ON A FREE INTERFACE PROBLEM MODELING SOLID COMBUSTION AND RAPID SOLIDIFICATION IN INFINITE MEDIUM

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ABSTRACT

We study a free interface problem related to combustion of condensed matter and some non-equilibrium exothermal phase transitions. The problem possesses a unique classical solution globally in time. In spite of a variety of non-trivial dynamical scenarios exhibited by the model the solutions are uniformly bounded and the interface velocity is a smooth function of time for arbitrary interface kinetics subject to some natural conditions. The model sustains a basic uniformly propagating wave that looses stability beyond a certain threshold value of the governing parameter resulting in Hopf bifurcation. We present numerical illustrations of complex auto-oscillatory regimes of the interface dynamics.

1 Introduction.

In the present article we study well-posedness and analytical properties of solutions of a free interface problem modeling combustion of condensed matter and some exothermic phase transitions. The most prominent feature of this model is the dynamical wealth exhibited by the interface motion which is due to the kinetic boundary condition reflecting the non-equilibrium character of the process.

We should point out right away this very important distinction from the classical (equilibrium) Stefan problems (see e.g[1]), where the temperature at the interface is given as a constant or as a prescribed function of its position. For the non-equilibrium phase transition the interface temperature is functionally related to its instantaneous velocity. This dependence is the key element of the thermal instability leading to a highly non-trivial dynamics which is, generally speaking, absent in the equilibrium case. In the context of solid combustion, the "phase transition" is represented by an exothermal chemical reaction in a solidified mixture of reactants, that occurs in a very narrow strip identified as flame front (interface), whereas the non-equilibrium condition reflects the dependence of the reaction rate on the temperature.

The ability of an exothermic free boundary to develope a variety of spacio-temporal patterns has been demonstrated numerically[2] for a simple "one-phase" problem., exhibiting a number of nontrivial self-oscillatory regimes as the governing parameter of the system moves into the instability region. The major dynamical scenarios observed in the evolution of the one-phase model include sequences of period doubling followed by chaotic pulsations, the so-called infinite period bifurcations etc.

It turns out that the two-phase problem (the "true" phase transition model, so to speak) that is considered in the present article, is capable of generating the same dynamical events. A detailed numerical evidence based on an entirely different method than in[2] will be presented elsewhere. We should only remark that numerical treatment of the two-phase problem is substantially more difficult due to the presence of the additional temperature field behind the propagating interface (in the product phase).

The dynamical wealth exhibited by the free interface problem motivates its thorough analytical investigation part of which we present below. The principal result establishes the global in time well-posedness for the classical solutions of the two-phase model. Perhaps the most relevant earlier result for our discussion can be found in[3] where the existence of weak solutions for the two-phase problem in bounded domain had been established. Our approach, however, is based on a systematic use of the classical heat potentials and follows rather closely that developed in[1] for the Stefan problem. This approach allows to reduce the free interface problem to the initial value problem for a Volterra type integral equation. It turns out that the non-equilibrium interface condition, while being the source of the non-trivial dynamics, is, in a sense, friendlier for the rigorous mathematical analysis, as it leads to a noticeably simpler integral equation than that for the classical Stefan problem.

It should be remarked that the most important element allowing us to obtain global results is a correct choice of conditions applied to the kinetic functions with which the global well-posedness becomes physically feasible and therefore amenable to mathematical treatment. Moreover, we are able to prove that solutions are *uniformly bounded for all times* no matter how complex (including the chaos) the dynamics of the interface may be. It is worth mentioning, that so far we were unable to obtain the uniform estimate for the same class of kinetics in the conceptually even simpler one-phase problem[4], which turns out to be somewhat less natural and, consequently, leads to a more complex integral representation for the solutions.

Another important analytical property of the solutions demonstrated below is the *smoothness of the interface velocity* that is also done via an appropriate integral equation for the interface acceleration. Higher time derivatives of the interface position function can be obtained in the same manner.

The results discussed below concern only the one-dimensional version of the problem. The two and three-dimensional problems generate an even more complex behavior as the geometry of the interface enters the picture and the non-equilibrium interface condition adds a curvature dependent term. One should not expect global results in this case, at least not for an arbitrary initial configuration of the interface while the governing parameter is in the instability region. We hope to be able to present a more comprehensive discussion of the multi-dimensional case elsewhere in the near future.

The paper is organized as follows. In Section 2 we explain how the free interface problem is related to the solid combustion. In Section 3 we formulate the main global existence and uniqueness result. The reduction of the original problem to the integral equation is carried out in Section 4. In the next two sections we obtain the *a priori* estimate and local existence of solutions for small times which allow to establish global well-posedness. Section 7 deals with the smoothness of the motion.

