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Influence of a simple magnetic bar on buoyancy-driven fingering of traveling autocatalytic reaction fronts

M. Mishra,[1] A. Thess,[2] and A. De Wit[3]
[1]*Department of Mathematics, Indian Institute of Technology (IIT) Ropar, 140001 Rupnagar, Punjab, India*
[2]*Institute of Thermodynamics and Fluid Mechanics, Ilmenau University of Technology, P.O. Box 100565, 98684 Ilmenau, Germany*
[3]*Nonlinear Physical Chemistry Unit, Service de Chimie Physique et Biologie Théorique, Faculté des Sciences, Université Libre de Bruxelles (ULB), CP 231, 1050 Brussels, Belgium*

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Magnetic fields have been shown experimentally to modify convective dynamics developing around traveling chemical fronts in presence of unfavorable density gradients. To understand the conditions in which such magnetic fields affect autocatalytic fronts, we study theoretically the influence of a simple magnetic bar on buoyancy-driven density fingering of a chemical front by numerical simulations of a reaction-diffusion-convection system. The model couples Darcy's law for the flow velocity to an evolution equation for the concentration of the autocatalytic product, which affects both the density of the solution and the magnetic force. The solutions of both products and reactants are assumed to be diamagnetic (i.e., negative magnetic susceptibility) and the magnetization is oriented perpendicularly to the plane in which the front travels. We show that, when aligned along the direction of front propagation, the magnetic force is able to suppress or enhance the convective instability depending on the value of the magnetic Rayleigh number of the problem. If the magnetic force is oriented transversely to the front propagation direction, tilted drifting convective patterns are obtained. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4768722]

I. INTRODUCTION

Magnetic fields are known to influence fluid motions in numerous instances but they can also affect reactive systems. The interplay between reactions and magnetic field has indeed already been shown to affect the morphology of protein crystals,[1] the control of cell migration during morphogenesis,[2] or pattern formation in reactive membranes,[3] for instance. In reactive systems, magnetic fields can as well modify the spatiotemporal evolution of autocatalytic traveling fronts.[4–9] Such fronts arise through the coupling between autocatalytic chemical reactions and diffusion, and correspond to self-organized miscible interfaces whereby products invade fresh reactants at a constant speed. The properties of these fronts have been studied in detail in gels used to avoid any convective motions. In absence of gels, density differences across the front can drive buoyancy-driven convective motions, which deform the front into fingers that have been largely studied both theoretically[10–16] and experimentally.[17–21]

If the autocatalytic reactions imply dia- or para-magnetic ions, magnetic fields due to the presence of permanent magnets or electromagnets can induce magnetic forces across the front able to accelerate the traveling speed of the front by several orders of magnitude[4] or lead to a spatial deformation of the otherwise planar interface.[4,5] In parallel, magnetic resonance imaging (MRI) techniques have been used to study the progression of fronts in vertical tubes.[6,7] By applying magnetic field gradients of different geometries using these magnetic resonance (MR) techniques, it has been shown that chemical fronts can be accelerated or decelerated by the use of magnetic forces.

In particular, fingering of autocatalytic fronts can be manipulated by magnetic fields as the magnetic force can oblige the fingers to follow paths imposed by the experimentalist.[8,9]

In this context, our goal is to construct a simple model describing how buoyancy-driven convective instabilities of an autocatalytic reaction front can be influenced and manipulated by a magnetic force due to the presence in the vicinity of the reactor of a simple magnetic bar. We derive a reaction-diffusion-convection model coupling Darcy's law describing the evolution of the flow velocity to an evolution equation for the concentration of the autocatalytic product. Across the traveling front, changes in concentrations induce a jump in density and in magnetic susceptibility responsible for the presence in the flow equation of a buoyancy force and of a magnetic force, respectively. This magnetic force is often referred to as a Kelvin force and its influence on convective systems has been extensively studied.[22]

In the case of the pure hydrodynamic Rayleigh-Taylor (RT) instability occurring when a denser fluid overlies a less dense one in the gravity field, a magnetic field has been used with success to control or even suppress the instability of the interface.[23–25] Magnetic fields also influence the properties of viscous fingering instabilities, for instance.[26–28] In the case of reactive systems, some simulations have been done using commercial computational fluid dynamics packages.[9] In this context, the model developed here shows in which conditions magnetic field effects can simply renormalize the buoyancy effects by yielding an effective Rayleigh number depending on the intensity of the magnetic field. We moreover show how a transverse magnetic field can tilt buoyancy-driven fingers and induce asymmetric drifting of the pattern in time. Our model suggests some simple experiments to be performed to test our predictions.

The article is organized as follows. In Sec. II, we present the experimental set-up we have in mind and the equations describing the problem. In Sec. III, the dynamics of the RT instability of the traveling front in absence of any magnetic field are explained. The influences of an axial or a transverse magnetic field on the dynamics of traveling fronts are discussed in Secs. IV and V, respectively, before concluding in Sec. VI.

