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ON A FREE BOUNDARY PROBLEM ARISING IN THE POLYMER INDUSTRY

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Abstract. A free boundary problem arising from a model for sorption of swelling solvent into glassy polymers is considered. Global existence and uniqueness of the solution are proved.

1. Introduction

In this paper we consider a free boundary problem arising from a model for sorption of swelling solvent in a glassy polymer. This model has been proposed 1978 by Astarita and Sarti [2]. It has been studied in different abstract forms (see [1], [3], [4], [5], [7], [8]).

This model is characterized by the presence of a threshold value for the concentration under which no sorption takes place. The polymer is then divided into two regions of different morphology, a swollen zone in which the concentration of the solvent exceeds the threshold value, and a glassy zone in which the concentration is negligibly small and actually taken zero in the model. The interface between these regions is the free boundary. The solvent is supposed to diffuse in the penetrated zone according to Fick's law.

The most interesting case from both the application and the mathematical point of view occurs when the polymer is initially unpenetrated.

Here we study one of the abstract free boundary problems arising from above model. It can be formulated as follows:

Problem (P): . Find a triple $(T, s(t), c(x,t))$ such that $T > 0, s(t) \in C^1[0,T]$, $c(x,t) \in C^{2,1}(D_T) \cap C(\overline{D}_T), D_T = \{(x,t) : 0 < x < s(t), 0 < t < T\}$, where ($\overline{D}_T$ is the closure of D_T), c_x is continuous up to $x = s(t)$, and the following equations and conditions are satisfied:

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$$c_{xx} - c_t = 0 \quad \text{in} \quad D_T, \tag{1.1}$$

$$s(0) = 0, \tag{1.2}$$

$$c(0,t) = g(t),\ g(0) = 1,\ 0 < t < T, \tag{1.3}$$

$$\dot{s}(t) = f(c(s(t),t)),\ 0 < t < T, \tag{1.4}$$

$$c_x(s(t),t) = -(c(s(t),t) + q)\dot{s}(t),\ 0 < t < T, \tag{1.5}$$

where q is a given nonnegative constant, f and g are given functions.

In this problem, $c(x,t) + q$ is the concentration at $(x,t), q$ is the threshold value. Condition (1.4) describes the penetration law, and condition (1.5) is the mass balance at the free boundary $x = s(t)$.

Throughout the paper the functions f and g will be supposed to satisfy the following assumptions:

$$f \in C^1(0,1],\ f'(c) > 0 \quad \forall c \in (0,1],\ f(0) = 0. \tag{1.6}$$

$$g \in C^1[0,+\infty),\ g(t) \geq 0\ \forall t \geq 0,\ 0 \geq g'(t) \geq -G_1\ \forall t \geq 0, \tag{1.7}$$

where G_1 is a given positive constant.

We notice that because of (1.6) there exists an inverse function Φ of f such that condition (1.4) can be rewritten in the following equivalent form:

$$c(s(t),t) = \Phi(\dot{s}(t)),\ 0 < t < T. \tag{1.4'}$$

2. Local existence

Set $\mathcal{R} = \{r(t) : r(t) \in C^1[0,T] \cap C^2(0,T),\ r(0) = 0,\ \dot{r}(0) = f(1),\ 0 \leq \dot{r}(t) \leq f(1) \text{ in } [0,T],\ |\ddot{r}(t)| \leq K \text{ in } (0,T)\}$, where T and K are some positive constants. Let $r(t)$ be a function of $\mathcal{R}$. We consider the following auxiliary problem:

AUXILIARY PROBLEM: . Find a function $c(x,t) \in C^{2,1}(D) \cap C(\overline{D}),\ D = \{(x,t) : 0 < x < r(t),\ 0 < t < T\}$, c_x is continuous up to $x = r(t)$, such that:

$$c_{xx} - c_t = 0 \qquad \text{in} \quad D_T, \tag{2.1}$$

$$c(0,t) = g(t), \; g(0) = 1, \; 0 < t < T, \tag{2.2}$$

$$c_x(s(t),t) = -(\Phi(\dot r(t)) + q)\dot r(t), \; 0 < t < T. \tag{2.3}$$

It is well-known that with the assumptions above, Problem (2.1) – (2.2) has a unique solution (see, for example, [6]). There are some a priori estimates of the solution of this Problem.