Finally, in the two last sections we briefly discuss the dynamics generated by the free interface problem and present two numerical examples of non-trivial interface motion. Large amplitude chaotic pulsations of the interface velocity with very sharp maxima illustrate well the necessity to answer such questions as global sustainability of the regime and smoothness of the evolution. Clearly, one can find the answers to this questions only through a rigorous argument.

2 Physical Origin and Formulation of the Problem.

We begin from a brief discussion of the physical origin of the free boundary problem and its formulation. In the context of combustion of condensed matter (known also as high-temperature synthesis) a propagating wave of exothermic chemical reaction transforms a solid combustible mixture directly into solid product. The most primitive one-dimensional model of gasless combustion involves a system of differential equations for the temperature u of the mixture and the relative concentration of the second reactant C (see e.g. Shkadinsky *et al.*[5]). In the one-dimensional formulation it takes the form:

$$\begin{aligned} u_t &= \kappa u_{xx} + qW(C,u), \\ C_t &= -W(C,u), \end{aligned}$$

where κ is the thermal diffusivity (everywhere in the sequel we will assume that units are normalized so that $\kappa = 1$), W is the chemical reaction rate, and q is the heat release.

Normally the process is characterized by a strong temperature dependence of the reaction rate, and by rather sharply defined narrow region where the bulk of chemical reaction and the heat release occur. The latter region is designated as the flame front. This suggests a natural alternative to the above model with distributed reaction which regards the chemical reaction as a point source (so called flame sheet approximation, see Zeldovich *et al.*[6]). The distributed reaction rate is then replaced by the δ-function,

$$W = g(u)\delta(x - s(t))$$

located at the interface $x = s(t)$ between the fresh ($C = 1$) and burnt ($C = 0$) material. One can carry out a formal derivation of the sharp interface model using matched asymptotics (see e.g. Matkowsky & Sivashinsky[7]). As a result of elimination of the concentration the temperature at the interface will depend on its velocity. The equations with the δ-function source are then transformed to the system of two heat equations coupled at the interface.

In the context of solidification of overcooled liquids (see, e.g.[8]) or the amorphous into crystalline transition[9], the free interface model studied below is conceptually even simpler and perhaps better known. The propagation of the product phase is governed by the heat diffusion in both phases. More specifically, the latent heat of the phase transition released at the interface must be diffused into the surrounding matter.

For some substances in the presence of strong supercooling of the original phase the phase transition temperature measured at the interface may deviate considerably from the equilibrium one and is functionally related to the interface velocity. This dependence called the interface attachment kinetics may be different for different substances due to various microscopic mechanisms responsible for the incorporation of the product at the interface into the crystalline lattice.

Therefore, we shall be concerned with the following appropriately non-dimensionalized free boundary problem: find $s(t)$ and $u(x,t)$ such that

$$u_t = u_{xx} - \alpha u, \quad x \neq s(t) \tag{2.1}$$

$$u(x,0) = u_0(x) \geq 0 \tag{2.2}$$

$$u(s(t),t) = g(v(t)) \quad \text{for}\, t > 0, \tag{2.3}$$

$$u_x^+(s(t),t) - u_x^-(s(t),t) = v(t) \quad \text{for}\, t > 0, \tag{2.4}$$

where $v(t)$ is the interface velocity, $s(t) = \int_0^t v(\tau)d\tau$ is its position, u is the temperature, and the derivatives u_x^+ and u_x^- are taken from right side and left side of the free interface respectively.

The last term in the heat equation 2.1 is due to the heat losses (that are sometimes present) into the medium surrounding the combustible or solidifying substance via Newton's cooling law with a non-dimensional coefficient $\alpha \geq 0$. The surrounding matter is assumed to be at the temperature of the fresh combustible mixture at $-\infty$ (the original phase in the phase transition interpretation). It should be remarked that the presence of the heat losses $\alpha > 0$ effects only positively the analytical properties of the solutions.

3 Global Existence and Uniqueness.

In this section we formulate the main result of the paper: global existence and uniqueness of uniformly bounded classical solutions of the free interface problem 2.1-2.4. The proof is carried out via the following scheme. First, the problem is reduced to a Volterra type integral equation for the interface velocity. Next, we establish a global uniform *a priori* estimate for the solutions of the integral equation. After that we show that for sufficiently small time intervals, the integral equation defines a contraction on an appropriately chosen closed set of continuous functions which yields local existence. The proof of the local existence closely parallels an analogous argument for the one-phase free boundary problem considered in[4], therefore many details of the proof are only sketched. The crucial ingredient of the global existence proof is the a priori estimate, it allows us to continue the local solution indefinitely.

