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Fingering in slow combustion

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Abstract

We report numerical simulations of the structure of advancing burning fronts under conditions where the dominating transport mechanisms is the diffusion of oxygen and heat. The model we study describes the interplay between two diffusive fields: one which accounts for the destabilizing mechanism leading to the production of a large surface area and the other for a stabilizing mechanism. The typical length scale associated with the observed fingering instability turns out to be a combination of the diffusion lengths associated with the two competing processes.

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1. Introduction

One of the most interesting aspects of non-equilibrium processes is their ability to generate complex behavior and produce an endless variety of shapes, due to the presence of dynamical instabilities determined by the coupling between thermodynamic and transport properties. Recently, there has been a surge of interest on the pattern formation occurring in a variety of growth phenomena such as solidification processes, viscous fingering, electrochemical deposition, formation of river networks, diffusion limited aggregation, dielectric breakdown and flame propagation [1,2]. In many of these processes the convection is negligible and the growth is controlled by diffusion. The classical example is represented by the diffusion of latent heat released when a liquid crystallizes. The velocity of the advancing solid–liquid interface depends on the rate at which the latent heat is diffused away. For many of these phenomena thermodynamics

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and hydrodynamics have provided a satisfactory framework and one can now relate the typical properties such as the velocity of the front, the length of the patterns to well studied quantities such as surface tension, chemical potential differences, temperature gradients [3,4].

The majority of the existing theoretical models employed in the study of flame propagation represent a coarse grained picture, are deterministic and disregard the nucleation of the growing phase. In the present paper, we will discuss a lattice model to simulate the dynamics of a propagating flame front in the absence of convection; the study has been motivated by a recent experiment performed by Zik et al. [5,6] with a two-dimensional apparatus. They used a thin rectangular chamber containing a sheet of paper and a hot wire at one end. Due to the limited amount of oxygen present in the space between the paper and the top of the cell the onset of convection is prevented and the rate of the burning process is governed by the oxygen diffusion towards the flame front. Under these conditions a fingering pattern can be observed. This situation is different from the more common case where convection is at work and flames propagate as a straight front. However, it stimulates interesting questions about the connection with the fingering instability of the combustion process and those related to dendritic growth and similar phenomena. In the combustion problem we cannot invoke the tools employed in the study of ordinary non-equilibrium phase transitions. The classical work of Mullins and Sekerka [7] demonstrated that a solid front advancing into the undercooled melt of its liquid undergoes a dynamic instability due to the competition between two different forces: the destabilizing action of the heat diffusion, which favors the formation of large surface areas, and the stabilizing tendency of the interfacial tension to minimize the surface area. What plays the role of the surface tension in a flame propagation problem? We shall show that the minimal mechanisms leading to the appearance of such a behavior can be accounted for by a simple lattice model in which two diffusive fields and a reactive field, proposed on a purely phenomenological basis, interact and give rise to a rich variety of patterns which can explain the observed experimental behavior.

The present paper is organized as follows: in Section 2 we introduce and motivate the model, in Section 3 we present the results of the simulations of the lattice model for various choices of the control parameters, in Section 4 we consider the continuum model version of the same problem and comment the similarities and the differences between the two approaches, finally in Section 5 we draw the conclusions.

2. Definition of a minimal model of flame propagation

Although, it is well known that a complete description of a combustion process may involve up to few dozens of chemical reactions [8], we shall confine ourselves to discuss a minimal model which contains only a limited number of dynamical variables, but has the ability of predicting the occurrence of the interfacial instabilities, that one observes in some experiments involving burning fronts. To illustrate our model of slow combustion we start by introducing the relevant fields, i.e., constitutive elements, and their evolution rules. Our point is that the salient features of the burning process can

be described only by three basic fields: the first representing the presence of oxygen, the second that of combustible and the third the heat released by the reaction.