II. MODEL

The model system is a Hele-Shaw cell of length L_x and width L_y placed vertically in the gravity field $\underline{g}$ oriented downwards along x and placed in a dc magnetic field (see Fig. 1). This magnetic field is assumed to be produced by a permanent magnet consisting of a long bar of uniformly magnetised material. If the magnetisation is oriented along the z-axis and the bar is infinitely

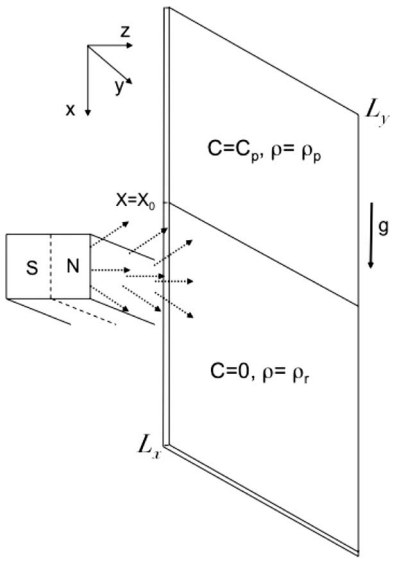

FIG. 1. Sketch of the system.

long, the resulting magnetic field has no component in the y-direction and can be represented as $\vec{B} = B_x(x, z)\vec{i}_x + B_z(x, z)\vec{i}_z$. The two components have to satisfy the condition $\nabla \cdot \vec{B} = 0$ which is expressed by $\partial_x B_x + \partial_z B_z = 0$.[29]

The magnetic force can be written explicitly as

$$\underline{F}_m = \chi(\underline{B} \cdot \nabla)\underline{B}, \tag{1}$$

where χ is the magnetic susceptibility parameter. In general, the two-dimensional magnetic field mentioned above gives rise to a two-dimensional Kelvin force with nonzero components in x- and z-direction. Here we assume that the magnetic force component perpendicular to the Hele-Shaw cell (in the z-direction) is balanced by a pressure gradient and does not affect the flow in the (x, y) plane. Therefore, only the x-component of the force needs to be taken into account, so we assume that in the region of interest, the x-component of the Kelvin force can be written as $\underline{B} \cdot \nabla \approx B_x \partial/\partial x$.

Initially, a solution of the autocatalytic product in concentration $c = c_p$ and of density ρ_p lies at $x = x_o$ above a miscible solution of the reactant, where $c = 0$ and of density ρ_r. The cell has a gap width $h \ll L_x, L_y$ sufficiently thin to consider as a model system that the incompressible velocity field $\underline{u} = (u_x, u_y)$ is two-dimensional and evolves following Darcy's law. Generalization of the problem to the use of Navier-Stokes equations for thicker cells is however straightforward. The flow equation is coupled to a reaction-diffusion-convection equation for the concentration c of the autocatalytic product[12,16] as

$$\underline{\nabla} \cdot \underline{u} = 0, \tag{2}$$

$$\underline{\nabla} p = -\frac{\mu}{\kappa}\underline{u} + F_m(c)\underline{i}_x + \rho(c) g \underline{i}_x, \tag{3}$$

$$\frac{\partial c}{\partial t} + \underline{u} \cdot \underline{\nabla} c = D\nabla^2 c - k\, c(c - c_p)(c + c_k), \tag{4}$$

where c_k is a constant and $\underline{i}_x$ is the unit vector along x. We assume that the solutions are dilute so that the density of the solution depends on c as

$$\rho(c) = \rho_r + (\rho_p - \rho_r)\frac{c}{c_p}. \tag{5}$$

If $\underline{u} = 0$, the reaction-diffusion system (4) has an analytical front solution whereby the product $c = c_p$ invades the reactant $c = 0$.[16]

If the above-mentioned magnetic field is applied close to the Hele-Shaw cell, a magnetic Kelvin force $\underline{F}_m = F_m(c)\underline{i}_x$ depending on c has to be added in Darcy's law (3). We moreover assume that the solutions of both products and reactants are diamagnetic (negative magnetic susceptibility). The difference in the magnetic susceptibility across the front is the important quantity as it leads to a resulting force acting on the solution across the interface. The magnetic susceptibility χ of Eq. (1) is assumed to depend linearly on concentration as

$$\chi(c) = \chi_0(1 + \alpha\frac{c}{c_p}), \tag{6}$$

where $\alpha = \frac{c_p}{\chi_0}\frac{\partial \chi}{\partial c}$ can be positive or negative depending on the magnetic susceptibility of the solution in the reactants and products. As an example, from the experimental work of Evans[9] it is found that the magnetic susceptibility dependence α on concentration of Co(II) is positive, while, in the case of Co(III), it is negative. Finally, we assume that the variation of the magnetic field in the x-direction is so slow (see Fig. 2) that we can approximate it by a Taylor series expansion around an arbitrary location of the unperturbed front x_0. By considering only the first order term in x it can be written as

$$B_x(x) = B_0 + x\frac{\partial B_x}{\partial x}|_{x=x_0}, \tag{7}$$

where $B_0 = B_x|_{x=x_0}$ is the z-intercept of the applied magnetic force (see Fig. 2). Hence, the axial component of the magnetic force in (1) becomes

$$F_{mx} = \chi_0(1 + \alpha\frac{c}{c_p})(B_0 + xG)G, \tag{8}$$

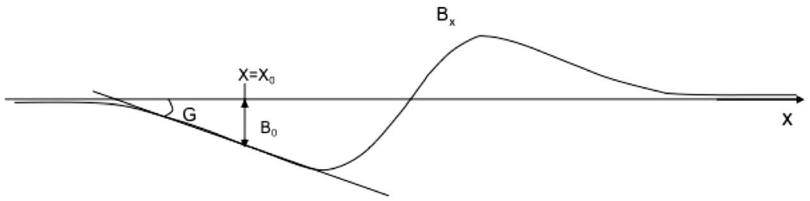

FIG. 2. Applied magnetic field.

