v}_0\|_{L^1} \le C_1\varepsilon^{(1+\alpha_2)}$ $(\alpha_1,\alpha_2 \ge 0)$ *then* $\|u_\varepsilon(.,t)-\bar{u}(.,t)\|_{L^1} \le \varepsilon(C_3\varepsilon^{\alpha_1}+C_4\varepsilon^{\alpha_2})$ *and* $\|v_\varepsilon(.t)-\bar{v}(.,t)\|_{L^1} \le C_5\varepsilon^{\alpha_1}+C_6\varepsilon^{\alpha_2}$. (The proof uses the same type of arguments as in Theorem 1).

(iii) Estimates (independent of $v_{0,\varepsilon}$ and $\bar{v}_0$) in $L^1(0,t:C(\bar{\Omega}))$: *Assume* $0 \le \bar{u}_0 \le u_{0,\varepsilon} \le 1$, $0 \le v_{0,\varepsilon} \le \bar{v}_0 \le 1$. *Then for any* $t>0$

$$0 \le \int_0^t (\bar{v}(x,\tau)-v_\varepsilon(x,\tau))d\tau \le \{\varepsilon + \lambda\|u_{0,\varepsilon}-\bar{u}_0\|_{L^\infty}\}w(x) \quad \text{a.e.}\, x \in \Omega$$

and

$$0 \le \int_\Omega (u_\varepsilon(x,t)-\bar{u}(x,t))dx \le \frac{\varepsilon|\Omega|}{\lambda} + \int_\Omega (u_{0,\varepsilon}(x)-\bar{u}_0(x))dx \ ,$$

where $w \in C^\infty(\Omega)$ *is the unique solution of*

$$\begin{cases} -\Delta w = 1 & \text{in } \Omega \\ w+\alpha\dfrac{\partial w}{\partial n} = 0 & \text{on } \partial\Omega \ . \end{cases}$$

(The idea of the proof is to integrate in time the equations of $\bar{v}$ and v_ε and in x the ones of $\bar{u}$ and u_ε). That extends previous results in [18] and [17].

4. On the solid conversion and the gas penetration.

Some particular examples show that if $f(s)=s^m$ with $0<m<1$ then the solid is partially converted for large time t and a free boundary F(t) appears defined as the boundary of the *conversion region* of the solid $\Omega_0^u(t):=\{x\in\overline{\Omega}:u(x,t)=0\}$. The following result gives a characterization of the functions f generating the free boundary F(t).

Theorem 2. *a) Assume* f(u) *such that*

$$\int_{0^+}\frac{ds}{f(s)} < \infty \qquad (9)$$

Then $\Omega_0^u(t)$ *is not empty for* t *large enough and in fact* $\Omega_0^u(t)=\Omega$ *after some suitable time* t_0.

b) If on the contrary

$$\int_{0^+}\frac{ds}{f(s)} = +\infty$$

then $u(x,t)>0$ *for any* $x\in\overline{\Omega}$ *and* $t>0$.

Idea of the proof. Define the auxiliary function

$$\theta(r) = \int_0^r\frac{ds}{f(s)}$$

Then it is easy to see that $u(x,t)=\theta^{-1}(\theta(u_0(x))-\int_0^t g(v(x,\tau))d\tau)$.
Finally as $v(x,t)\longrightarrow 1$ if $t\longrightarrow\infty$ and $\theta^{-1}(0)=0$ we obtain the conclusion (a). The proof of (b) uses a comparison argument. ▪

We also introduce the **overcall conversion at time t** by

$$\gamma(t) = \frac{\int_\Omega (u_0(x)-u(x,t))dx}{\int_\Omega u_0(x)dx}$$

It is easy to see that $\gamma(t)\nearrow 1$ as $t\nearrow\infty$, and that if (9) holds then there exist a t_0 (called **full conversion time**) at which $\gamma(t_0)=1$.

Theorem 3. *Let* $u_0\equiv 1$, $0\le v_0\le 1$, f *satisfying* (9) *and* g *such that there exists* g_1 *and* g_2 *Lipschitz nondecreasing functions with* $g_1(1)=g_2(1)=g(1)$ *and satisfying* $g_1(r)\le g(r)\le g_2(r)$ $\forall r\in[0,1]$. *Then we have the estimate*

$$\frac{1}{g(1)}(I+M_2\lambda\|w\|_{L^\infty}) \le t_0 \le \frac{1}{g(1)}(I+M_1\lambda\|w\|_{L^\infty})$$

where $I=\theta(1)$, $M_1=\sup\{g_1(r):r\in[0,1]\}$, $M_2=\inf\{g_2(r):r\in[0,1]\}$.

