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A REACTION-DIFFUSION SYSTEM MODELLING THE POST IRRIDIATION OXYDATION OF AN ISOTACTIC POLYPROPYLENE

1. Introduction

The spontaneous oxydation of isotactic polypropylene is a reaction of great interest not only in practice but also in modelling theory.

So in [2] a kinetic submodel have been studied, describing the reaction of post-irridiaton between oxygen and the most stable macro-radical alkyl, produced by irridiation in vacuum, of an isotactic polypropylene.

Here, we are concerned with the reaction-diffusion system originated from [2] which is the following weakly coupled system of nonlinear partial differential equations

$$(1.i)\qquad \partial_t u_i - \mu_i \partial_{xx} u_i = F_i(U), \quad i\in I:=\{1,2,3,4\};\ x\in\Omega:=(0,1),\ t\geq 0,$$

in the concentration functions u_1, u_2, u_3, u_4 with

$U = (u_1,u_2,u_3,u_4)$,

$F_1(U) = -k_1u_1u_2 + k_2u_3^2$,

$F_2(U) = -k_1u_1u_2 - 2k_6u_2^2 - k_5u_3u_2 + k_3u_3 + k_8u_4$,

$F_3(U) = k_1u_1u_2 - k_5u_2u_3 - (k_2+k_3)u_3 - 2k_3u_3^2$,

$F_4(U) = 2k_3u_3^2 - (k_7+k_8)u_4$.

The diffusivitivities μ_i, $i\in I$, and the coefficients k_j, $j=1,\dots,8$, are positive constants, they are factors of proportionality and measure the proportion of reactional collisions.

The concentrations u_i, $i \in I$, satisfy the boundary conditions

(1.5) $$\begin{cases} u_i(t,0) = u_i(t,1) = 0, & t>0 \text{ for } i=2,3,4, \\ \partial_x u_2(t,0) = \partial_x u_2(t,1) = 0, & t>0, \end{cases}$$

and the initial data

(1.6) $$u_i(0,x) = \phi_i(x), \qquad x \in \bar{\Omega}, \qquad i \in I,$$

where $\phi_i(0) = \phi_i(1) = 0$, $i = 1,2,3$, and $\phi_2'(0) = \phi_2'(1) = 0$.

Given are the initial concentrations $\phi_i(x)$, $i \in I$, all of which are assumed to be nonnegative and continuous on $[0,1]$.

Our aim will be to establish the existence of global bounded solutions to problem (1.1)-(1.6) and analyze their large time behaviour by methods of functional analysis, specifically the theory of semigroups of linear operators in Banach spaces. We shall prove that for problem (1.1)-(1.6) there exists a unique nonnegative solution which goes to 0, uniformly in every closed interval contained in Ω, for each $i \in I$. Moreover, we shall show that there exist a positive T large enough, positive constants $K_i > 0$, $i = 1,3,4$, and α, C_0, C_1 with $C_0 < C_1$ such that for any $t \geq T$, we have

$$\|u_i\|_\infty \leq K_1 \exp(-\alpha t) \quad \text{for} \quad i=1,3,4,$$

$$C_0/t \leq \|u_2\|_\infty \leq C_1/t.$$

It is worth noting that the problem (1.1)-(1.6) can be analyzed neither in the framework of the theory of monotonic or quasimonotonic systems nor by the techniques of comparison [1], [4], [8], [12].

2. Notations and preliminary results

In order to study the problem (1.1)-(1.6), we introduce the Banach space $X := C([0,1],\mathbb{R}^4)$ (of the functions $u = (u_1,u_2,u_3,u_4)^T$ (T for transpose) which are continuous in $[0,1]$), endowed with the norm

$$\|U\| = \sum_{j=1}^{4} \sup|u_j(x)| \quad \text{for} \quad x \in [0,1].$$

Let A be defined as the linear operator $D\partial_{xx} = \mathrm{diag}(\mu_1,\mu_2,\mu_3,\mu_4)\partial_{xx}$ with domain

$$D(A) := \{U \in X \mid D\partial_{xx}U \in X \ \text{and} \ u_i(t,0) = u_i(t,1) = 0,\ i=1,3,4,$$
$$\partial_x u_2(t,0) = \partial_x u_2(t,1) = 0\}$$

and let F denote the nonlinear operator

$$F(U) = (F_1(U),F_2(U),F_3(U),F_4(U))^T \in X \text{ for } U \in X,$$

so (1.1)-(1.6) can be written in the abstract form

$$(2.1) \qquad d/dt(U(t)) = AU(t) + F(U(T)),$$

$$(2.2) \qquad U(0) = \phi \in X_+,$$

where X_+ denotes the set of nonnegative functions from X.