PROPOSITION 2.1. *Let c be a solution of Problem (2.1) – (2.3). Then*

$$c(x,t) \le 1 \qquad \forall (x,t) \in \overline{D} \setminus \{0,0\}, \tag{2.4}$$

$$-f(1)(1+q) < c_x(x,t) \le M \qquad \forall (x,t) \in \overline{D} \setminus \{0,0\}, \tag{2.5}$$

where

$$M = \max_{(x,t)\in\overline{D}} \{ \frac{e^{2t}}{x+1} \max_{0\le\tau\le T} (-e^{-2\tau} g'(\tau))\} \ge 0.$$

PROOF: Using the maximum principle we can prove (2.4) and the first inequality in (2.5). In order to prove the last inequality in (2.5) we set $v(x,t) = e^{-2t}(x+1)c_x(x,t)$. Then v is a solution of the following problem:

$$L(v) = v_{xx} - \frac{2}{x+1} v_x + [\frac{2}{(x+1)^2} - 2]v - v_t = 0 \quad \text{in} \quad D, \tag{2.6}$$

$$v(0,t) - v_x(0,t) = -e^{-2t} g'(t), \; 0 < t < T, \tag{2.7}$$

$$v(r(t),t) = -e^{-2t}\dot r(t)(r(t)+1)(\Phi(\dot r(t)) + q), \; 0 < t < T. \tag{2.8}$$

We notice that there are three cases: If $v \le 0$ in $\overline{D}$, then $c_x \le M$ in $\overline{D}$. If v has a positive maximum in D or in $t = T$ at (x_0, t_0), then

$$v_t(x_0,t_0) \ge 0, \; v_x(x_0,t_0) = 0, \; v_{xx}(x_0,t_0) < 0.$$

Since $\frac{2}{(x+1)^2} - 2 \le 0$, we have $L(v(x_0,t_0)) < 0$, which contradicts (2.6). So v attaints its positive maximum only on $x = 0$. Hence

$$c_x(x,t) \le \frac{e^{2t}}{x+1} \max_{0\le\tau\le T} (-e^{-2\tau} g'(\tau)) \le M.$$

REMARK 2.1: For any positive constant $c_0 < 1$ and $f(1) > \dot{r}_0 > 0$, a small enough $T > 0$ can be found such that

$$c(x,t) \geq c_0 > 0, \qquad (x,t) \in D, \tag{2.9}$$

$$\dot{r}(t) \geq \dot{r}_0 > 0, \qquad 0 \leq t \leq T. \tag{2.10}$$

PROPOSITION 2.2. *Let c be a solution of Problem (2.1) – (2.3), then $c \in C^{2,1}(\overline{D})$, $c_{xt} \in C(\overline{D} \setminus \{0,0\})$ and there exists a positive constant Q depending on f, g, K such that*

$$|c_t(x,t)| \leq Qt + G_1, \ (x,t) \in \overline{D} \setminus \{0,0\}. \tag{2.11}$$

PROOF: Set $w = c_t$, then w is a solution of the following problem:

$$w_{xx} - w_t = 0 \qquad \text{in} \qquad D, \tag{2.12}$$

$$w(0,t) = g'(t), \ 0 < t < T, \tag{2.13}$$

$$\begin{aligned}(w_x + w\dot{r})_{|x=r(t)} &= -\ddot{r}(t)[q + \Phi(\dot{r}(t)) + \Phi'(\dot{r}(t))\dot{r}(t)] \\ &\equiv h(t), \ 0 < t < T.\end{aligned} \tag{2.14}$$

We notice that $w \in C(\overline{D})$ and that

$$|h(t)| \leq K[q + 1 + f(1) \max_{0 \leq y \leq f(1)} \Phi'(y)] \equiv H.$$

By the same way as in [3] we obtain

$$0 \leq v(x,t) \leq \max\{G_1, \frac{H}{\dot{r}_0}\} \equiv G_0, \ (x,t) \in \overline{D}.$$

Then (2.14) yields

$$\begin{aligned}|w(x,t)| = |w(0,t) + \int_0^x w_\xi(\xi,t)d\xi| \leq \\ \leq G_1 + (H + G_0 f(1))f(1)t \equiv G_1 + Qt.\end{aligned}$$

Denote by $c_i(x,t)$ the solution of problem (2.1) – (2.3) with respect to $r_i \in \mathcal{R}$ and $D_i = \{(x,t) : 0 < x < r_i(t), \ 0 < t < T\}$, $i = 1, 2$.