We shall not give the proof of this result but we point out an interesting application to the case of $g(v)=v^p$, $p>0$:(i). If $p=1$ then $t_0=I+\lambda\|w\|_{L^\infty}$ (*Sohn-Szekely law of additive times*); (ii) If $p<1$ then $I+p\lambda\|w\|_{L^\infty}\le t_0\le I+\lambda\|w\|_{L^\infty}$; (iii) If $p>1$, $I+\lambda\|w\|_{L^\infty}\le t_0\le I+p\lambda\|w\|_{L^\infty}$. Similar results holds for the pseudo-steady-state problem. These results generalize the ones of [19] and [17].

Finally another free boundary may occur as the boundary of the *gas penetration region* $\Omega_0^v(t)=\{x\in\Omega: v(x,t)\neq 0\}$.

Theorem 4. *Assume* g *such that*

$$\int_{0^+}\frac{ds}{\sqrt{G(s)}}<\infty \qquad , \; G(s):=\int_0^s g(r)dr \; .$$

Then for any $t>0$ *there exists* $C(t)>0$ *such that*

$$\overset{v}{\Omega}_0(t)\supset\{x\in\Omega\text{-supp}v_0 : d(x,\text{supp}v_0)\geq C(t)\}.$$

On the other hand, if

$$\int_{0^+}\frac{ds}{\sqrt{G(s)}} = +\infty$$

then $v(x,t)>0$ *for any* $t>0$ *and* $x\in\overline{\Omega}$.

The proof uses some results in the literature (see [4],[5],[7],[8],[9],[10]). We refer the reader to [11] for details and other additional results.

References

[1] Benilan, Ph. Equations d'evolution dans un espace de Banach quelconque el applications. Doctoral Thesis, Orsay (1972).

[2] Benilan, Ph. Crandall, M.G. and Pazy, A. Nonlinear Evolution Equations Governed by Accretive Operators. Book to appear in Springer-Verlag.

[3] Crandall, M.G. "Nonlinear semigroups and evolution governed by accretive operators". In Nonlinear Functional Analysis and Its Applications. F.E. Browder ed. Proceedings of Symposia in Pure Mathematics. Vol 45-Part 2. AMS, (1986). 305-339.

[4] Bandle, C. Sperb, R.P. and Stakgold, I. "Diffusion-reaction with monotone kinetics". Nonlinear Analysis, 8 (1984), 321-333.

[5] Bandle, C. and Stakgold, I. "The formation of the dead core in parabolic reaction-diffusion equations". Trans. Amer. Math. Soc. 286 (1984), 275-293.

[6] Diaz, J.I. "Solutions with compact support for some degenerate parabolic problems". Nonlinear Analysis 3 (1979), 831-847.

[7] Diaz, J.I. Nonlinear partial differential equations and free boundaries Vol. 1, Elliptic Equations. Pitman, London, 1985.

[8] Diaz, J.I. and Hernandez, J. "On the existence of a free boundary for a class of reaction diffusion systems" SIAM J. Math. Anal. 15 (1984), 670-685.

[9] Diaz, J.I. and Hernandez, J. "Some results on the existence of free boundaries for parabolic reaction-diffusion systems". In Trends in Theory and Practice of Nonlinear Differential Equations, V. Lakshmikantham ed. Marcel Dekker, (1984), 149-156.

[10] Diaz, J.I. and Hernandez J. "Qualitative properties of free boundaries for some nonlinear degenerate parabolic equations". In Nonlinear parabolic equations: qualitative properties of solutions. L. Boccardo and A. Tesei eds. Pitman (1987), 85-93.

[11] Diaz, J.I. and Stakgold, I. "Mathematical aspects of the combustion of a solid by a distributed isothermal gas reaction". (To appear).

[12] Diaz, J.I. and Vrabie, I.I. (Article in preparation).

[13] Froment, G.F. and Bischoff, K.B. Chemical reactor design and analysis. Wiley and Sons, (1979).

[14] Ladde, G.S., Lakshmikantham, V. and Vatsala, A.S. Monotone iterative techniques for nonlinear differential equations. Pitman, (1985).

[15] Pao, C.V. "Mathematical analysis of enzyme-substrate reaction diffusion in some biochemical systems". Nonlinear Analysis, 4, (1980), 369

[16] Stakgold, I. "Gas-solid reactions" in Dynamical Systems II. A.R. Bednarek and L. Cesari eds. Academic Press, (1982).

[17] Stakgold, I."Reaction-diffusion problems in chemical engineering". In Nonlinear Diffusion Problems. A. Fasano and M. Primicerio. eds. Springer, (1986), 119-152.

[18] Stakgold, I., Bischoff, K.B. and Gokhale, V. "Validity of the pseudo-steady-state approximation". Int. J. of Eng. Sci. 21 (1983), 537-542.

[19] Stakgold, I. and McNabb, A. "Conversion estimates for gas-solid reactions". Math. Modelling, 5 (1984), 325-330.

[20] Stakgold, I. and DiLiddo, A. "Isothermal combustion with two moving fronts". (To appear).

[21] Szekely, J., Evans, J.W. and Sohn, H.Y. Gas-solid reactions. Academic Press, (1976).