Therefore, the result is as follows:

Theorem 1 *Suppose that the kinetic functions g satisfies the following assumptions:*

(A1) $g^{-1}(u)$ *is a continuously differentiable, monotone decreasing, negative function on* $(0, \infty)$ *with* $g^{-1}(0) = -v_0$ *for some velocity* $-v_0 < 0$;

(A2) $g^{-1}(u)$ *is sublinear:* $\lim_{u\to\infty} g^{-1}(u)/u = 0$;

and that the following conditions hold for the initial data $u_0(x)$*:*

(1) $u_0(x)$ *is bounded and has a continuous bounded derivative,*

(2) $\dfrac{\partial u_0}{\partial x}$ *approaches* 0 *at* $\pm\infty$ *and is consistent with the boundary conditions at* $x = 0$,

$$\frac{\partial u_0^+}{\partial x}(0) - \frac{\partial u_0^-}{\partial x}(0) = g^{-1}u_0(0).$$

Then there exists one and only one classical solution of the free interface problem 2.1-2.4. The solution is uniformly bounded for all $t > 0$.

[We say that $u(x,t), v(t)$ form a *classical solution* of 2.1-2.4 if (i) $u(x,t)$ and $v(t)$ are continuous for $t \geq 0$; (ii) u_{xx} and u_t are continuous for $x \neq s(t),\ t > 0$; (iii) Eqs. 2.1-2.4 are satisfied.]

Remark 1 The above results with only minor modifications of the proofs can be obtained for a somewhat more general jump condition than the Stefan condition 2.4:

$$u_x^+(s(t),t) - u_x^-(s(t),t) = h(v(t)) \quad \text{for t} > 0,$$

where $h(v)$ is a monotone Lipshitz continuous function with Lipshitz continuous inverse.

4 The integral equation.

In this section the problem is reduced to solving an integral equation for the interface velocity. First, it is convenient to get rid of the heat losses term. Let $w(x,t) = e^{\alpha t}u(x,t)$. In terms of w the problem takes the form

$$w_{xx} = w_t, \quad x \neq s(t) \tag{4.5}$$

$$w(x,0) = w_0(x) = u_0(x) \geq 0, \tag{4.6}$$

$$w(s(t),t) = e^{\alpha t} g(v(t)) \quad \text{for}\, t > 0, \tag{4.7}$$

$$w_x^+(s(t),t) - w_x^-(s(t),t) = e^{\alpha t} v(t) \quad \text{for}\, t > 0. \tag{4.8}$$

Let G be the fundamental solution of the heat equation

$$G(x,t,\xi,\tau) = \exp\{-\frac{(x-\xi)^2}{4(t-\tau)}\}[4\pi(t-\tau)]^{-1/2}. \tag{4.9}$$

Then, the solution of the of the Cauchy problem 4.5-4.6 with the matching condition 4.8 along a *given* interface $s(t)$ is well known:

$$w(x,t) = \int_{-\infty}^{\infty} G(x,t,\xi,0) w_0(\xi) d\xi - \int_0^t G(x,t,s(\tau),\tau) e^{\alpha\tau} v(\tau) d\tau, \tag{4.10}$$

Recalling the definition of w we obtain:

$$u(x,t) = e^{-\alpha t} \int_{-\infty}^{\infty} G(x,t,\xi,0) u_0(\xi) d\xi - \int_0^t G(x,t,s(\tau),\tau) e^{-\alpha(t-\tau)} v(\tau) d\tau, \tag{4.11}$$

The integral representation for the solution u with the heat source of constant density along the interface $s(t)$ is physically quite transparent. We, however, do not know the location of the interface. Instead we have the additional condition 2.3 that "overdefines" the problem. Thus, taking the limit of 4.11 as $x \to s(t)$ and using the kinetics condition 2.3, we are able to obtain the following reduction of the original problem to an integral equation in terms of v only:

$$g(v(t)) = e^{-\alpha t} \int_{-\infty}^{\infty} G(s(t),t,\xi,0) u_0(\xi) d\xi - \int_0^t G(s(t),t,s(\tau),\tau) e^{-\alpha(t-\tau)} v(\tau) d\tau, \tag{4.12}$$

where $s(t) = \int_0^t v(\tau)d\tau$.

Indeed we have already shown that every solution u, s of 2.1-2.4 satisfies 4.12. Conversely, let v be a continuous solution of 4.12. Then, it is not difficult to show that $v(t)$ and $u(x,t)$ defined by 4.11 form the solution of the original problem. First, it is quite obvious that 2.1-2.3 hold. Secondly, we differentiate $u(x,t)$ with respect to x and take $x \to s(t) - 0$ and $x \to s(t) + 0$ respectively. Then, using the well-known jump property of the derivative of the heat potential we obtain 2.4.