where $G = \partial B_x/\partial x|_{x=x_0}$. By neglecting the second order gradient of the magnetic field we get

$$F_{mx} = \chi_0 B_0 G(1 + \alpha \frac{c}{c_p}), \tag{9}$$

where χ_0, α, B_0, and G are given constants known from the experiments.[9] Now using Eqs. (5) and (9), Eq. (3) becomes

$$\underline{\nabla} p = -\frac{\mu}{\kappa}\underline{u} + \chi_0 B_0 G(1 + \alpha \frac{c}{c_p})\underline{i}_x + \rho_r(1 + \Delta\rho \frac{c}{c_p})g\underline{i}_x, \tag{10}$$

where $\Delta\rho = (\rho_p - \rho_r)/\rho_r$. To nondimensionalize the equations, we define a time scale $\tau_c = 1/kc_p^2$, length scale $L_c = \sqrt{D\tau_c}$, and velocity scale $U = D/L_c$. Using the following non-dimensional variables:

$$u^* = u/U, v^* = v/U, x^* = x/L_c, y^* = y/L_c, t^* = t/\tau_c, p^* = (\kappa/D\mu)p, c^* = c/c_p, \rho^* = \rho/\rho_r,$$

and by dropping the (*) from the variables, the governing equations for velocity and concentration become

$$\underline{\nabla} p = -\underline{u} + M_R(1 + \alpha c)\underline{i}_x + \frac{\kappa\rho_r L_c g}{D\mu}(1 + \Delta\rho c)\underline{i}_x, \tag{11}$$

$$\frac{\partial c}{\partial t} + \underline{u} \cdot \underline{\nabla} c = \nabla^2 c - c(c-1)(c+d), \tag{12}$$

where $d = c_k/c_p$ and

$$M_R = \frac{\kappa L_c \chi_0 B_0 G}{\mu D} \tag{13}$$

is the magnetic number. Without loss of generality, we can define a new pressure gradient $\underline{\nabla} p'$, such that

$$\underline{\nabla} p' = \underline{\nabla} p - M_R \underline{i}_x - \frac{\kappa\rho_r L_c g}{D\mu}\underline{i}_x. \tag{14}$$

Introducing the stream function $\psi(x, y)$, such that $u = \partial\psi/\partial y$ and $v = -\partial\psi/\partial x$ and taking the curl of Eq. (11),[31] the final equations read

$$\nabla^2\psi = (M_R\alpha + Ra)\frac{\partial c}{\partial y}, \tag{15}$$

$$\frac{\partial c}{\partial t} + \frac{\partial\psi}{\partial y}\frac{\partial c}{\partial x} - \frac{\partial\psi}{\partial x}\frac{\partial c}{\partial y} = \nabla^2 c - c(c-1)(c+d), \tag{16}$$

where the Rayleigh number Ra is defined as

$$Ra = \frac{\kappa L_c \Delta\rho g}{D\nu}, \quad \nu = \frac{\mu}{\rho_r}. \tag{17}$$

These equations are similar to the evolution equation for the Rayleigh-Taylor buoyancy-driven instability of autocatalytic fronts[12,16,30] with an effective Rayleigh number

$$R = M_R\alpha + Ra. \tag{18}$$

In absence of any magnetic field ($M_R = 0$), the RT instability is observed as soon as $Ra > 0$, i.e., when a denser solution overlies a less dense one. If $M_R \neq 0$, we still effectively have the equivalent of an unstable density stratification leading to a Rayleigh-Taylor instability as soon as $R > 0$, and a stabilization of a buoyantly unstable situation if $R < 0$. This shows that not only can a magnetic field influence or stabilize a genuinely already buoyantly unstable situation obtained when $Ra > 0$, it can also destabilize buoyantly stable situations. For a given reaction (α constant), the intensity of the velocity change due to the magnetic field depends on the intensity of the magnetic field M_R. For a same value of M_R, the response will vary depending on the reaction at hand, i.e., depending on the value of α. Interestingly, Eq. (15) shows moreover that the effect of the magnetic field is possible only if $\partial c/\partial y \neq 0$, i.e., if concentration gradients perpendicular to the direction of propagation of the front are present. This clarifies theoretically experimental observations[4,6] showing that inhomogeneity of both the solution and of the magnetic field are essential to observe any effect and that, as stated by Boga *et al.*[4] "a pattern very similar to the one observed in the gravity effect would be expected" if the front travels along the magnetic field vector. It is also consistent with experimental quantitative observations by Evans *et al.*[6] that the influence of magnetic effects on the velocity of autocatalytic waves depends on the direction of the magnetic field gradient and that no magnetic effects were observed for flat interfaces: a deformation of the front (induced in their case by buoyancy-driven fingering) and a gradient of concentration in another direction than the propagating one was needed for the magnetic field to operate. Let us now show numerically which nonlinear dynamics can be expected in presence of magnetic fields and what type of experiments could be performed. To do so, Eqs. (15) and (16) are solved using a pseudo-spectral method.[16,31] The initial condition is $\psi = 0$ (no flow) everywhere. The concentration $c = 0$ except in two bands where $c = 1$ located on top and bottom of the system to yield ascending and descending fronts whereby the product at $c = 1$ invades the reactant $c = 0$.