By Theorem 1.5 of [10] we can show that a unique local classical solution of problem (2.1)-(2.2) exists; by this we mean that

$$U \in (C^{2,1}(\Omega \times (0,T),R) \cap C^{1,0}(\bar{\Omega} \times (0,T),R))^4$$

and satisfies system (1.1)-(1.6). This can be proved using general results concerning the semigroup theory because each operator $\mu_i\partial_{xx}$ generates an analytic semigroup on $C(\bar{\Omega})$.

Moreover, we have the following alternative:

$$(\text{Alt.}) \quad \begin{cases} \text{Either} \quad T = \infty, \\ \text{or} \quad T < \infty \quad \text{and} \quad \lim_{t \to \infty} \|U\| = \infty. \end{cases}$$

An important fact is that U(t,x) remains nonnegative, if $\phi_i(x) \geq 0$, $i \in I$.

Lemma 2.1. Let U be a local solution to (1.1)-(1.6). If $\phi_i(x) \geq 0$, $i \in I$, then $u_i \geq 0$, $i \in I$, on $[0,T) \times \Omega$.

Proof. Observe first that u_4 satisfies the inequality

$$\partial_t u_4 - \mu_4 \partial_{xx} u_4 + (k_7 + k_8) u_4 = 2k_3 u_3^2 \geq 0$$

as $\phi_4 \geq 0$, by the maximum principle $u_4 \geq 0$.

Now we restrict ourselves to $t \in [0, T_0]$ for a fixed $T_0 < T$. By the local result, we can choose a constant $C = C(T_0)$ such that $p(t,x) = C + k_1 u_1(t,x) \geq 0$, then we can write

$$\partial_t(\exp(-Ct)u_1) - \mu_1 \partial_{xx}(\exp(-Ct)u_1) + p(t,x)(\exp(-Ct)u_1) = k_3 \exp(-Ct) u_3^2 \; 0$$

so, again by the maximum principle $u_1 \geq 0$.

For the positivity of u_2 and u_4, we multiply (1.2) by $u_2(t)^- = (|u_2(t)| - u_2(t))/2$ and (1.3) by $u_3(t)^-$ and integrating over Ω to obtain

$$\int_\Omega u_2' u_2^- = \mu_2 \int_\Omega \partial_{xx} u_2 u_2^- - k_1 \int_\Omega u_1 u_2 u_2^- - 2k_6 \int_\Omega u_2^2 u_2^- - k_5 \int_\Omega u_2 u_3 u_2^- +$$

$$+ k_3 \int_\Omega u_3 u_2^- + k_8 \int_\Omega u_4 u_2^-$$

and

$$\int_\Omega u_3' u_3^- = \mu_3 \int_\Omega \partial_{xx} u_3 u_3^- + \int_\Omega u_1 u_2 u_3^- - k_5 \int_\Omega u_2 u_3 u_3^- -$$

$$- (k_2 + 2k_4) \int_\Omega u_3 u_3^- - 2k_3 \int_\Omega u_3^2 u_3^-,$$

where we have suppressed the dependence on t.

By the local existence, we clearly have

$$u_3 \in C^1((0,T);\, L^2(\Omega)) \quad \text{and} \quad u_3 \in C((0,T);\, H_0^1(\Omega)).$$

Thus $\int_\Omega u_3' u_3^- = -\frac{1}{2} d/dt \int_\Omega (u_3^-)^2$ and $\int_\Omega \partial_{xx} u_3 u_3^- = \int_\Omega |\partial_x u_3^-|^2$.