5 *A Priori* Estimate

In this section we establish global boundedness of the classical solutions of the free boundary problem. In view of the fact that it is in a sense much less predictable a property than the local existence we present a detailed proof.

Theorem 2 *Let $u(x,t), v(t)$ be a classical solution of 2.1-2.4 and $\sup_{-\infty<x<\infty}|u_0(x)| = M$, then*

$$|u(x,t)| \le 2(M + \frac{v_1}{v_0}), \tag{5.13}$$

where v_1 is a constant dependent on the kinetic function g.

Proof. First we prove the estimate for the temperature at the interface: $\psi(t) = g(v(t)) = I_1 - I_2$, where I_1 and I_2 are the two parts of the right hand side of 4.12.

It is rather obvious that $|I_1| \le M$. Now, since the kinetic function satisfies the condition (A2), for any $N > 0$ there exists $v_1 > 0$ such that $|g^{-1}(\psi)/\psi| \le N$ if $g^{-1}(\psi) \le -v_1$We subdivide I_2 as follows:

$$\begin{aligned} I_2 &= \int_{\chi_1} G(s(t), t, s(\tau), \tau)e^{-\alpha(t-\tau)}g^{-1}(\psi(\tau))d\tau \\ &+ \int_{\chi_2} G(s(t), t, s(\tau), \tau)e^{-\alpha(t-\tau)}g^{-1}(\psi(\tau))d\tau = I_3 + I_4, \end{aligned} \tag{5.14}$$

where $\chi_1 = \{\tau| - v_1 < g^{-1}(\psi(\tau)) < -v_0, 0 < \tau < t\}$ and $\chi_2 = (0,t) \setminus \chi_1$.

For I_3 we have:

$$\begin{aligned} & |\int_{\chi_1} G(s(t), t, s(\tau), \tau)g^{-1}(\psi(\tau))d\tau| \\ \le\ & v_1|\int_0^t G(s(t), t, s(\tau), \tau)d\tau| \\ =\ & v_1|\int_0^t \exp\{-\frac{(s(t)-s(\tau))^2}{4(t-\tau)}\}(2\sqrt{\pi(t-\tau)})^{-1}d\tau| \\ \le\ & v_1|\int_0^\infty -\frac{2}{v_0\sqrt{\pi}}\exp\{-v_0^2\frac{t-\tau}{4}\}d(\frac{v_0\sqrt{t-\tau}}{2})| = \frac{v_1}{v_0}. \end{aligned} \tag{5.15}$$

Here we have used the observation that $|s(t) - s(\tau)| = |v(\xi)|(t-\tau) \geq v_0(t-\tau)$ for some $\tau \leq \xi \leq t$.

Now, let us interpret $I_4(t) = P\psi(t)$ as a mapping; then, for its norm the following estimate holds:

$$\begin{aligned} \|P\psi\| &\leq |\int_{\chi_2} G(s(t),t,s(\tau),\tau)g^{-1}(\psi(\tau))d\tau| \\ &\leq |\int_{\chi_2} G(s(t),t,s(\tau),\tau)N\psi(\tau)d\tau| \\ &\leq N\|\psi\|\,|\int_0^\infty G(s(t),t,s(\tau),\tau)d\tau| \leq \frac{N}{v_0}\|\psi\|. \end{aligned} \tag{5.16}$$

Since $\psi = I_1 - I_3 - P\psi$, we have $\|\psi + P\psi\| = |I_1 - I_3| \leq M + v_1/v_0$. On the other hand, choose $N = v_0/2$, then have $\|P\| \leq \frac{1}{2}$, and therefore $|\psi(t)| \leq 2(M + v_1/v_0)$.

Finally, by the maximum principle we are able to extend the uniform *a priori* estimate to the field variable

$$\begin{aligned} |u(x,t)| &\leq \max\{\|u_0\|, |u(s(t),t)|\} \\ &= \max\{\|u_0\|, |\psi(t)|\} \leq 2(M + \frac{v_1}{v_0}). \end{aligned} \tag{5.17}$$

■

6 Local Existence.

We continue to carry out the scheme outlined at the beginning of Section 3. It is convenient to rewrite 4.12 as follows:

$$v(t) = g^{-1}\left[e^{-\alpha t}\int_{-\infty}^{\infty} G(s(t),t,\xi.0)u_0(\xi)d\xi - \int_0^t G(s(t),t,s(\tau),\tau)e^{-\alpha(t-\tau)}v(\tau)d\tau\right] \tag{6.18}$$