III. TRAVELING FRONT IN ABSENCE OF MAGNETIC FIELD

In absence of magnetic field, a Rayleigh-Taylor destabilization of a chemical front in a vertical set-up occurs when the denser side of the front is placed on top of the less dense one in the gravity field. Depending on the type of chemical reaction at hand[32] this occurs for ascending fronts if the fresh reactant solution on top is denser than the product solution beneath it (which corresponds to $Ra < 0$ here). This is typically the case for the well studied iodate-arsenous acid reaction (IAA) for which buoyancy-driven destabilization of ascending fronts has been much studied both experimentally[18,21] and theoretically[12,13,16] and also for which model (22-23) is a good quantitative model. Density fingering occurs on the contrary for descending fronts provided the product solution on top is denser than the reactant solution at the bottom. This is the case of the chlorite-tetrathionate fronts, for instance.[14,19,20] Figure 3 shows the dynamics of both cases in absence of magnetic field ($M_r = 0$) for $Ra = 1$ and $Ra = -1$, respectively. If $Ra < 0$, then $\Delta\rho < 0$ and the reactant density ρ_r is larger than the product density ρ_p so that the ascending front is unstable, while the descending front, which features the reverse stable density stratification, is stable (Fig. 3(b)). The reverse situation shown in Fig. 3(a) is obtained for $Ra > 0$, i.e., descending (ascending) fronts are unstable (stable). Let us now analyze the influence on these dynamics of the presence of a magnet placed close to the Hele-Shaw cell as sketched in Fig. 1.

IV. AXIAL MAGNETIC FIELD

If the magnet is placed horizontally such that the magnetic force is exerted in the axial direction of propagation of the front (see Fig. 1), the magnetic field acts as if it were renormalizing the effective density difference across the front and the new effective Rayleigh number of the problem becomes indeed $R = M_R\alpha + Ra$. As α can be both positive or negative depending on the reaction at hand, it is quite obvious that the magnetic field can then stabilize buoyantly unstable cases while destabilizing stable cases.

This is confirmed on Figs. 4 and 5, which show the influence of increasing magnetic fields for the respectively unstable descending front ($Ra > 0$) with positive and negative magnetic susceptibility.

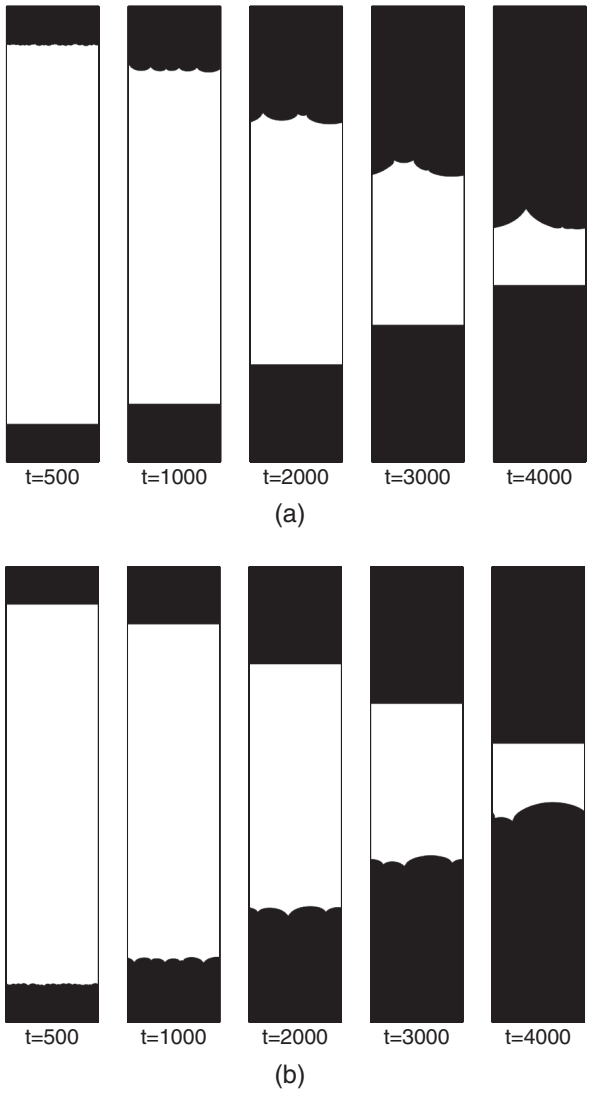

FIG. 3. Nonlinear dynamics of autocatalytic chemical fronts at successive times in absence of any magnetic field ($M_R = 0$) with (a) $Ra = 1$ (unstable descending front) and (b) $Ra = -1$ (unstable ascending front).