The first formula above follows from the definition of u_3^- (multiply out $(u_3^-)^2$) and the second formula from well-known facts about $\partial_x u_3^-$ ([5], section 7.4). Once again we restrict ourselves to T_0. By the local existence, there exists a constant $C = C(T_0)$ such that

$$d/dt \int_\Omega (u_2(t)^-)^2 + (u_3(t)^-)^2 \leq 2C \int_\Omega (u_2(t)^-)^2 + (u_3(t)^-)^2$$

it follows that

$$\int_\Omega (u_2(t)^-)^2 + (u_3(t)^-)^2 \leq \int_\Omega (\phi_2^-)^2 + (\phi_3^-)^2.$$

By assumption, $\phi_2 \geq 0$ and $\phi_3 \geq 0$, so $\phi_2^- = 0$ and $\phi_3^- = 0$ for a.e. $x \in \Omega$ and $u_2 \geq 0$, $u_3 \geq 0$ for a.e. $x \in \Omega$. Since $T_0 > 0$ is arbitrary, the result is true for all $t \in [0,T)$.

3. Global existence and precompactness of trajectories

First, let us establish the following preparatory lemma.

Lemma 3.1. Denoting by $\| \ \|_1$ the $L^1(\Omega)$ norm, we have

$$(3.1) \qquad \|u_i(t)\|_1 \leq C, \quad i \in I,$$

$$(3.2) \qquad \int_0^t \| u_j(t)\|_1 \, d\sigma \leq C, \quad j = 3,4.$$

P r o o f . Integrating (1.1) and (1.3) over $Q_t = \Omega \times (0,t)$ and adding up, we find

$$\int_\Omega (u_1 + u_3) + k_3 \int_{Q_t} u_3^2 + (k_5 + 2k_4) \int_{Q_t} u_3 \leq -k_5 \int_{Q_t} u_2 u_3 + \int_\Omega (\phi_1 + \phi_3) \leq C,$$

hence, in particular, (3.1) for $i = 1,3$ and (3.2) for $j = 3$. Moreover, we have

$$(3.3) \qquad \int_{Q_t} u_3^2 \leq C.$$

On the other hand, integrating (1.1) over Q_t, we have

$$\int_\Omega u_1 + k_1 \int_{Q_t} u_1 u_2 \leq \int_\Omega \phi_1 + k_3 \int_{Q_t} u_3^2 \leq C,$$

hence, in particular

$$\int_{Q_t} u_1 u_2 \leq C. \tag{3.4}$$

Now, integrating (1.3) over Q_t, we get

$$\int_\Omega u_3 + (k_2 + k_4) \int_{Q_t} u_3 \leq \int_\Omega \phi_3 + k_1 \int_{Q_t} u_1 u_2,$$

hence, by (3.4)

$$\int_{Q_t} u_3 \leq C. \tag{3.5}$$

Finally, adding up (1.2) and (1.4) and integrating over Q_t, we find

$$\int_\Omega (u_2 + u_4) + k_7 \int_{Q_t} u_4 \leq \int_\Omega (\phi_2 + \phi_4) + k_3 \int_{Q_t} u_3 + 2k_3 \int_{Q_t} u_3^2.$$

Using (3.3) and (3.5), we deduce

$$\int_\Omega (u_2 + u_4) + k_7 \int_{Q_t} u_4 \leq C,$$

so, in particular (3.1) for $i = 2, 4$ and (3.2) for $j = 4$.

Theorem 3.2. Let U be a local solution of (1.1)-(1.6). Then we have

$$u_i \in C_B(R^+; C(\bar{\Omega})), \quad i \in I, \tag{3.6}$$

where $C_B(R^+; C(\bar{\Omega}))$ is the space of continuous bounded functions from R^+ to $C(\Omega)$.

Proof. We are going to show only how to obtain $u_2 \in C_B(R^+; C(\bar{\Omega}))$, because the same analysis works to obtain $u_i \in C_B(R^+; C(\bar{\Omega}))$, $i=1,3,4$. With $w = u_2$ and $\mu = \mu_2$, the equation (1.2) can be written in the following form

$$(3.7) \qquad \partial_t w - (\mu \partial_{xx} - b)w = M(t,x),$$

where $M(t,x) = bu_2(t,x) + F_2(U(t,x))$, and $w(0,x) = \phi_2(x)$. Without loss of generality it may be assumed that $\phi_2 \in C^2(\bar{\Omega})$, for otherwise the initial value problem starting at $t = \delta > 0$ may be considered. There is a constant $C > 0$ such that

$$\|M(t)\|_\infty \leq C, \quad t \geq 0.$$

By [11, p.88], for some b, under homogeneous boundary conditions $\partial_{xx} - bI$ generates an analytic semigroup in $L^p(\bar{\Omega})$ for $p > 1$, and with the associated operator A, there is a $\delta > 0$ such that $\mathrm{Re}\,\sigma(A) > \delta$ (where $\sigma(A)$ denotes the spectrum of A).

