

A MATHEMATICAL MODEL FOR NON ISOTHERMAL CRYSTALLIZATION

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In this paper we will consider heat conduction in composite materials in the presence of partial crystallization. Consider for example the case of a volcanic magma, or of a polymer. It is known that when the temperature is in a given range  $\theta \in (\theta_g, \theta_m)$ , then nucleation occurs throughout the material, i.e. elementary crystals appear of approximately spherical shape and of radius  $r_0$ , ranging from  $10^{-2}$  microns to some microns, depending on the material and on the temperature.

Then, each of these crystals starts growing with a radial speed depending on temperature until it hits another growing crystal. In this case, both crystals stop growing on the contact surface. If the material is kept at a temperature  $\theta \in (\theta_g, \theta_m)$  for a sufficiently long time, a fraction  $w_m$  of it will crystallize, while the remaining fraction  $1-w_m$  will remain in a glassy (amorphous) state. The maximum crystallinity  $w_m$ , as well as  $r_0$ , the rate of nucleation  $\dot{N}_0$  (average number of nuclei appearing per unit time and unit volume in a completely melted material) and the radial growth speed  $\dot{R}_0$  depend on temperature. For the sake of simplicity we assume  $w_m$  constant throughout this note.

Crystallization influences heat conduction, since the change of phase is associated with the release of a latent heat  $L$ . We will start by modelling isothermal crystallization to make our exposition more clear.

The key point is to take the effect of impingement into account. To this end we first follow the classical argument of Kolmogorov [3] and Avrami [2]. We assume  $r_0=0$ ,  $w_m=1$ , and we confine our attention to the one dimensional case, since the reasoning can be easily repeated in  $n$  dimensions.

Consider points  $P_0 \equiv (x_0, t_0)$  and  $P_1 \equiv (x_0, t_1)$  in the half plane  $\mathbb{R} \times \mathbb{R}^+$  and assume that no crystals are present at  $t=0$  and that nucleation and growth rates are constant.

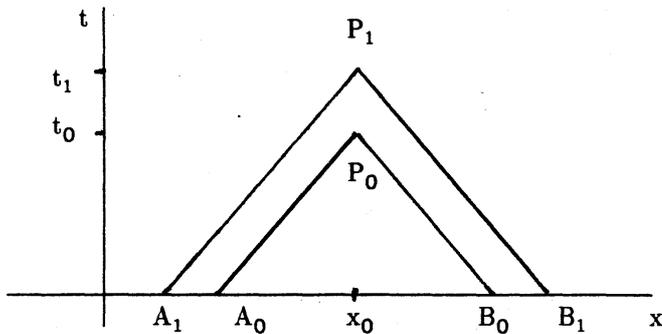


fig. 1

If point  $x_0$  is at amorphous state at time  $t=t_0$  and in crystalline state for  $t=t_1$  it means that no nucleation has occurred in the triangle  $A_0B_0P_0$  in the event plane (see fig. 1), but at least one nucleation occurred in  $A_1B_1P_1$ .

Thus (neglecting higher order terms) this probability of transition is  $\dot{N}_0$  times the difference of the areas of the two triangles.

The probability of having crystallization at  $x_0$  between  $t_0$  and  $t_1$  is the product of the probability of transition times the probability  $1-w(t_0)$  that  $x_0$  is still glassy at time  $t=t_0$ .

Hence

$$(1.1) \quad w(t_1) - w(t_0) = (1 - w(t_0)) \dot{N}_0 \dot{R}_0 (t_1^2 - t_0^2) + o(t_1 - t_0).$$

Dividing by  $t_1 - t_0$  and letting the difference tend to zero we obtain

$$(1.2) \quad \dot{w} = 2(1-w) \dot{N}_0 \dot{R}_0 t,$$

i.e.

$$(1.3) \quad w(t) = 1 - \exp[-\dot{N}_0 \dot{R}_0 t^2],$$

while for 2 and 3 space dimension the analogous of (1.2) and (1.3) are

$$(1.4) \quad \dot{w} = \omega_n (1-w) \dot{N}_0 \dot{R}_0^n t^n,$$

and

$$(1.5) \quad w = 1 - \exp\left[-\frac{\omega_n}{n+1} \dot{N}_0 \dot{R}_0^n t^{n+1}\right],$$

where  $\omega_n$  is the volume of the unit ball in  $\mathbb{R}^n$ ,

$$(1.6) \quad \omega_n = \begin{cases} 2, & \text{for } n=1, \\ \pi, & \text{for } n=2, \\ \frac{4}{3}\pi, & \text{for } n=3. \end{cases}$$

The argument relies heavily on the constancy of  $\dot{R}_0$ ,  $\dot{N}_0$  (and also on the assumption  $r_0=0$ ) even if it can be generalized to cover cases in which  $\dot{R}_0$  depends on the "age" of the crystal and so on (see [5], [6]).