Let Kv denote the action of the right hand side of 6.18. Our next step is to show that

Proposition 1 *Transformation $\omega = K\varphi$ is a contraction of an appropriate subset of $C[0,\sigma]$ for some σ and therefore has a unique fixed point $v = Kv$.*

Proof. In the Banach space $C_\sigma = C[0,\sigma]$ with uniform norm we consider the closed set

$$B_{M,\sigma} = \{\varphi \in C_\sigma,\ \varphi \leq -v_0\ |,\ \|\varphi\| = \sup_{0\leq t\leq\sigma} |\varphi| \leq M\}$$

with M to be specified later on. First we prove that the operator K maps the set $B_{M,\sigma}$ into itself. Indeed, for any $\varphi \leq 0$ the expression in the brackets on the right hand side of 6.18 is non-negative, then, in view of the property $(A1)$ of the kinetics g we see that $\omega \leq -v_0$.

Also, since $|\varphi(\tau)| < M$, we have

$$\|g(K\varphi)\| \leq \|u_0\| + M|\int_0^t G(s(t),t,s(\tau),\tau)d\tau| \leq \|u_0\| + \frac{2M}{\sqrt{2\pi}}\sqrt{\sigma}. \tag{6.19}$$

If now M is taken to be $M = \max\{2\sup|u_0(x)|, N\}$, where N is a large enough number such that $|g^{-1}(u)| \leq |u|$ for $|u| \geq N$ (property $A2$ of g), and $\sigma \leq \pi/8$, then $||g(K(\varphi)|| \leq M$. Assumption $\omega > M \geq N$ leads to contradiction, since, then $|\omega| \leq |g(\omega)| \leq M$. Thus, the set $B_{M,\sigma}$ is mapped into itself.

Now we sketch the prove that K is a contraction on $B_{M,\sigma}$. Let $\omega = K\varphi$ and $\omega' = K\varphi'$, then

$$\begin{aligned} |\omega - \omega'| &\leq L_1|e^{-\alpha t}\int_{-\infty}^{\infty}[G(s,t,\xi,0) - G(s',t,\xi,0)]u_0(\xi)d\xi \\ &+ \int_0^t[G\varphi' - G\varphi]e^{-\alpha(t-\tau)}d\tau| = L_1|W_1 + W_2|. \end{aligned} \tag{6.20}$$

where L_1 is the Lipschitz constant for g^{-1}.

It is not difficult to show that

$$|G(s(t),t,s(\tau),\tau) - G(s'(t),t,s'(\tau),\tau)| \leq C\|\varphi - \varphi'\|(t-\tau)^{1/2} \tag{6.21}$$

for some constant $C > 0$. This allows to estimate the second term in 6.20:

$$\begin{aligned} |W_2| &= |\int_0^t \Delta G\varphi e^{-\alpha(t-\tau)}d\tau \\ &+ \int_0^t G(s'(t),t,s'(\tau),\tau)(\varphi' - \varphi)e^{-\alpha(t-\tau)}d\tau| \\ &\leq A_1\|\varphi - \varphi'\|t^{3/2}\frac{2}{3}V_0 + A_2\|\varphi - \varphi'\|t^{1/2} \\ &= (A_3t^{3/2} + A_4t^{1/2})\|\varphi - \varphi'\|. \end{aligned} \tag{6.22}$$

The estimation for the first term in 6.20 is similar although a little more involved. Assuming $s(t) < s'(t)$ we split the integral for W_1 into three integrals:

$$W_1 = e^{-\alpha t}(\int_{-\infty}^{s} \delta G u_0 d\xi + \int_{s}^{s'} \delta G u_0 d\xi + \int_{s'}^{\infty} \delta G u_0 d\xi) \tag{6.23}$$

where $\delta G = G(s(t),t,\xi,0) - G(s'(t),t,\xi,0)$ and estimate them separately. The result will be

$$|W_1| \leq A_5t^{1/2}\|\varphi - \varphi'\| \, \|u_0\|. \tag{6.24}$$

Combination of 6.22 and 6.24 yields the following contraction estimate

$$\begin{aligned} \|K\varphi - K\varphi'\| &\leq A_3\sigma^{3/2}\|\varphi - \varphi'\| + A_4\sigma^{1/2}\|\varphi - \varphi'\| \\ &+ A_5\sigma^{1/2}\|\varphi - \varphi'\| \, \|u_0\|, \end{aligned} \tag{6.25}$$

where all the constants A_3, A_4 and A_5 depend only on bounds and Lipschitz constants of g^{-1} . If $\sigma < 1$ and such that $\sigma^{1/2}[A_3 + A_4 + MA_5] < 1$, then K is a contraction on $B_{M,\sigma}$. Therefore, K has a unique fixed point $v(t)$ in $B_{M,\sigma}$. ■

It is easy to show (see Friedman[1]) that any solution of the integral equation 4.12, regardless of whether it is bounded by M or not, must coincide with v in their common interval of existence. We have proved the existence and uniqueness of solutions $v(t)$ of the integral equation 4.12 for $0 \leq t < \sigma$. If $t_1 < \sigma$ then by repeating the argument with the initial data $u_0(\xi) = u(\xi, t_1)$ (where u is calculated according to 4.11) we can construct a solution of the integral equation for $t_1 \leq t \leq t_2$.