It is found for $\alpha > 0$ that, when increasing the magnetic Rayleigh number M_R, the fronts become more unstable as fingering starts earlier (see Fig. 4) and the mixing length L of the fingers is larger (Fig. 6). The dynamics of the fingers for a given effective R is exactly the same as that obtained in absence of a magnetic field for $Ra = R$. As an example, the dynamics in Fig. 4(a) obtained for $Ra = 1, \alpha = 0.25, M_R = 2\,(R = 1.5)$ is exactly the same as that obtained for $Ra = 1.5, M_R = 0$. In the case of a negative susceptibility $\alpha = -0.25$ and $Ra = 1$ then the propagating descending front unstable at $M_R = 2$ is stabilized at $M_R = 4\,(R = 0)$, while $M_R = 6$ gives rise to an unstable propagating ascending front (see Fig. 5).

V. TRANSVERSE MAGNETIC FIELD

Now, if the magnet is applied along the gravity field direction, i.e., perpendicularly to the situation depicted in Fig. 1, it can be represented as $\vec{B} = B_y(y, z)\vec{i}_y + B_z(y, z)\vec{i}_z$. It is straightforward to understand that, under the same assumptions as above, the magnetic force $\underline{F}_m$ will now apply

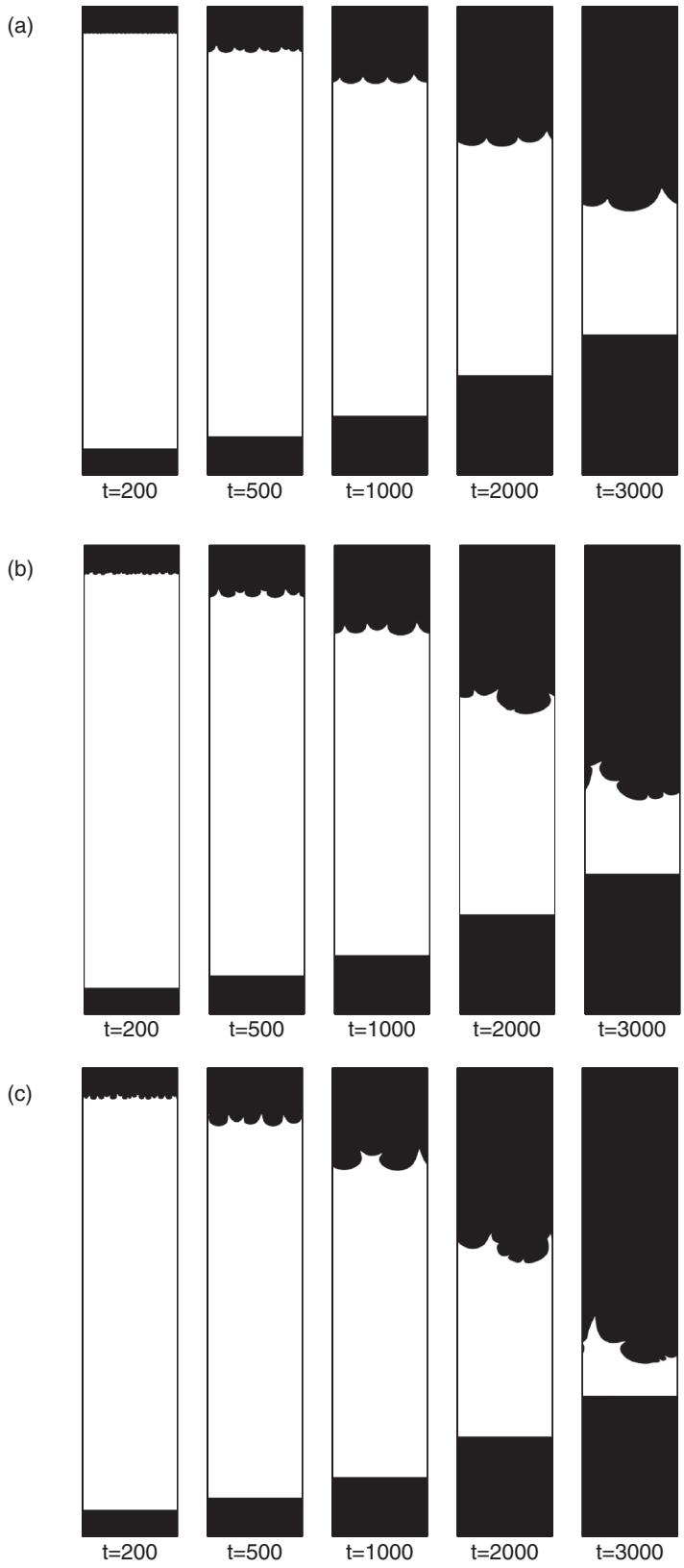

FIG. 4. Nonlinear dynamics of autocatalytic chemical fronts at successive times with $Ra = 1$, $\alpha = 0.25$, and (a) $M_R = 2$, (b) $M_R = 4$, and (c) $M_R = 6$.