Consequently, applying (1.4) and (1.5) to non-isothermal processes is a nontrivial problem which is often circumvented using the so-called additivity rule approximation (see e.g. [7]).

Our approach is to introduce an equivalent growth rate which incorporates the effect of impingement assuming that it is a function of the crystallinity  $w$ , decreasing monotonically from  $\dot{R}_0$  to zero as  $w$  grows from 0 to  $w_m$ .

Also the nucleation rate is assumed to depend in a similar way on  $w$ , the linear case  $\dot{N} = \dot{N}_0(1 - \frac{w}{w_m})$  being the simplest approximation which has an obvious geometrical interpretation, at

least when  $w_m=1$ .

Set

$$(1.7) \quad \delta = 1 - w/w_m ,$$

$$(1.8) \quad \dot{N} = \dot{N}_0 f(\delta) ,$$

$$(1.9) \quad \dot{R} = \dot{R}_0 g(\delta) ,$$

with  $f(1)=g(1)=1$  ,  $f(0)=g(0)=0$  ,  $f'>0$  ,  $g'>0$ .

Thus the crystallization kinetics in the isothermal case is described by

$$(1.10) \quad w(t) = \int_0^t \dot{N}_0 f(\delta(\tau)) \omega_n \left( r_0 + \int_{\tau}^t \dot{R}_0 g(\delta(s)) ds \right)^n d\tau ,$$

yielding the following integro-differential equation

$$(1.11) \quad -w_m \dot{\delta}(t) = \omega_n r_0^n \dot{N}_0 f(\delta(t)) + n\omega_n \dot{R}_0 g(\delta(t)) \int_0^t \dot{N}_0 f(\delta(\tau)) \left( r_0 + \int_{\tau}^t \dot{R}_0 g(\delta(s)) ds \right)^{n-1} d\tau .$$

$$(1.12) \quad \delta(0) = 1 .$$

Let us note that this formulation can include in a natural way the variation of  $\dot{N}_0$ ,  $r_0$  and  $\dot{R}_0$  with temperature (see below); furthermore it is expected that functions  $f(\delta)$  and  $g(\delta)$  can take into account not only geometrical effects but also other factors (segregation of molecules of low molecular weight, lamellar structure, etc.).

As far as just the geometrical effects are concerned and  $w_m=1$  , as in Avrami's model, we already noted that  $f(\delta)=\delta$ . Concerning  $g(\delta)$ , numerical simulation and comparison with (1.5) suggest a form  $g(\delta)=\delta^q$  ,  $q$  being approximately equal to 0.7 for  $n=1$  (see [1]).

So far melting has received less attention than solidification. But our model allows to take into account melting effects by a simple extension of the ideas described above. In Section 2 we prove existence and uniqueness of the solution to this scheme.

We can expect that for  $\theta > \theta_m$  (and  $\delta < 1$ ) melting takes place at a positive rate, depending on temperature only. Of course the melting rate vanishes for  $\theta = \theta_m$  and/or  $\delta = 1$  . Hence a possible extremely simple choice could be  $\delta = \min(\delta_1, 1)$  , where e.g.

$$(1.13) \quad \delta_1 = K(\theta - \theta_m) , \quad \theta \geq \theta_m ,$$

for some  $K > 0$ , leaving (1.11) unchanged for  $\theta < \theta_m$  .