Let us briefly recall the phenomenon which has inspired our work. The experiment of Ref. [5] consists of a plane sheet of paper ignited uniformly from one side. The burning reaction proceeds slowly because the air supply is scarce and oxygen has to be driven by diffusion towards the front. In fact, turbulent motions are prevented by the restricted geometry of the experimental apparatus. This situation is not observed frequently in nature, because in this case the air circulation, caused by the heat production determines a turbulent mixing of the reactants, however it might be relevant in some peculiar cases such as flames propagating in narrow gaps between confining walls. As stated before our minimal description of the reaction requires three fields. Two of these have reactive character, namely the oxygen and the combustible, and due to the ignition process go from an unstable state to an adsorbing state; on the other hand, only the oxygen can be transported by diffusion. The third field is the heat released at a burning site and is purely diffusive.

To account for the spatial structure of the system we assign to each site of a two-dimensional square lattice, labelled by i, three dynamical variables A_i, C_i and H_i to represent, respectively, the oxygen concentration (A_i), the presence of combustible C_i, taking on the values 0 or 1, to represent the fact that it can be unburned or burned, and H_i the heat amount.

The evolution consists of the following steps:

(1) a site i is chosen randomly.
(2) if both the local heat and oxygen variables H_i and A_i are above some fixed thresholds (T_h and T_O) the discrete variable C_i will change its state ($0 \rightarrow 1$) in an irreversible fashion, i.e., the site lights up.
(3) A certain quantity of heat is released by the reaction and some oxygen is consumed as represented by the following scheme:

$$C_i' = C_i + 1\,,$$

$$H_i' = H_i + \Delta H\,,$$

$$A_i' = A_i - \Delta A\,.$$

(4) The oxygen molecules diffuse towards the reaction zone, while the heat diffuse away from there. The two processes occur at different rates.

This model is inspired to earlier work, where we gave a microscopic description of a system undergoing a first-order phase transition with emission of latent heat [9]. However, three main differences occur with respect to the solidification problem:

(a) in solidification the analogue of the material field may undergo either the liquid to solid transformation or the reverse.
(b) The oxygen field can only be consumed but never produced.
(c) There is no surface energy cost between neighboring sites belonging to different states.

The absence of detailed balance, implied by (a) and (b) is a consequence of the fact that an adsorbing burned state always propagates irreversibly into the unstable phase.

To summarize the present model is a combination of two reactive–diffusive fields (A and H) with a purely reactive field (C). The burning sites represent sinks for A and sources for H. Let us analyze in some detail what are the stabilizing and destabilizing mechanisms.

- The ignition at a site i, cannot start unless a sufficient amount of thermal energy (heat) is present. This reflects the sensitivity of the typical chemical reactions to the temperature. In the spirit of providing a minimal description of the burning process we deliberately chose a simple threshold mechanism, instead of the usual Arrhenius thermally activated mechanism.
- Heat is released by combustion and is diffused. If the associated thermal diffusion length is sufficiently short the result will be that of a stabilizing force similar to a surface tension, because only sites close to a site which is releasing heat can light up.
- If the oxygen supply is not present at a given spot, it must be transported there by diffusion. Thus the need for fresh oxygen tends to favor the formation of a large surface area, i.e., a burning site will be surrounded preferentially by unburned sites.

The balance between these two competing forces leads to a dynamical instability of the propagation front between the burned and unburned material, whose characteristics will change according to the experimental conditions.

3. Results

We performed simulation runs of the reactive–diffusive model described above in the xy plane, using a 512×512 lattice. In the initial configuration the sites are unburned ($C = 0$) and cold ($H = 0$), and the oxygen concentration is uniform ($A = A_0$). To start the combustion a thin strip ($x < x_0$) at the left border of the domain is prepared in the "hot" state ($H = 1$). In our simulations we fixed the oxygen concentration threshold at $T_O = 1$; the release of heat and the oxygen consumption are assumed to be $\Delta H = \Delta A = 1$. The combustion process was followed up to N Monte Carlo steps (MCS), until a well defined regime was attained. The diffusion mechanism for the heat and oxygen fields was simulated via a random exchange between a site and its nearest neighbors; the two different diffusive time scales were controlled by iterating independently, for each MCS, the related attempt frequencies. The resulting frequencies of attempt (per MCS) will be indicated in as f_h and f_o, respectively. To present the numerical results all lengths will be measured in lattice spacing units and the non-dimensional time will be expressed in MCS.