Both solutions for $0 \leq t \leq t_1$ and for $t_1 \leq t \leq t_2$ generate solutions of the free boundary problem 2.1-2.4 on these intervals. It can be shown than that they are glued together smoothly with respect to t across the line $t = t_1$. Thus, we obtain a unique solution defined for $0 \leq t \leq t_2$. This process can be repeated indefinitely. Thanks to the uniform *a priori* estimate, we can choose an appropriate the time step, $\frac{2}{3}\sigma$ for instance, to obtain the global existence and uniqueness.

7 Smoothness of the Interface Motion: the Acceleration Equation.

In this section we sketch the proof of the differentiability of the interface velocity. Normally results of this type are based on the regularity of the field variable and a rather technically involved analysis of the limits of appropriate derivative from the interior of the domain (see e.g.[10] where smoothness of the free interface for the classical Stefan problem is established). We, however, are able to fully utilize the reduction of the original problem 2.1-2.4 to the integral equation 4.12 for the interface velocity.

The basic idea of the proof is to obtain an integral equation for the acceleration of the interface and to show that its solutions are the true accelerations for the solutions of 2.1-2.4. Therefore, differentiation with respect to t of Eq.4.12 requires justification. We note that a straightforward differentiation with respect to time gives rise to a non-integrable singularity in the integral, therefore we modify it as follows. *Assuming* that he velocity is differentiable the differentiation in the integral can be switched to the velocity and we derive an integral equation for the acceleration $a(t) = \dot{v}(t)$.

Next, considering the acceleration equation independently we show that it generates a contraction mapping in the space of continuous functions on a sufficiently short time interval. This yields local existence and uniqueness for the acceleration equation. Finally, we are able to demonstrate that the integral of the acceleration obtained as the solution of the acceleration equation must, in fact, coincide with the interface velocity thus concluding the regularity proof.

The acceleration equation is derived in the following

Proposition 2 *Let $v(t)$ be a continuously differentiable solution of the integral equation 4.12, $s(0) = 0$. Let the initial data u_0 be twice differentiable in $x < 0$ and $x > 0$ with bounded derivatives and satisfy the matching conditions:*

$$g(v(0)) = u_0(0) \tag{7.26}$$

(which is also a matching condition for the original integral equation 4.12) and

$$v(0) = \frac{\partial u_0^+}{\partial x}(0) - \frac{\partial u_0^-}{\partial x}(0) \equiv v_0 \tag{7.27}$$

Then the acceleration a satisfies the following integral equation:

$$\begin{aligned} g'(v)a &= e^{-\alpha t}\int_{-\infty}^{\infty} G(u_{0_{\xi\xi}} + v(t)u_{0_\xi} - \alpha u_0)d\xi \\ &+ \int_0^t G_\xi[v(t) - v(\tau)]e^{-\alpha(t-\tau)}v(\tau)d\tau \\ &- \int_0^t Ge^{-\alpha(t-\tau)}(a(\tau) + \alpha v(\tau))d\tau \end{aligned} \tag{7.28}$$

where $v(t) = v(0) + \int_0^t a(\tau)d\tau$, $s(t) = \int_0^t v(\tau)d\tau$; in all the temporal integrals the arguments of G are $s(t), t, s(\tau), \tau$. Any solution of 7.28 automatically satisfies the initial conditions:

$$g'(v)a|_{t=0} = \frac{1}{2}(u^+_{0_{\xi\xi}}(0) + u^-_{0_{\xi\xi}}(0)) + \frac{1}{2}v(0)(u^+_{0_\xi}(0) + u^-_{0_\xi}(0)) - \alpha u_0(0) \tag{7.29}$$

Proof. First we note that

$$\begin{aligned} &\frac{d}{dt}\int_{-\infty}^{\infty} G(s(t), t, \xi, 0)u_0(\xi)d\xi = \int_{-\infty}^{\infty}(\dot{s}G_x + G_t)u_0 d\xi \\ = \;&\dot{s}(t)\int_{-\infty}^{\infty} Gu_{0_\xi}d\xi + \int_{-\infty}^{\infty} Gu_{0_{\xi\xi}}d\xi + G(s(t), t, 0, 0)(u^+_{0_\xi} - u^-_{0_\xi}) \end{aligned} \tag{7.30}$$

The above result follows from smoothness of G via a simple integration by parts.