The integral solution of (3.7) is

$$(3.8) \qquad w(t) = \exp(-At)\phi_0 + \int_0^t \exp(-A(t-s))M(s)ds.$$

From [7, p.26], for $\sigma > 0$, we have

$$(3.9) \qquad \|A^\sigma \exp(-At)\| \leq C(\sigma)t^{-\sigma} \exp(-\delta t).$$

Taking some $\sigma \in (0,1)$ and applying A^σ to (3.8), we obtain

$$\|A^\sigma w(t)\|_\infty \leq \|A^\sigma \exp(-At)\|_\infty \|\phi_2\|_\infty + \int_0^t \|A^\sigma \exp(-A(t-s))\|_\infty \, ds \leq$$

$$\leq C(\sigma)t^{-\sigma} \exp(-\delta t)\|\phi_2\|_\infty + \max_{0 \leq s \leq t} \|M(s)\|_\infty \int_0^t (t-s)^{-\sigma} \exp(-\delta(t-s))ds,$$

by (3.9). It follows from the bounds on ϕ_2 and M that there is a constant C such that

$$(3.10) \qquad \|A^\sigma w(t)\|_\infty \leq C, \quad (t \geq 1).$$

From the definition of the fractional space $(L^p)^\sigma$ [7, p.29], and a standard imbedding theorem [7, p.39], we have

(3.11) $$\|A^{\sigma} w(t)\| = \|w(t)\|_{(L^p)^{\sigma}},$$

(3.12) $$\|w(t)\|_{C^{\nu}} \leq k\|w(t)\|_{(L^p)^{\sigma}}, \quad 0 \leq \nu \leq 2\alpha - 1/p,$$

where k is independent of w. Taking $\alpha = 1/2$ and $p = 1$, $\nu = 0$ we obtain the result on combining (3.10), (3.11) and (3.12). By the alternative (Alt.), not only the solution exists for all $t > 0$ but also is bounded.

4. Large time behaviour

Lemma 4.1. If $\phi \in C^1(R^+)$, $\phi \geq 0$ is such that

$$\int_0^{\infty} \phi(t)\, dt < \infty \quad \text{and} \quad d/dt(\phi) \leq C < \infty, \ t \geq 0,$$

then $\lim_{t \to \infty} \phi(t) = 0$.

Proof. Let $\varepsilon > 0$, then there is $M = M(\varepsilon) > 0$ such that $\int_M^{\infty} \phi(t) dt \leq \varepsilon$. Let t and $t_e \geq M$ with $t > t_e$. Integrating by parts, we get

$$\int_{t_e}^{t} (s - t_e)\, d/ds(\phi) ds = (t - t_e)\, \phi(t) - \int_{t_e}^{t} \phi(s)\, ds,$$

hence

(*) $$(t - t_e)\, \phi(t) = \int_{t_e}^{t} \phi(s)\, ds + \int_{t_e}^{t} (s - t_e)\, d/ds(\phi)\, ds \leq$$

$$\leq \int_{t_e}^{t} \phi(s)\, ds + \frac{C}{2} (t - t_e)^2,$$

where $C > 0$ is a constant such that $d/ds\, \phi \leq C$. From (*) and choosing t_e such that $t - t_e = \varepsilon^{1/2}$ we obtain

$$(**)\qquad \phi(t)\leqslant \frac{1}{\sqrt{\varepsilon}}\int_M^\infty \phi(s)ds + \frac{C}{2}\frac{1}{\sqrt{\varepsilon}} \leqslant \frac{1}{\sqrt{\varepsilon}}\left(1+\frac{C}{2}\right)$$

hence, $\varepsilon > 0$ given, there is $T_M = M + \frac{1}{\sqrt{\varepsilon}}$ such that (**) holds as soon as $t = t_e + \frac{1}{\sqrt{\varepsilon}} \geqslant M + \frac{1}{\sqrt{\varepsilon}}$.