The structures developed in the combustion process strongly depend on the diffusion rate of the oxygen field. In Fig. 1a, b we show, at the instant $t = 8000$, the patterns which arise for two different values of the frequency f_o, having fixed $f_h = 0.2$; the oxygen concentration is $A_0 = 0.75$ and the heat threshold is $T_h = 0.30$. Fig. 1a ($f_o = 1$) shows the typical cellular structure which characterizes the diffusion limited growth of

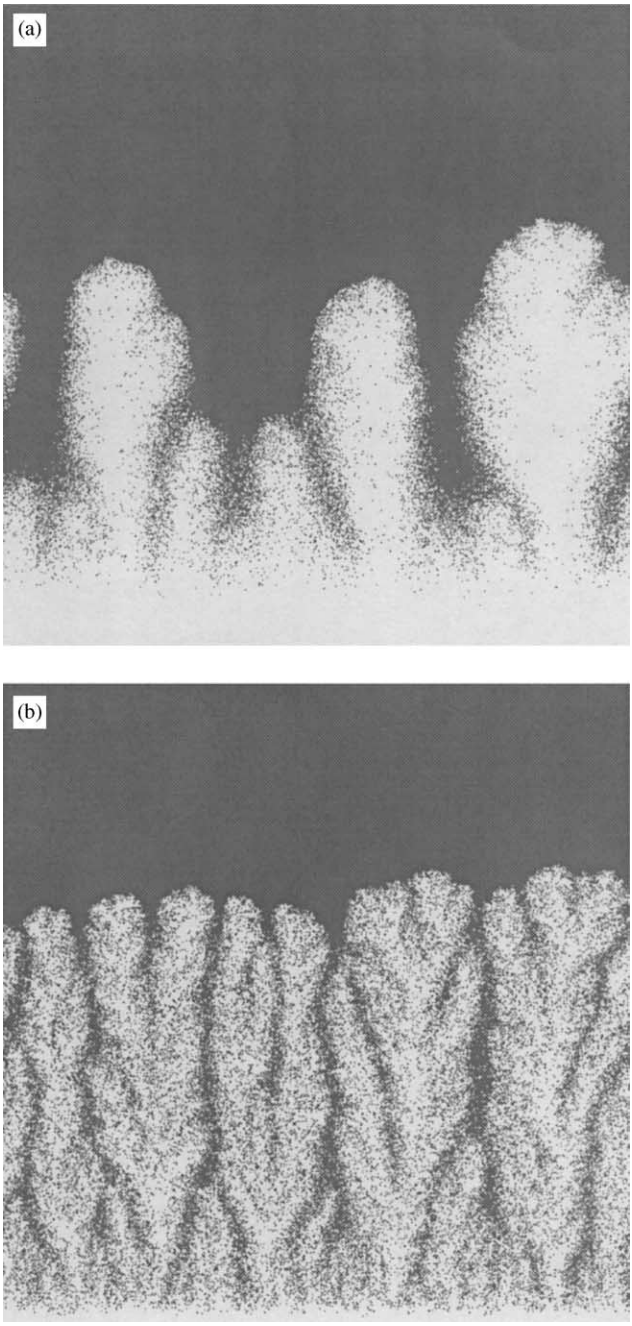

Fig. 1. The combustion field at $t = 8000$ MCS. In both images the initial oxygen concentration is $A_0 = 0.75$, and the frequency of attempt for the heat fields is $f_h = 0.2$. The frequency of attempt for the oxygen field is $f_o = 1$ (upper graph) and $f_o = 0.1$ (lower graph).