PROPOSITION 2.3. *Under the above assumptions, the constants $T_0 > 0$ and $N > 0$ can be found such that for any $T \in (0, T_0)$,*

$$|c_1(r_1(t), t) - c_2(r_2(t), t)| \leq NT \| r_1 - r_2 \|_{C^1(0,T)}, \; 0 < t < T. \qquad (2.15)$$

Moreover, N depends on f, g, q, K.

PROOF: As in [7].

Now we prove the local existence of solutions of Problem (P).

Let α be a positive constant, $\alpha < 1$. Denote by $\gamma(t)$ a positive non-increasing function defined for $t > 0$. Let $X(K, T, \gamma)$ be the set of all functions $r(t) \in \mathcal{R}$ such that

$$|\ddot{r}(t_1) - \ddot{r}(t_2)| \leq \gamma(\tau)(t_1 - t_2)^{\alpha/2}, \; 0 < \tau \leq t_1 \leq t_2 \leq T. \qquad (2.16)$$

Note that this set is closed in $C^1[0, T]$. For any function $r(t) \in X$, we define $c(x, t)$ to be a solution of the auxiliary problem (2.1) - (2.3). Let $\bar{r}$ be a solution of the problem:

$$\dot{\bar{r}}(t) = f(c(r(t), t)), \; 0 < t < T, \qquad (2.17)$$

$$\bar{r}(0) = 0. \qquad (2.18)$$

Then we can define the transformation

$$\mathcal{F} : r \in X \mapsto \bar{r} \in C^1[0, T] \cap C^2(0, T).$$

PROPOSITION 2.4. *There are positive constants K, T and a function γ such that the transformation $\mathcal{F}$ is a contractive mapping from X into itself.*

PROOF: It is clear that $\dot{\bar{r}}(0) = f(1)$ and $0 \leq \dot{\bar{r}}(t) \leq f(1)$. We only need to show that K, T can be chosen such that $|\ddot{\bar{r}}(t)| \leq K$ for all $t \in (0, 1)$ and then verify (2.16).

Taking the derivative with respect to t on both sides of (2.17), we get

$$\ddot{\bar{r}}(t) = f'(c(r(t), t)).[c_x(r(t), t)\dot{r}(t) + c_t(r(t), t)].$$

Hence

$$|\ddot{r}(t)| \leq F_1[Pf(1) + (Qt + G_1)], \; 0 < t < T,$$

where $F_1 = \max\limits_{c \in [c_0,1]} f'(c)$, $P = \max\{f(1)(1+q), M\}$. If we choose $K = 2F_1(Pf(1) + G_1)$ and T small enough then $|\ddot{r}(t)| \leq K \quad \forall t \in (0,T)$. Hence we can proceed as in [7] to complete the proof.

THEOREM 2.1. *There exists a positive constant T_0 such that Problem (P) has a solution for $T \leq T_0$. Moreover,*

$$c \in C^{2,1}(\overline{D}_T), \; c_{xt} \in C(\overline{D}_T \setminus \{0,0\}), \; s \in C^2[0,T].$$

PROOF: The existence follows from Proposition 2.4 and Banach's fixed point theorem. The regularity properties of c and s follows from Proposition 2.3 and the definition of X.

3. Uniqueness

First we prove the monotone dependence of $s(t)$, from which we get the uniqueness of the solution.