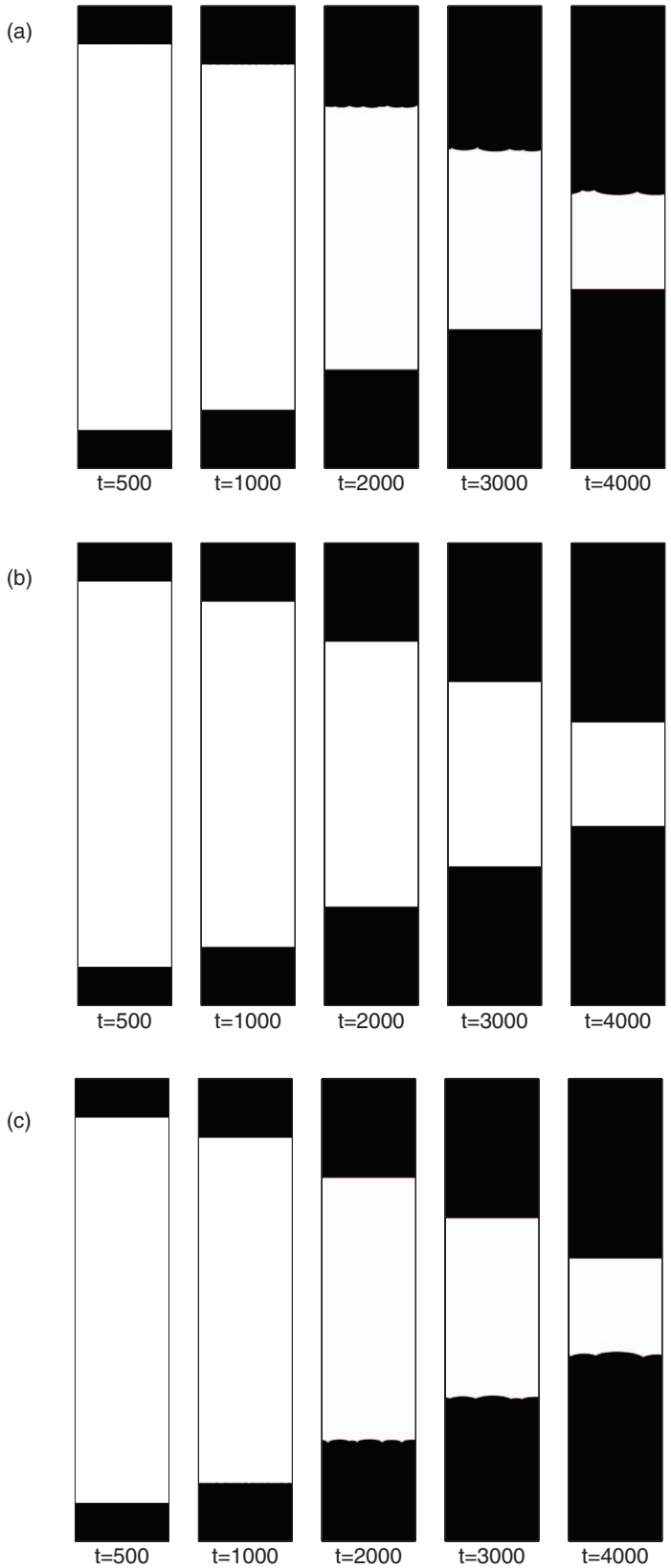

FIG. 5. Nonlinear dynamics of autocatalytic chemical fronts at successive times with $Ra = 1$, $\alpha = -0.25$, and (a) $M_R = 2$, (b) $M_R = 4$, and (c) $M_R = 6$.

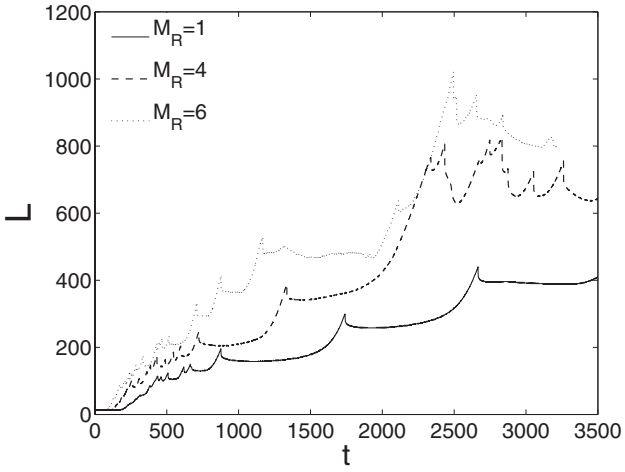

FIG. 6. Mixing length of the finger of autocatalytic chemical front for different magnetic numbers with $Ra = 1$, $\alpha = 0.25$ (corresponds to Fig. 4, unstable descending front).