a stable phase into a metastable one. We can observe, for example, the competitive dynamics between adjacent cells, and the tip splitting mechanism characteristic of these regimes. Similar structures are observed in the directional solidification of binary alloys beyond the onset of the Mullins–Sekerka instability. The characteristic length of the pattern is selected through the competition of the stabilizing effect of the surface tension and the necessity to develop a large interface area to reject (or to draw) a conserved quantity. Both these phenomena can be described through natural length scales (the capillary length d_0 and the diffusion length l_d, respectively), and the wavelength of the pattern emerges as $\lambda \sim \sqrt{d_0 l_d}$. Our model does not account for an interface energy cost, nevertheless a stabilizing effect is still present, as the ignition is prevented when the local heat field is below the threshold T_h. However, the heat released at a burning site is dissipated through a diffusion mechanism, in such a way that too fragmented structures are disfavored when either T_h is high, or the thermal diffusion length is low. Hence, we expect that, as the diffusion length of the oxygen field decreases, (that means to reduce the frequency f_o) the combustion pattern should develop thinner structures, and eventually assume a dendritic morphology. This is the situation depicted in Fig. 1b ($f_o = 0.1$). We observe that the wavelength of the pattern has been strongly decreased, and the side-branching activity is indicative of the local nature of the diffusion field.

Notice that the concentration of oxygen is initially fixed at a value below the threshold T_O, and to sustain the combustion some oxygen must be drawn towards the interface. Then, a compact front would be slowed down with the growth rate decaying as $t^{-1/2}$. Instead, a cellular or dendritic structure can develop at constant growth rate if the burnt sites left behind the advancing front cover an area fraction equal to the initial oxygen concentration. This is precisely the situation shown in Fig. 2, where the "mass" of the combusted sites is represented versus time. The straight lines refer (from top to down) to the dendritic and cellular growth addressed in Fig. 1a, b: we observe that the combustion front advances at constant growth rate. For comparison we also show a curve representative of compact growth conditions ($f_o = 1$, $T_h = 0.45$, $A_0 = 0.60$). In the latter case we observe a typical diffusion-limited behavior, with the combustion front advancing as $\sim t^{\alpha}$: the deviation of the best fit value $\alpha = 0.53$ from the pure diffusional value $\alpha = 0.5$ can be attributed to a residual porosity of the combusted phase.

We observed that the morphology of the combusted phase is strongly affected by the length scale of the oxygen field. The latter in turn depends on the frequency of attempts f_o, which is the microscopic counterpart of the macroscopic diffusion coefficient D. To elucidate this point we show in Fig. 3 the oxygen concentration along the growth direction, at $t = 8000$. The two curves refer to the same sets of data employed in Fig. 1a, b, and the oxygen field is averaged over the y direction. We note that the width of the transition zone from the low concentration area (behind the combustion front) to the high concentration sites is much larger for cellular growth ($f_o = 0.8$) than for dendritic growth ($f_o = 0.1$).

Fig. 4 shows the diffusion length L_o of the oxygen field versus f_o. The two curves refer (from top to bottom) to $T_h = 0.4$ and 0.3. L_o has been estimated as the length required for the transition from 20% to 90% of the concentration at infinity. Notice that a larger value of T_h reflects on a lower velocity of the process, and we recover the well known result that the diffusion length diminishes as the growth rate increases.