Let c_i, s_i $(i = 1,2)$ be a solution of the problem:

$$\begin{aligned}
&c_{ixx} - c_{it} = 0, \quad 0 < x < s_i(t), \; t_i < t < T, \\
&s_i(t_i) = 0, \\
&c_i(0,t) = g(t), \quad t_i < t < T, \\
&\dot{s}_i(t) = f(c_i(s_i(t),t)), \quad t_i < t < T, \\
&c_{ix}(s_i(t),t) = -(c_i(s_i(t),t) + q)\dot{s}_i(t), \quad t_i < t < T.
\end{aligned}$$

LEMMA 3.1. *If $t_1 < t_2$, then*

$$s_1(t) > s_2(t), \quad t_2 < t < T.$$

PROOF: Set

$$u(x,t) = -\int_x^{s(t)} [c(y,t) + q]dy. \tag{3.1}$$

This transformation carries (1.1) – (1.5) into a Stefan-like problem:

$$u_{xx} - u_t = 0 \quad \text{in} \quad D_T, \tag{3.2}$$

$$s(0) = 0, \tag{3.3}$$

$$u_x(0,t) = g(t) + q, \ 0 < t < T, \tag{3.4}$$

$$u(s(t),t) = 0, \ 0 < t < T, \tag{3.5}$$

$$u_x(s(t),t) = \Phi(\dot{s}(t)) + q, \ 0 < t < T. \tag{3.6}$$

Consider the function $u_i(x,t)$ obtained from $c_i(x,t)$ by (3.1). Suppose that there exists $t_0 = \min_{t_2<t<T}\{t : s_1(t) = s_2(t)\}$. Then

$$\dot{s}_1(t_0) \leq \dot{s}_2(t_0). \tag{3.7}$$

Set $w(x,t) = u_1(x,t) - u_2(x,t)$. By the strong maximum principle, we obtain $w_x(s_2(t_0),t_0) > 0$, hence $\dot{s}_1(t_0) > \dot{s}_2(t_0)$, which contradicts (3.7).

THEOREM 3.1. *Problem (P) has at most one solution.*

PROOF: Let (T_1, s_1, c_1) and (T_2, s_2, c_2) be two maximal solutions of Problem (P). Suppose that $T_2 \leq T_1 \leq +\infty$. For any $\epsilon > 0$, we define

$$s_\epsilon(t) = s_2(t-\epsilon), \ c_\epsilon(x,t) = c_2(x,t-\epsilon)$$

$$s_{-\epsilon}(t) = s_2(t+\epsilon), \ c_{-\epsilon}(x,t) = c_2(x,t+\epsilon)$$

Because of the time invariance of Problem (P), $(T_2 + \epsilon, s_\epsilon, c_\epsilon)$ and $(T_2 - \epsilon, s_{-\epsilon}, c_{-\epsilon})$ are also solutions corresponding to the data $s_\epsilon(\epsilon) = 0, \ s_{-\epsilon}(-\epsilon) = 0$. Applying Lemma 3.1, we get

$$s_\epsilon(t) < s_1(t), \ \epsilon \leq t \leq T_2$$

$$s_1(t) < s_{-\epsilon}(t), \ 0 \leq t \leq T_2 - \epsilon.$$

Letting ϵ tend to 0, we obtain $s_1(t) \equiv s_2(t), \ 0 \leq t \leq T_2$, and $c_1(x,t) \equiv c_2(x,t)$ in $\overline{D}_{T_2}$. Because of the assumption on maximality, we obtain $T_1 = T_2$.

REMARK 3.1: From Theorem 2.1 and Theorem 3.1 it follows that for any solution of Problem (P), a positive constant $T_0 > 0$ can be found such that

$$c \in C^{2,1}(\overline{D}_{T_0}), \ c_{xt} \in C(\overline{D}_{T_0} \setminus \{0,0\}), \ s \in C^2[0,T_0].$$

4. Regularity, convexity, global existence

PROPOSITION 4.1. *Let s, c be a solution of Problem (P) for a given $T < +\infty$. Then there exists $c_0 = c_0(T) > 0$ such that*

$$c_0 \le c(x,t) < 1,\ 0 < x \le s(t),\ 0 < t < T, \tag{4.1}$$

$$0 < f(c_0) \equiv \dot{r}_0 \le \dot{s}(t) \le f(1),\ 0 < t < T, \tag{4.2}$$

$$-f(1)(1+q) < c_x(x,t) < M,\ \text{in } D_T. \tag{4.3}$$

PROOF: If $c(x,t)$ attains the value 0 (we need only to consider on $x = s(t)$) for the first time at some point $(s(t_0), t_0)$, then $c_x(s(t_0), t_0) = 0$. On the other hand, $(s(t_0), t_0)$ is a minimum of $c(x,t)$ in $\overline{D}_{t_0}$. Using the strong maximum principle we get $c_x(s(t_0), t_0) = 0$, which leads to a contradiction. Hence $c(x,t) \ge c_0 > 0$ and $\dot{s}(t) = f(c(s(t),t)) \ge f(c_0) = \dot{r}_0 > 0$. The last inequalities in (4.1), (4.2) follow as in the proof of Proposition 2.1. Similarly, we can prove the inequality (4.3).