Another possibility, closer to the microscopic approach we adopt, is to substitute (1.8), (1.9) with

$$(1.14) \quad \dot{N}(\theta, \delta) = \dot{N}_0(\theta) f(\delta),$$

$$(1.15) \quad \dot{R}(\theta, \delta) = \begin{cases} \dot{R}_0(\theta) g(\delta), & \theta \leq \theta_m, \\ \dot{R}_0(\theta), & \theta > \theta_m, \end{cases}$$

where  $\dot{R}_0$  and  $\dot{N}_0$  are positive in  $(\theta_g, \theta_m)$ .  $\dot{N}_0$  vanishes outside this interval, while  $\dot{R}_0(\theta) = 0$  if  $\theta \leq \theta_g$ , and  $\dot{R}_0(\theta)$  has the sign of  $\theta_m - \theta$  for  $\theta > \theta_g$ ;  $f$  and  $g$  are as in (1.8), (1.9).

Clearly, some correction is needed in (1.10) to avoid the possibility of crystals with negative radii, due to the form of  $\dot{R}_0$ .

It is evident that, once a crystal has disappeared, the model should not bear any memory of its history. Hence the expression of the crystallinity should be:

$$(1.16) \quad w(x, t) = \int_0^t \dot{N}(\theta, \delta)(x, \tau) \omega_n R^n(x, t, \tau) H\left(\inf_{\tau \leq s \leq t} R(x, s, \tau)\right) d\tau,$$

where

$$(1.17) \quad R(x, t, \tau) = r_0(\theta(x, \tau)) + \int_{\tau}^t \dot{R}(\theta, \delta)(x, s) ds,$$

$$(1.18) \quad H(s) = \begin{cases} 1 & s > 0, \\ 0 & s \leq 0. \end{cases}$$

Here  $x \in \mathbb{R}^n$  is a space variable; the assumptions on the geometry of the problem and on the regularity of the constitutive functions will be precised in the next section.

In order to simplify the mathematical treatment of the crystallization kinetics, we replace (1.16) with

$$(1.19) \quad w(x, t) = \int_0^t \dot{N}(\theta, \delta)(x, \tau) \omega_n [R(x, t, \tau)]_+^n d\tau,$$

where  $a_+ = \max(a, 0)$ .

We remark that, unlike equation (1.16), (1.19) allows crystals whose radius becomes non positive at a time  $t_0$ , to reappear at a time  $t_1 \geq t_0$ , provided their radius becomes positive again for  $t > t_1$ .

Equation (1.9) can be therefore assumed to be a good approximation of (1.16) in cases when this unphysical occurrence does not change much the actual value of  $w$ . This point will be further investigated in the future, both from the theoretical and the experimental point of view.

## 2. Existence and uniqueness

Let  $\Omega$  be a smooth open bounded domain in  $\mathbb{R}^n$ ,  $n=1, 2, 3$ . If  $\theta(x,t)$  denotes the thermal field,  $(x,t) \in \Omega_T \equiv \Omega \times (0, T)$ ,  $T > 0$ ,  $k(x,t)$  and  $c(x,t)$  are the conductivity and the heat capacity, and  $L > 0$  is the constant latent heat of crystallization (per unit volume), and if we consider  $w_m$  to be constant for  $\theta < \theta_m$ , the heat balance is

$$(2.1) \quad c(x,t) \frac{\partial \theta}{\partial t} = \operatorname{div}[k(x,t) \operatorname{grad} \theta] - L w_m \frac{\partial \delta}{\partial t}, \quad \text{in } \Omega_T.$$

From (1.19) we have that  $\delta = \delta(x,t)$  satisfies the integro-differential equation

$$(2.2) \quad -w_m \frac{\partial \delta(x,t)}{\partial t} = \omega_n [r_0^n(\theta) \dot{N}_0(\theta, \delta)](x,t) + \\ + n \omega_n [\dot{R}_0(\theta, \delta)](x,t) \int_0^t \dot{N}(\theta, \delta)(x, \tau) [R(x, t, \tau)]_+^{n-1} d\tau, \quad (x,t) \in \Omega_T$$

where  $\dot{N}$ ,  $\dot{R}$ ,  $R$  are defined respectively in (1.14), (1.15), (1.17).

In (2.2) we define  $R_+^{n-1} = 0$  if  $R \leq 0$  even in the case  $n=1$ .