The differentiation of the temporal integral in 4.12 is a little more involved. One can form a difference quotient for the time derivative and through a change of variables transfer the quotient from G to the velocity (which is assumed to be differentiable). Then one can employ the Lebesgue dominated convergence to pass to the limit in the difference quotient. We then obtain the following expression for the time derivative of the temporal integral:

$$\frac{d}{dt}\int_0^t G(s(t),t,s(\tau),\tau)e^{\alpha\tau}v(\tau)d\tau = \int_0^t G(s(t),t,s(\tau),\tau)(\alpha v(\tau)+\dot{v}(\tau))e^{\alpha\tau}d\tau$$
$$-\int_0^t G_\xi(s(t),t,s(\tau),\tau)[\dot{s}(t)-\dot{s}(\tau)]e^{\alpha\tau}v(\tau)d\tau + G(s(t),t,0,0)v(0). \qquad (7.31)$$

Therefore, differentiating 4.12 and applying 7.30, 7.31 combined with the matching condition 7.27 we arrive to the integral equation 7.28.The initial condition 7.29 follows from the easily seen fact that for any Lipschitz continuous $s(t)$ with $s(0) = 0$ and for any piece-wise continuous ϕ $\lim_{t\to 0}\int_{-\infty}^{\infty} G(s(t),t,\xi,0)\phi(\xi)d\xi = \frac{1}{2}[\phi(0-)+\phi(0+)]$. ■

The next step in the scheme outlined at the beginning of this section: the local existence for the acceleration equation is rather technically involved but it follows the same routine as that for the velocity equation. One can show that $g'^{-1}K$ (where K is the action of the right hand side of 7.28) is a contraction for small times on an appropriate closed subset in the space of continuous functions. This leads to the following result:

Theorem 3 *Let the condition (A1) hold and, additionally, the derivative of the inverse function $(g^{-1})'$ be Lipschitz continuous. Then the solution of the velocity equation 4.12 is differentiable.*

Proof. Let $a(t)$ and $v(t)$ be solutions of the acceleration equation 7.28 and the velocity equation 4.12 respectively. Then we can reverse the procedure of derivation of 7.28 and show that $\tilde{v}(t) = v_0 + \int_0^t a(\tau)d\tau$ satisfies 4.12 up to a constant:

$$\begin{aligned} g(\tilde{v}(t)) &= e^{-\alpha t}\int_{-\infty}^{\infty} G(s(t),t,\xi,0)u_0(\xi)d\xi \\ &- \int_0^t G(x,t,s(\tau),\tau)e^{-\alpha(t-\tau)}\tilde{v}(\tau)d\tau + C \end{aligned} \qquad (7.32)$$

As $t \to 0$ the time integral vanishes while it is easy to show that the spatial integral approaches $u_0(0)$ which, in view of the matching condition 7.26 yields $C = 0$. Thus, $\tilde{v}$ satisfies the velocity equation 4.12. Then, in view of uniqueness for 4.12, $v = \tilde{v}$. ■

We conclude this section by a remark concerning higher order derivatives of the velocity. The acceleration equation can be dealt with exactly in the same fashion as with the velocity equation. Namely, it can be "differentiated" to yield an integral equation for the second derivative of the velocity. The estimates sufficient for the solvability of the corresponding integral equation can be obtained if $(g^{-1})''$ is Lipschitz continuous.

8 Basic Propagation Mode and Its Stability.

In this section we briefly discuss the basic propagation mode generated by the model and the loss of stability beyond a certain threshold value of the governing parameter. The instability results in Hopf bifurcation and leads to a variety of non-trivial self oscillatory regimes exhibited by the interface evolution. For simplicity we assume that the heat losses are absent ($\alpha = 0$) and rewrite the non-equilibrium interface condition 2.3 in the form:

$$u(s(t)) = 1 + \nu K(v(t)). \tag{8.33}$$

We shall assume that the function $K(v) = g(v) - 1$ is normalized in such a way that

$$K(-1) = 0, \qquad K'(-1) = -1, \tag{8.34}$$

which can be achieved by rescaling the variables.

In order to clarify the meaning of the (positive) parameter ν it will suffice to mention that for the Arrhenius type kinetics ν is related to the activation energy of the exothermic chemical reaction that occurs at the interface. Note, however, that the global existence and boundedness of the solutions are absolutely independent of the parameter ν.