THEOREM 4.1. *Let (T, s, c) be a solution of Problem (P). Then $s \in C^2[0,T]$. Moreover, if $f \in C^\infty(0,1]$, then $s \in C^\infty(0,T]$.*

PROOF: We can apply the iterative technique of [9] to the equivalent problem (3.2) – (3.6) with (3.6) rewritten in the form:

$$\dot{s}(t) = f(u_x(s(t),t) - q),\ 0 < t < T.$$

From the continuity of f', it follows the continuity of $\ddot{s}$ in $(0,T]$. If $f \in C^\infty(0,1]$, we similarly get $s \in C^\infty(0,T]$. The continuity of $\ddot{s}(t)$ at $t = 0$ is already known in Remark 3.1.

Now we prove the convexity of the free boundary.

THEOREM 4.2. *Assume that (T, s, c) solves Problem (P). Then*

$$\ddot{s}(t) < 0,\ 0 \le t \le T. \tag{4.4}$$

PROOF: The continuity of c_t in $\overline{D}_T$ and the continuity of c_{xt} in $\overline{D}_T \setminus \{0,0\}$ follow from Theorem 4.1.

Define

$$v(x,t) = [\ln(c+q)]_{xx},$$

we can see that v is continuous in $\overline{D}_T$ and v_x is continuous in $\overline{D}_T \setminus \{0,0\}$. Moreover v is a solution of the problem:

$$v_{xx} + 2[\ln(c+q)]_x v_x + 2v^2 - v_t = 0 \text{ in } D_T, \tag{4.5}$$

$$v(0,t) = \{(c_{xx}(c+q) - c_x^2)/(c+q)^2\}_{|x=0}, \ 0 < t < T, \tag{4.6}$$

$$v(s(t),t) = \{\ddot{s}(t)\Phi'(\dot{s}(t))\}/(\Phi(\dot{s}(t)) + q), \ 0 < t < T. \tag{4.7}$$

Because $\ddot{s}(t)$ is continuous at $t = 0$ and $\ddot{s}(0) = f'(1)[g'(0) - f^2(1)(1+q)] < 0$ there exists $t_0 > 0$ such that $\ddot{s}(t)$ and consequently $v(s(t),t)$ is negative in $[0,t_0]$. Notice that $c_{xx}(0,t) = c_t(0,t) = g'(t) \leq 0$, we get $v(0,t) < 0$. If $v(s(t_0),t_0) = 0$, then $v_x(s(t_0),t_0) > 0$ by the strong maximum principle. Because of (4.7) we obtain $\ddot{s}(t_0) = 0$. Note that

$$v_x(s(t),t) = \{[-\ddot{s}(t)(c+q) + \dot{s}(t).\ddot{s}(t)\Phi'(\dot{s}(t))]/(c+q)\}_{|x=s(t)}.$$

Then $v_x(s(t_0),t_0) = 0$, a contradiction.

Corollary 4.1. *Let (T,s,c) be a solution of Problem (P). Then*

$$c_t(x,t) \geq -G_1, \qquad (x,t) \in \overline{D}_T. \tag{4.8}$$

Proof: As in the proof of Proposition 2.2, we set $w = c_t$ and get (2.12) – (2.14). Since $\ddot{s}(t) < 0$, $h(t) \geq 0$. So w cannot attain negative minimum on $x = s(t)$. Hence

$$w(x,t) \geq \min_{0\leq\tau\leq T} g'(\tau) \geq -G_1, \ (x,t) \in \overline{D}_T.$$

Corollary 4.2. *If (T,s,c) is a solution of Problem (P), then*

$$\ddot{s}(t) \geq -[f^2(1)(q+1) + G_1] \max_{c\in[c_0,1]} f'(c), \ 0 < t < T. \tag{4.9}$$