along the transverse y-direction. The non-dimensional model equations therefore become

$$\nabla^2\psi = -M_R\alpha\frac{\partial c}{\partial x} + Ra\frac{\partial c}{\partial y}, \tag{19}$$

$$\frac{\partial c}{\partial t} + \frac{\partial \psi}{\partial y}\frac{\partial c}{\partial x} - \frac{\partial \psi}{\partial x}\frac{\partial c}{\partial y} = \nabla^2 c - c(c-1)(c+d). \tag{20}$$

The transverse magnetic field provides therefore a source of convection related to concentration gradients perpendicular to those triggering buoyancy-driven flows. Figure 7 compares the convective deformation of the front in absence of the magnetic field and with a transverse magnetic field of same $M_R = 15$ but $\alpha = +0.25$ or -0.25. Interestingly, a transverse magnetic field leads to a tilting of the convective fingers to the right or to the left for α positive or negative, respectively. This is analogous to the tilting of the tip of fingers obtained when applying transverse magnetic field as done by Evans *et al.*[8] in their study of manipulation and control of chemical waves by inhomogeneous magnetic fields. Moreover, in our simulations, we observe that, instead of the general coarsening obtained when $M_R = 0$, more frequent tip splittings lead to an increased number of smaller fingers. This can be appreciated in Fig. 8 showing a space time plot of the dynamics obtained by following the extrema of the transversely averaged profiles $\langle c(y,t)\rangle = \int_0^{L_x} c(x,y,t)dx$, where L_x is the width of the integration domain.[16] This space-time map shows that the location of the extrema is drifting laterally when a transverse magnetic field is applied. The drifting speed increases with the magnitude of the field.

It is interesting to note that, for a positive or negative susceptibility parameter α, we observe that fingers tilt initially towards the left and right sides, respectively, but that, later in time, a reversal of the direction of propagation is observed (Figures 7 and 8). There appears to be a different response to B depending whether the system is in an early destabilizing regime or in a later fully nonlinear one. This is further evidenced in Fig. 9 where we see, by inspecting the temporal growth of the mixing length L in time, that the most unstable system, i.e., the one that destabilizes first is the one with $M_R = 0$. Whether α is positive or negative, the system destabilizes roughly at a same later time when $M_R \neq 0$ (as seen in Fig. 9(b)). However, once fingers start to develop and the nonlinear regime is reached, fingers elongate much more in the presence of a transverse magnetic field. In the long run, their asymptotic mean length is however similar.

Eventually, we end up by insisting on the fact that transverse magnetic fields can only act if the system is genuinely buoyantly unstable, i.e., if $Ra \neq 0$. Indeed, as observed experimentally[4,6] gradients of concentration along y are needed for the transverse magnetic force to operate and such gradients can only be present if a Rayleigh-Taylor instability operates first. Hence, in absence of any