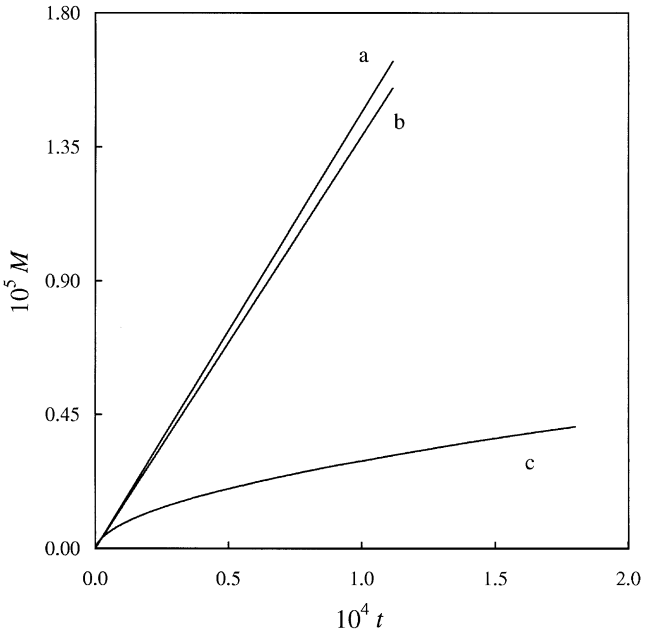

Fig. 2. The mass of the combusted sites represented versus time. The straight lines refer to the dendritic (a) and cellular (b) growth addressed in Fig. 1. The lower curve (c) is representative of compact (diffusion limited) growth conditions, obtained with $A_0 = 0.60$, $f_o = 1$, $f_h = 0.2$ and $T_h = 0.45$.

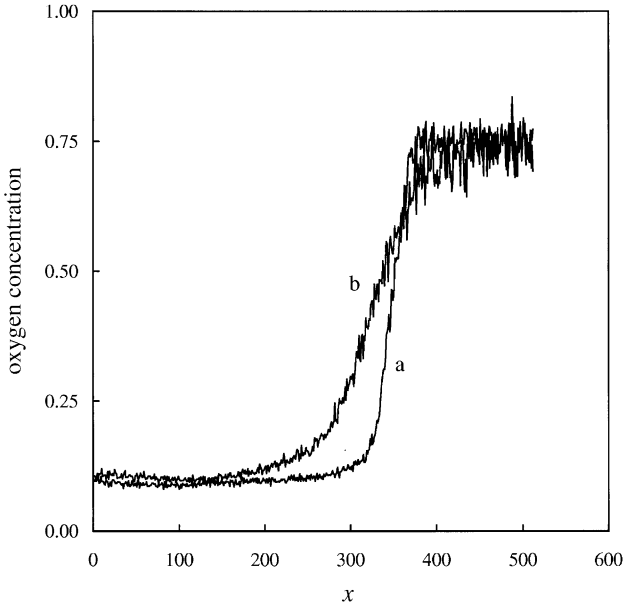

Fig. 3. The profile of the oxygen field for the dendritic (a) and cellular growth (b) addressed in Fig. 1.

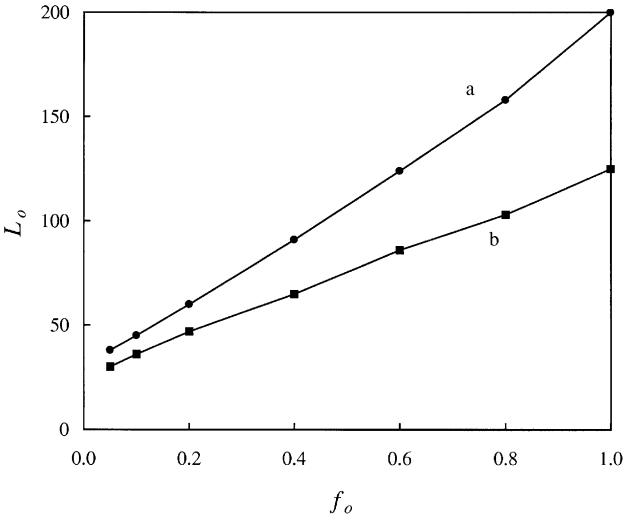

Fig. 4. The diffusion length of the oxygen field versus the frequency of attempt f_o. The two curves were obtained with (a) $T_h = 0.4$ and (b) 0.3.