For the sake of brevity we will write (2.2) as

$$(2.2)' \quad - \frac{\partial \delta}{\partial t}(x,t) = F[\theta, \delta](x,t).$$

The problem is completed by initial and boundary conditions

$$(2.3) \quad \delta(x,0) = 1, \quad x \in \Omega,$$

$$(2.4) \quad \theta(x,0) = \theta_0(x), \quad x \in \Omega,$$

$$(2.5) \quad \theta(x,t) = \theta_1(x,t), \quad x \in \partial\Omega, \quad t \in (0, T).$$

We give the following

**Definition 2.1** A solution to problem (2.1)-(2.5) in  $(0,T)$  is a pair  $(\theta, \delta)$ ,  $\theta \in C^{2,1}(\Omega_T) \cap C^{0,0}(\bar{\Omega}_T)$ ,  $\delta \in C^{0,1}(\bar{D}_T)$ , such that (2.1)-(2.5) are satisfied in the classical sense.

We use the following notation: If  $A \subset \mathbb{R}^n$  and  $A_T \equiv A \times (0,T)$ ,  $C^{l,l/2}(A_T)$  is the set of bounded functions admitting bounded continuous derivatives  $\left(\frac{\partial^{s_1}}{\partial x_1^{s_1}}\right) \dots \left(\frac{\partial^{s_n}}{\partial x_n^{s_n}}\right) \left(\frac{\partial^r}{\partial t^r}\right)$  for  $E \equiv \sum_{i=1}^n s_i + 2r \leq l$ ; if  $l$  is non-integer, such a derivative is also requested to be Hölder continuous with exponent  $l-E$  w.r.t. space variables, and with exponent  $\frac{l-E}{2}$  w.r.t. the time variable (provided such exponents are less than 1).

$C^{l,l/2}(A_T)$  is a Banach space under the natural norm.

$C^l(A)$  is defined similarly; it coincides with the subset of  $C^{l,l/2}(A_T)$  containing only and all the functions independent of  $t$ .

We also denote by  $C^{0,1}(A_T)$  the set of functions continuous in  $A_T$ , with continuous first derivative w.r.t. the time variable.

Finally  $C^{\beta,1}(A_T)$ ,  $\beta \in (0,1)$ , is the space of functions continuous in  $A_T$ , and satisfying a uniform Hölder condition with exponent  $\beta$  w.r.t. the space variables, and a Lipschitz condition w.r.t. the time variable.

Now we list the assumptions we will use in the proof of the existence and uniqueness theorem, noting that they are far from being minimal;

(H<sub>1</sub>) heat coefficients:  $M^{-1} < c, k < M$  in  $\Omega_T$ . For some  $\alpha \in (0,1)$ ,  $c, k \in C^{1+\alpha, \frac{1+\alpha}{2}}(\bar{\Omega}_T)$ , with norms bounded by  $M > 1$ ;

(H<sub>2</sub>) initial and boundary temperature:  $\theta_0 \in C^{2+\alpha}(\bar{\Omega})$ ,  $\theta_1$  is the restriction to  $\partial\Omega \times (0,T)$  of a function  $\tilde{\theta}_1 \in C^{2+\alpha, 1+\frac{\alpha}{2}}(\bar{\Omega}_T)$ . Their norms are bounded by  $M$ . Moreover

$$(2.6) \quad \theta_0(x) \geq \theta_m, \quad x \in \bar{\Omega};$$

we assume also that standard compatibility conditions of order  $(2+\alpha)$  are satisfied for (2.1) by  $\theta_0$  and  $\theta_1$  on  $\partial\Omega \times \{0\}$  (see [4] Chapter IV).

(H<sub>3</sub>) critical radius, nucleation and growth rates:  $\dot{N}_0, \dot{R}_0, r_0 \in C^1(\mathbb{R})$ ;  $\dot{N}_0, \dot{R}_0$  are given as prescribed in (1.14)-(1.15) and  $r_0 \geq 0$  on  $\mathbb{R}$ . Their norms are bounded by  $M$ ;

(H<sub>4</sub>) functions  $f(\delta)$  and  $g(\delta)$ :  $f, g: \mathbb{R} \rightarrow [0,1]$  are nondecreasing Lipschitz continuous functions,  $f(z) = g(z) = 0, z = 0, f(z) = g(z) = 1, z \geq 1$ ; the Lipschitz norms of  $f$  and  $g$  are bounded by  $M$ .

In the following we will assume that  $(H_1)$ - $(H_4)$  are satisfied.