Theorem 4.1. There holds $\lim_{t\to\infty}\|u_i(t)\|_\infty = 0$, $i \in I$.

Proof. Integrating (1.3) and (1.4) over Ω we get

$$d/dt\int_\Omega u_3 \leqslant k_1\int_\Omega u_1u_2 \quad\text{and}\quad d/dt\int_\Omega u_4 \leqslant 2k_3\int_\Omega u_3^2.$$

As U is bounded, we can write

$$d/dt\int_\Omega u_3 \leqslant C \quad\text{and}\quad d/dt\int_\Omega u_4 \leqslant C.$$

On the other hand, from Lemma 3.1,

$$\int_\Omega u_3 \in L^1(R^+) \qquad\text{and}\quad \int_\Omega u_4 \in L^1(R^+),$$

so, using Lemma 4.2, we obtain

$$(4.1)\qquad \lim_{t\to\infty}\|u_j(t)\|_1 = 0, \qquad j = 3,4.$$

Now multiplying (1.2) by u_2, integrating the result over Ω, we get

$$d/dt\left(\int_\Omega u_2^2\right) + 2\mu_2\int_\Omega (\partial_x u_2)^2 \leqslant 2\int_\Omega (k_3u_3 + k_8u_4)u_2,$$

hence

$$d/dt\left(\int_\Omega u_2^2\right) \leqslant C.$$

On the other hand, integrating (1.2) over $\Omega\times(0,t)$ we can state that $\int_\Omega u_2^2(t) \in L^1(R^+)$. So, by Lemma 4.2, we find

$$(4.2)\qquad \lim_{t\to\infty}\|u_2(t)\|_2 = 0.$$

Finally, multiplying (1.1) by u_1 and integrating the result over Ω, we have

$$(4.3)\qquad d/dt\left(\int_\Omega u_1^2\right) + 2\mu_1\int_\Omega (\partial_x u_1)^2 \leqslant 2k_3\int_\Omega u_3^2 u_1 \leqslant C_\varepsilon \int_\Omega u_3^4 + \varepsilon \int_\Omega u_1^2.$$

Using Poincaré's inequality, choosing ε, and integrating over $(0,t)$, we obtain by (3.3)

$$\int_\Omega u_1^2 + C_0\int_0^t\int_\Omega u_1^2 \leqslant C\int_0^t\int_\Omega u_3^2 + \int_\Omega \phi_1^2 \leqslant C.$$

Hence

$$\int_\Omega u_1^2 \in L^1(R^+).$$

Now, from (4.3), we have $d/dt \int_\Omega u_1^2 \leqslant C$. So, by Lemma 4.2 we get

$$(4.4)\qquad \lim_{t\to\infty} \|u_1(t)\|_2 = 0.$$

Finally, as the trajectories $\bigcup_{t>0}\{u_i(t)\}$, $i \in I$ are precompact in $C(\bar\Omega)$, so (4.1), (4.2) and (4.4) lead to

$$\lim_{t\to 0} \|u_i(t)\|_\infty = 0, \quad i \in I.$$

Theorem 4.2. Let U be a solution of (1.1)-(1.6). Then there exist strict positive constants T, α, K_i, $i \in I$, C_0, C_i with $C_0 < C_i$ such that for any $t \geqslant T$ we have $\|u_i(t)\|_\infty \leqslant K_i \exp(-\alpha t)$, $i = 1,3,4$ and $C_0/t \leqslant \|u_2(t)\|_\infty \leqslant C_1/t$.

Proof. Let

$$A_i := \mu_i \partial_{xx}, \quad i = 1,2,$$

$$A_3 := \mu_3\partial_{xx} - (k_3+2k_4)\cdot Id =: \mu_3\partial_{xx} - \lambda_3 Id,$$

$$A_4 := \mu_4\partial_{xx} - (k_7+k_8)\cdot Id =: \mu_4\partial_{xx} - \lambda_4\cdot Id,$$

where Id := identity. It is simple to show for any $t \geqslant 0$ the following estimates

$$\|\exp(t A_1)\| \leq \exp(-(\pi/1)^2 t),$$

$$\|\exp(t A_2)\| \leq 1,$$

$$\|\exp(t A_i)\| \leq \exp(-\lambda_i t), \quad \text{for} \quad i=3,4.$$

So, if we set $\delta = \mathrm{Min}\left\{(\pi/1)^2, \lambda_3, \lambda_4\right\} > 0$ we will have