The above considerations suggest that a morphological phase diagram could be drawn in a T_h, f_o plane, where regions characterized by a large growth rate or low oxygen diffusivity should correspond to dendritic growth, whereas cellular patterns are likely to be found at large values of T_h and f_o. This kind of diagram is shown in Fig. 5, for an initial oxygen concentration $A_0 = 0.75$.

Recently, Grant and coworkers [10,11] studied the nucleation problem in slow combustion by a different method, i.e., modeling the process by a differential equation of the reactive–diffusive type for the temperature field type coupled to a purely reactive equation describing the consumption of the reactants. In their approach the limiting mechanism provided by a newtonian cooling due to a heat reservoir kept a fixed temperature T_0. Probably such a mechanism is not sufficient to produce a front instability of the kind of that observed in experiments of Olami et al. [5]. We also notice that the ignition process of reference contains an Arrhenius factor [11], while in the present model we employed a fixed threshold.

4. Description by means of a continuum model

An interesting question is whether the same behavior observed in a lattice simulation survives also when we model the system by a continuum approach and if it is true which are the differences.

Reaction–diffusion models based on continuous partial differential equations had many applications in recent years in describing the formation of non equilibrium interfacial patterns such as alloy solidification, viscous fingering. A particularly well known model of growth kinetics is the so called Phase field [12], where two fields are

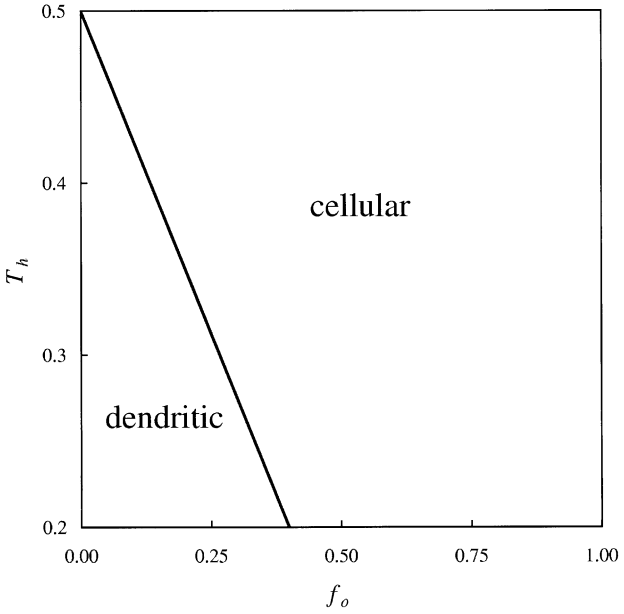

Fig. 5. The morphological phase diagram for the pattern of the combustion field in the T_h, f_o plane.

employed to describe the solidification of an undercooled melt. One of the two fields is purely diffusive, while the other is an ordering field. Their interplay gives raise under suitable condition to complex patterns such as dendrites, cellular phases etc. An analogous description is possible also for the combustion problem as we illustrate hereafter.

The combustible field has a purely reactive behavior, which may be represented in the simplest form as

$$\frac{\partial C}{\partial t} = F[a_1(A - T_O) + a_2(C - C^2)] \,. \tag{1}$$

The dynamic force which drives the system towards the stable burnt state ($C = 1$) increases with increasing local temperature and oxygen concentration; the prefactor F is set to zero (and the reaction is prevented) when either the temperature or the oxygen field are below the threshold level.

The heat and oxygen fields follow a reactive–diffusive dynamics, described by

$$\frac{\partial H}{\partial t} = D_H \nabla^2 H + (\Delta H) \frac{\partial C}{\partial t} \,, \tag{2}$$

$$\frac{\partial A}{\partial t} = D_A \nabla^2 A - (\Delta A) \frac{\partial C}{\partial t} \,. \tag{3}$$

This set of differential equations can reproduce the rich phenomenology of the burning process, i.e., the fingering instability, the cellular regime as well as the transition to