Assuming a traveling wave propagation one can easily find that the problem has a unique (basic) solution in the form of the exponential profile:

$$u_b = \begin{cases} exp(x+t), & x \le -t \\ 1, & x > -t \end{cases}, \qquad s_b = -t \tag{8.35}$$

provided that K is monotone. Moreover, it is not difficult to verify that the basic solution 8.35 becomes unstable when the parameter ν decreases below a certain threshold value. A routine linear stability analysis of the basic solution yields the following dispersion relationship[7]:

$$4\nu^2\omega^2 + (\nu^2 + 4\nu - 1)\omega + \nu = 0 \tag{8.36}$$

where the perturbations of the basic solutions are of the form $v - v_b = Ae^{\omega t}, \quad u - u_b = W(x)Ae^{\omega t}$.

Therefore, the instability develops as ν decreases below the threshold value $\nu_{cr} = \sqrt{5} - 2$ and a pair of complex conjugate eigenvalues crosses the imaginary axis. This is the well known condition for the Hopf bifurcation to occur. One can easily check that another well-known condition for a non-degenerate Hopf bifurcation is also satisfied

$$\frac{\partial Re\omega}{\partial \nu}(\nu_{cr}) < 0 \tag{8.37}$$

(or, in other words, the two conjugate eigenvalues cross the imaginary axes with non-zero speed). It can be shown that the continuous spectrum of the linearized problem is contained within a parabola in the negative half-plane $Re\omega < -a(Im\omega)^2, \quad a > 0$, while the origin $\omega = 0$ is an accumulation point for the spectrum.

Furthermore, if the problem is considered in certain functional spaces with exponentially growing weights, the continuous spectrum can be separated from the origin and moved into the strictly negative half plane. This allows to separate the subspaces corresponding to the continuous and the discrete parts of the spectrum and to prove a version of the Hopf theorem. For details the reader is referred to[11] or[12] where a more detailed treatment of the Hopf bifurcation for the one-phase model is presented. However, for some kinetics the Hopf bifurcation appears to be subcritical, and the corresponding branch of periodic solutions is unstable which leads to entirely different types of dynamics (see below).

9 Numerical illustrations.

The rigorous analysys presented above serves an important purpose of providing a solid basis for the numerical simulation of the free interface problem 2.1-2.4. As we have mentioned in the introduction the two-phase model exhibits a variety of complex auto-oscillatory regimes. The oscillations sometimes have a strongly manifested relaxational character with sharp spikes and velocity variations of significant magnitude. Thus, the uniform global boundedness and smoothness of solutions demonstrated rigorously give an additional measure of confidence in the numerical results obtaining which is not entirely a routine matter for a two-sided free interface problem.

Figure 1: Chaotic pulsations of the interface velocity

Remarkably, the rigorous analysis in our case turns out also to be extremely helpful for the numerical simulations in a direct way: by providing us with a rather efficient numerical approach to the problem. It should be noted that the two-phase problem 2.1-2.4 is much less convenient for the numerical treatment than its one-phase counterpart. Although we have developed a finite-differences based numerical code similar to the one used in[2], it proved to be rather time consuming and not sufficiently robust. However, if one uses directly the contracting property of operator in 6.18 to calculate the velocity of the interface the situation improves considerably. A detailed discussion of the numerical simulation of the two-phase problem will be presented elsewhere.

We present just two illustrations of the interface motion generated by problem 2.1-2.4. Figure 1 demonstrates well developed chaotic pulsations of the interface velocity. This state corresponds to the supercritical Hopf bifurcation and a sequence of period doubling precedes the fully chaotic motion as the governing parameter ν is moved deeper into the instability region.

Figure 2: Interface velocity vs. time: infinite period bifurcation

Figure 2 depict the interface velocity profile $v(t)$ corresponding to a drastically different dynamical scenario than the one described above. Here the Hopf bifurcation is subcritical and the simple near harmonic oscillations branching from the basic solution just past the stability threshold are unstable, hence, cannot be observed. Consequently, the period doubling cascade leading to chaos does not occur either.

Instead we have a behavior strongly reminiscent of what is known as infinite period bifurcation for the finite dimensional dynamical systems. The solution is strictly periodic with a prolonged relatively small-amplitude "accumulation phase" where the velocity oscillates only slightly about that of the basic solution. The accumulation phase is followed by a strongly manifested deceleration-acceleration in which the interface velocity first drops to a small fraction of the uniform propagation, then increases significantly beyond that and finally returns to a near basic state which completes the cycle. In this infinite period bifurcation scenario the total period tends to infinity as ν approaches the stability range of the basic solution.

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