(4.5) $$\|\exp(t A_i)\| \leq \exp(-\delta t), \quad i = 1,3,4, \quad \text{for any } t \geq 0.$$

On the other hand, by Theorem 4.1, we state that for any $\varepsilon > 0$ there exists $T > 0$, such that for any $t \geq T$, we have

(4.6) $$\|u_i(t)\|_\infty \leq \epsilon, \quad i \in I.$$

Using (4.6), the positivity of semigroups $\exp(t A_i)$, $i \in I$, and the integral formulation of the solution, we have for any $t \geq 0$

$$u_1(t) \leq \exp((t-T)A_1)\, u_1(T) + \varepsilon k_3 \int_T^t \exp((t-\sigma)A_1) u_3(\sigma)\, d\sigma,$$

$$u_3(t) \leq \exp((t-T)A_3)\, u_3(T) + \varepsilon k_1 \int_T^t \exp((t-\sigma)A_3) u_1(\sigma)\, d\sigma,$$

Now, setting $\psi(t) := \|u_1(t)\|_\infty + \|u_3(t)\|_\infty$ and using (4.5), we have

$$\exp(\delta t)\psi(t) \leq \psi(T) \exp(\delta T) + \varepsilon (k_1 + k_3) \int_T^t \exp(\delta\sigma)\psi(\sigma)\, d\sigma.$$

By Gronwall's lemma, we have

$$\psi(t) \leq \psi(T) \exp(-(t-T)(\delta - \varepsilon(k_1 + k_2))).$$

Hence, for any $t \geq T$

(4.7) $$\|u_i(t)\|_\infty \leq K_i \exp(-\alpha t), \quad i = 1,3,$$

where we set

$$\alpha = \delta/2, \quad K_1 = K_3 = \exp((\delta - \varepsilon(k_1 + k_3))T)\psi(T), \quad \varepsilon = \delta(k_1+k_3)/2.$$

On the other hand, using (4.5), (4.6) and (4.7) yields

$$\| u_4(t)\|_\infty \leq \exp(\delta(t-T))\|u_4(T)\|_\infty + 2k_3k_1 \int_T^t \exp(-\delta(t-\sigma/2))d \quad .$$

Hence, for any $t \geq T$,

$$\|u_4(t)\|_\infty \leq K_4 \exp(-\alpha t). \tag{4.8}$$

Finally, for any $t \geq T$, there holds $\partial_t u_2 - \mu_2 \partial_{xx} u_2 \geq -2k_6 u_2^2$, so, u_2 is a sup-solution of

$$(d/dt)v(t) = -2k_6 v(t)^2, \quad v(T) = \operatorname*{Min}_x u_2(T,x).$$

A simple integration, yields $u_2(t,x) \geq v(t) \geq C_0/t$.

On the other hand, multiplying (1.2) by u_2^m, and integrating over Ω, one can find a constant $\gamma>0$ such that

$$(1/m)x\left\{(d/dt\,\phi(t))/\phi(t)^{(m+1)/m}\right\} \leq -\gamma,$$

where we set $\phi(t) = \int_0^t u^m(t,x)dx$. Integrating this inequality over (t,T), one can find a positive constant C_1 such that $\|u_2(t)\|_\infty \leq C_1/t$ for any $t \geq T > 0$. Hence, we have found constants $T \geq 0$, $C_1 \geq C_0 \geq 0$ such that $C_0/t \leq \|u_2(t)\|_\infty \leq C_1/t$ for any $t \geq T$.

Remarks:

1) If instead of $F_2(U)$, we have $F_2(U) - a\, u_2$, for example, with $a > 0$, then we would have $\|u_2(t)\|_\infty \leq K_4 \exp(-\alpha t)$ for any $t \geq T$ and for certain positive constants K_4, α.

2) It will be interesting to have the same result for a domain Ω in R^2 or in R^3.

3) It will be interesting to have an estimate of the form $\|u_i - \beta_i\|_{L^\infty((0,+\infty;L^2(\Omega))} \leq C(\mu_i)$, $i = 1,2,3,4$, where β_i, $i \in I$, are the solutions of the corresponding ordinary differential system.

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