**Lemma 2.1** Let  $T > 0$ , and let  $\hat{\theta} \in C(\bar{\Omega}_T)$ . Then there exists a unique solution to

$$(2.7) \quad \begin{cases} -\hat{\delta}_t(x,t) = F[\hat{\theta}, \hat{\delta}](x,t), & \text{in } \Omega_T, \\ \hat{\delta}(x,0) = 1, & \text{in } \Omega. \end{cases}$$

Moreover

$$(2.8) \quad 0 \leq \hat{\delta}(x,t) \leq 1, \quad \text{in } \bar{\Omega}_T,$$

$$(2.9) \quad |\hat{\delta}(x_1, t_1) - \hat{\delta}(x_2, t_2)| \leq C(M, T) \{ \|\hat{\theta}(x_1, \cdot) - \hat{\theta}(x_2, \cdot)\|_{t_1} + |t_1 - t_2| \}, \\ \forall 0 \leq t_1 \leq t_2 \leq T, x_1, x_2 \in \Omega$$

(here  $\|f\|_t = \sup_{0 \leq s \leq t} |f(s)|$ ,  $\forall f: [0, T] \rightarrow \mathbb{R}$ ).

**Proof** For any given  $x_0 \in \Omega$  and continuous  $d: [0, T] \rightarrow [-M, M]$ , let  $\tilde{d}(x_0, \cdot)$  be the solution of

$$(2.10) \quad \begin{cases} -\tilde{d}_t(x_0, t) = F[\hat{\theta}, d](x_0, t), & t \in (0, T), \\ \tilde{d}(x_0, 0) = 1. \end{cases}$$

It is obvious that  $\forall (x, t) \in \Omega_T$ ,

$$|F[\hat{\theta}, d](x, t)| \leq C(M, T),$$

implying

$$(2.11) \quad |\tilde{d}(x_0, t) - 1| \leq C(M, T) T, \quad t \in (0, T).$$

Moreover, if we consider two different functions  $d_1, d_2: [0, T] \rightarrow [-M, M]$  we see that

$$(2.12) \quad \|\tilde{d}_1(x_0, \cdot) - \tilde{d}_2(x_0, \cdot)\|_T \leq T \|F[\hat{\theta}, d_1](x_0, \cdot) - F[\hat{\theta}, d_2](x_0, \cdot)\|_T \leq C(M, T) T \|d_1 - d_2\|_T.$$

This means that a  $T_0 \leq T$  can be chosen so that the transformation  $d \rightarrow \tilde{d}(x_0, \cdot)$  maps the ball  $\{z \in C^0([0, T]) : \|z\|_T \leq M\}$  into itself, in a contractive way.

Clearly we can choose  $T_0$  independent of  $x_0$ .

Then we have proved existence of a unique solution to (2.7) in  $(0, T_0)$ .

The first inequality in (2.8) follows from

$$\frac{\partial \hat{\delta}}{\partial t}(x, t) = -F[\hat{\theta}, \hat{\delta}](x, t) \geq -C(M) \hat{\delta} - \chi_{\{\hat{\theta} \leq \theta_m\}}(x, t) C(M, T) \dot{R}(\hat{\theta}, \hat{\delta})(x, t) \geq -C(M, T) \hat{\delta}.$$

The second inequality follows from the immediate consequence of (2.7)

$$\hat{\delta}(x,t) = 1 - \frac{\omega_n}{w_m} \int_0^t \dot{N}(\hat{\theta}, \hat{\delta})(x, \tau) [R(x; \tau)]_+^n d\tau.$$

Owing to the a priori estimate (2.8), we may iterate the argument above to cover the whole interval  $(0, T)$ , since the choice of  $T_0$  is independent of the initial condition.

It remains to prove (2.9). From (2.7) and assumptions  $(H_3)$ ,  $(H_4)$  we get

$$|\hat{\delta}_t(x_1, t) - \hat{\delta}_t(x_2, t)| \leq C(M, T) \{ \|\hat{\theta}(x_1, \cdot) - \hat{\theta}(x_2, \cdot)\|_t + \|\hat{\delta}(x_1, \cdot) - \hat{\delta}(x_2, \cdot)\|_t \}.$$

Thus, from Gronwall's Lemma we obtain

$$(2.13) \quad |\hat{\delta}(x_1, t) - \hat{\delta}(x_2, t)| \leq C(M, T) \|\hat{\theta}(x_1, \cdot) - \hat{\theta}(x_2, \cdot)\|_t,$$

and hence (2.9). □

Of course, if  $\hat{\theta}$  is chosen in particular to satisfy (2.4)-(2.5) and to belong to  $C^{\beta, \beta/2}(\bar{\Omega}_T)$  for some  $\beta \in (0, 1)$ , then Lemma 2.1 implies that  $\hat{\delta} \in C^{\beta, 1}(\bar{D}_T)$ .