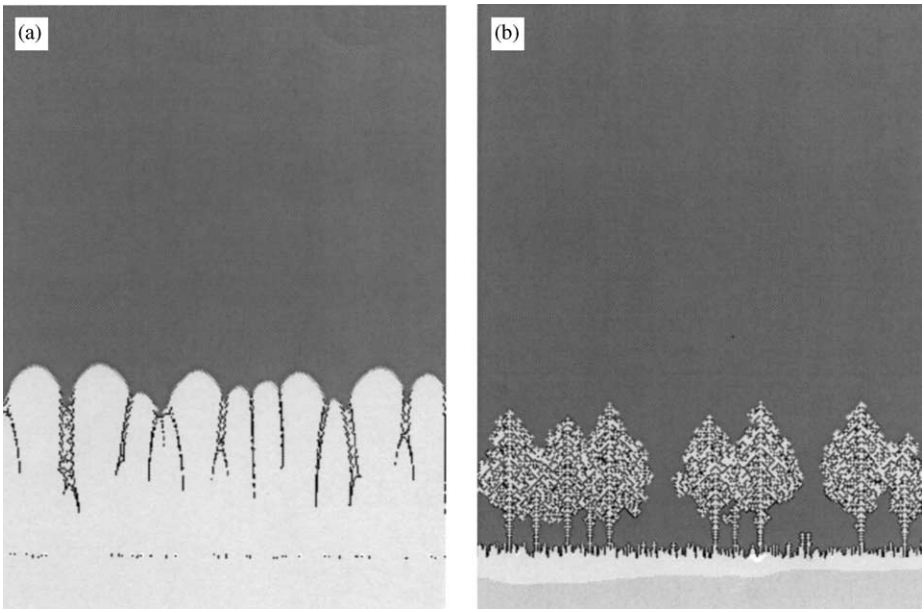

Fig. 6. Cellular and dendritic growth with the continuum model. We chose $D_H=0.01$, $D_A=10$, $\Delta H=\Delta A=1$, $T_O=0.5$, $a_1=2.5$, $a_2=1$. Cellular growth was obtained with an oxygen concentration at infinity $A_0=1$, and $F=0.2$, while for dendrites we have $A_0=0.65$, $F=0.8$.

dendritic growth. As a general trend we notice that the shorter the diffusion length of the A field, the stronger is the side branching activity of the burned structures. This corresponds to the onset of dendritic growth regime. On the other hand, it is clear from the comparison between the discrete and the continuous model that going from the first to the second one looses the granularity typical of the lattice version. Such a granularity is due to the existence of pieces of combustible which did not find the sufficient conditions to burn; we believe that they represent a real effect under conditions of low oxygen concentration.

In order to illustrate the method we show in Fig. 6 the pattern of the combustion field obtained with $D_H=0.01$, $D_A=10$, $\Delta H=\Delta A=1$, $T_O=0.5$, $T_h=0.01$, $a_1=2.5$, $a_2=1$. The two pictures refer to different values of the initial oxygen concentration; $A_0=1$ (cells) and $A_0=0.65$ (dendrites). As usual, dendritic growth requires a strongly localized diffusion field; this is achieved forcing a dynamics of the reactive field ($F=0.8$) faster than that for cellular growth ($F=0.2$).

5. Conclusions

In the present paper, we have discussed a new lattice model which describes the formation and the advancing of combustion front in the absence on convection. With respect to existing approaches to the problem we adopted a microscopic point of view,

in the sense that we have not assumed from the start a coarse grained differential equation. Instead we have assumed a set of stochastic evolution rules for the microscopic variables A_i, H_i, C_i representing, respectively, the local values of the oxygen, the heat and the combustible, (a) a local rule for the C_i variable, (b) two diffusive rules for A_i and C_i plus source and sink terms, respectively. The numerical simulations carried out in two dimensions at various diffusion rates display the trapping phenomenon of unburned combustible, and a complex morphology of the interfacial patterns which has allowed us to construct a phase diagram of the phenomenon. We have also compared our results with a continuous version of the same model and found fairly good agreement as far as the large scales are concerned and differences with respect to the small scales.

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