Proof: Using Proposition 2.1 and Corollary 4.1, we obtain

$$\ddot{s}(t) = \{f'(c)[c_x\dot{s}(t) + c_t]\}_{|x=s(t)} \geq \max_{c\in[c_0,1]} f'(c)[-f^2(1)(q+1) - G_1]$$

THEOREM 4.3. *Problem (P) admits a solution for arbitrary $T > 0$.*

PROOF: Assume that there exists $T^* > 0$ such that the solution cannot be continued beyond T^*. Because of the nonotonicity of s and $\dot{s}$, $\lim_{t\to T^*-} s(t)$, $\lim_{t\to T^*-} \dot{s}(t)$ both exists. Using the transformation (3.1), we obtain the following free boundary problem:

$$\begin{aligned}
&u_{xx} - u_t = 0,\ 0 < x < s(t), T^* < t < \widehat{T},\\
&u_x(0,t) = g(t) + 1,\ T^* < t < \widehat{T},\\
&u(s(t),t) = 0,\ T^* < t < \widehat{T},\\
&u_x(s(t),t) = \Phi(\dot{s}(t)) + q,\ T^* < t < \widehat{T}.
\end{aligned}$$

with initial data

$$s(T^*) = \lim_{t\to T^*-} s(t),\ u(x,T^*) = \lim_{t\to T^*-}\{-\int_x^{s(t)} [c(t,t) + q]dy\}.$$

This problem has a unique solution for suitable $\widehat{T} > T^*$ (see [6]), which leads to a contradiction.

5. Asymptotic estimates

Let $s(t)$, $c(x,t)$ be a solution of Problem (P) for any $T > 0$. Using Green's inequality

$$\oint_{\partial D_t} Pd\tau + Qdx = \int_{D_t}\int (\frac{\partial Q}{\partial \tau} - \frac{\partial P}{\partial x})dxd\tau,\ 0 < t < T,$$

with $P = xc_x - c$, $Q = xc$, we get

$$\frac{1}{2}qs^2(t) + \int_0^{s(t)} x\dot{c}(x,t)dx + \int_0^t c(s(\tau),\tau)d\tau = \int_0^t g(\tau)d\tau. \tag{5.1}$$

Set $L = \int_0^{+\infty} g(\tau)d\tau$.

THEOREM 5.1. *If $q > 0$, then*

$$\lim_{t\to+\infty} s(t) \leq \sqrt{\frac{2L}{q}}. \tag{5.2}$$

Moreover, if $L < +\infty$, then

$$\lim_{t\to+\infty} \dot{s}(t) = 0. \tag{5.3}$$

PROOF: To prove (5.2), it suffices to use the positivity of c and (5.1). To prove (5.3), we note that there exist $\lim_{t\to+\infty} s(t)$, $\lim_{t\to+\infty} \dot{s}(t)$ because of the monotonicity of $s, \dot{s}$ and of (5.2). But from (5.2) it follows that $\lim_{t\to+\infty} \dot{s}(t) = 0$.

THEOREM 5.2. *If $q = 0$, $g(t) \geq G > 0$ for all $t > 0$, G is a given constant, then*

$$\lim_{t\to+\infty} s(t) = +\infty. \tag{5.4}$$

PROOF: From (5.1) we obtain

$$\int_0^{s(t)} xc(x,t)dx = \int_0^t [g(\tau) - c(s(\tau), \tau)]d\tau. \tag{5.5}$$

Since $\ddot{s}(t) < 0$, there exists $\lim_{t\to+\infty} \dot{s}(t)$, and consequently $\lim_{t\to+\infty} c(s(t),t)$. Suppose that $\lim_{t\to+\infty} s(t) < +\infty$. Then $\lim_{t\to+\infty} \dot{s}(t) = 0$ which implies $\lim_{t\to+\infty} c(s(t),t) = 0$. It then follows

$$g(\tau) - c(s(\tau), \tau) \geq G - c(s(\tau), \tau) \geq G^* > 0,$$

if τ is large enough. Hence, the right hand side of (5.5) tends to $+\infty$ when t tends to $+\infty$. But the left hand side of (5.5) is bounded. Hence we have a contradiction.

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