Next we prove

**Lemma 2.2** *The problem*

$$(2.14) \quad c(x, t) \frac{\partial \tilde{\theta}}{\partial t} - \operatorname{div}(k(x, t) \operatorname{grad} \tilde{\theta}) = L w_m F[\hat{\theta}, \hat{\delta}](x, t), \text{ in } \Omega_T,$$

with conditions (2.4)-(2.5), where  $\hat{\theta} \in C^{\beta, \beta/2}(\bar{\Omega}_T)$ ,  $\beta \in (0, \alpha]$ , satisfies (2.4)-(2.5), has a unique solution  $\tilde{\theta} \in C^{2+\beta, 1+\beta/2}(\Omega_T)$  such that

$$(2.15) \quad \|\tilde{\theta}\|_{C^{\bar{\alpha}, \bar{\alpha}/2}(\bar{\Omega}_T)} \leq C(M, T),$$

where  $\bar{\alpha}$  depends on  $\alpha$  and  $M$  of assumptions  $(H_1)$  and  $(H_2)$ .

**Proof** Because of assumptions  $(H_3)$ ,  $(H_4)$ , the norm  $\|F[\hat{\theta}, \hat{\delta}]\|_{C^{\beta, \beta/2}(\bar{\Omega}_T)}$  is bounded by a constant  $C(M, T)$ .

At this point Schauder type estimates (as obtained in [4] Theorem 1.1 p. 419) imply estimate (2.15) for the solution  $\tilde{\theta}$ , whose existence and uniqueness in the required class follow from standard parabolic theory (see [4] chapter IV). □

According to Lemma 2.2, if we choose  $\hat{\theta} \in C^{\bar{\alpha}, \bar{\alpha}/2}(\bar{\Omega}_T)$ , with  $\bar{\alpha}$  as in (2.15), we have defined a mapping  $\hat{\theta} \rightarrow \tilde{\theta}$  of a closed ball of this space into itself, because the constant in (2.15) does not depend

on the choice of  $\hat{\theta}$ , provided  $\theta$  is Hölder continuous.

Moreover, if we choose  $\hat{\theta}_1$  and  $\hat{\theta}_2$  in such a ball and we calculate the corresponding  $\hat{\delta}_1$  and  $\hat{\delta}_2$ , we can obtain  $\tilde{\theta}_1$  and  $\tilde{\theta}_2$  solving problems of type (2.14). This means that the difference  $\tilde{\theta}_1 - \tilde{\theta}_2$  solves a conduction problem with zero data and with source bounded in terms of  $\|\hat{\theta}_1 - \hat{\theta}_2\|_t$  and of  $\|\hat{\delta}_1 - \hat{\delta}_2\|_t$ . The latter is in turn estimated by means of (2.13).

Finally

$$(2.16) \quad \|\tilde{\theta}_1 - \tilde{\theta}_2\|_t \leq C(M, T) \|\hat{\theta}_1 - \hat{\theta}_2\|_t, \quad t \in (0, T).$$

Thus the continuity of the transformation and Schauder fixed point theorem prove the existence of a solution.

Clearly, uniqueness follows from (2.16).

So we proved

**Theorem 2.3** *Under the assumptions (H<sub>1</sub>)-(H<sub>4</sub>), the problem (2.1)-(2.5) has one unique solution in  $\Omega_T$ .*

**Remark 2.1** Note that the assumption  $f, g$  Lipschitz continuous in  $[0, 1]$  may be weakened allowing a non-Lipschitz behaviour at  $\delta=0$  (e.g.  $g(\delta)=\delta^q$ ,  $0 < q < 1$ ). This can be done if in proving the theorem we can restrict ourselves to intervals  $(0, T)$  such that  $\delta$  is uniformly positive for  $t \in (0, T)$  (we recall that  $\delta(x, 0)=1$ ).

Of course, in this case,  $\delta$  may attain the value  $\delta=0$  in a finite time.

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