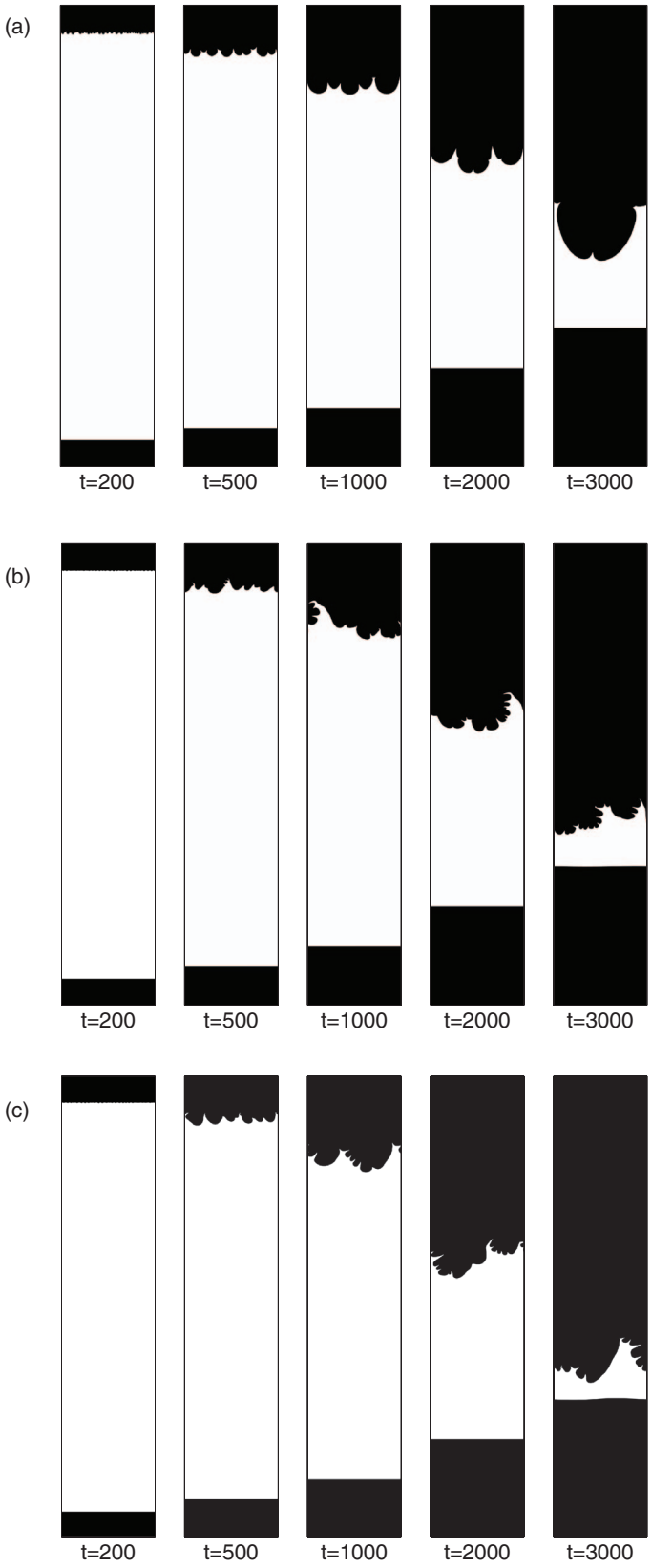

FIG. 7. Nonlinear dynamics of autocatalytic chemical fronts at successive times when $Ra = 2$, and a transverse magnetic field is applied with (a) $M_R = 0$, (b) $M_R = 15, \alpha = 0.25$, and (c) $M_R = 15, \alpha = -0.25$.

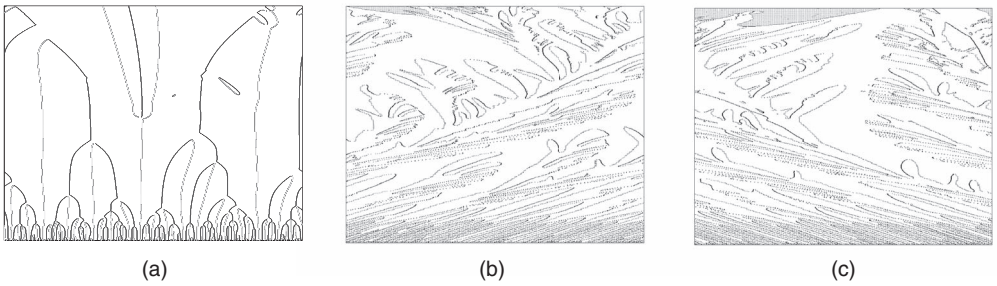

FIG. 8. Space-time maps of the locations of the maxima (black) and the minima (grey) of the transversed averaged profiles $\langle c(y,t)\rangle$ as a function of time for $Ra = 2$ and a transverse magnetic field with (a) $M_R = 0$, (b) $M_R = 15, \alpha = 0.25$, and (c) $M_R = 15, \alpha = -0.25$. The horizontal direction corresponds to the length in the y-direction ($l_y = 2048$), while time is increasing upward from $t = 0$ up to $t = 3000$.

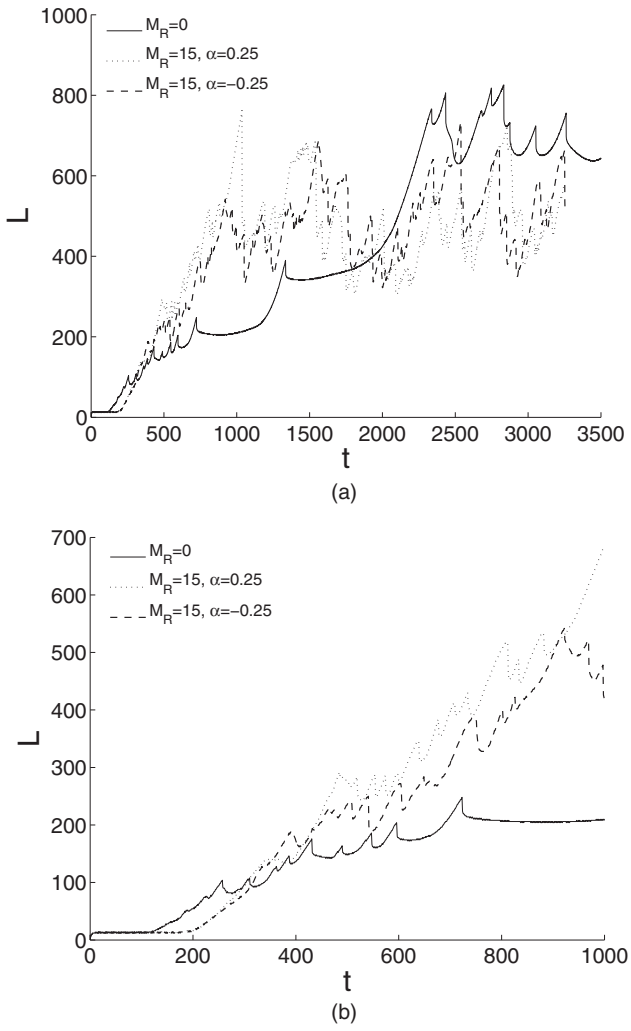

FIG. 9. (a) Mixing length of the finger of autocatalytic chemical front in presence of a transverse magnetic field for $Ra = -2$ (corresponds to Fig. 7), (b) magnification of (a) at early time t.

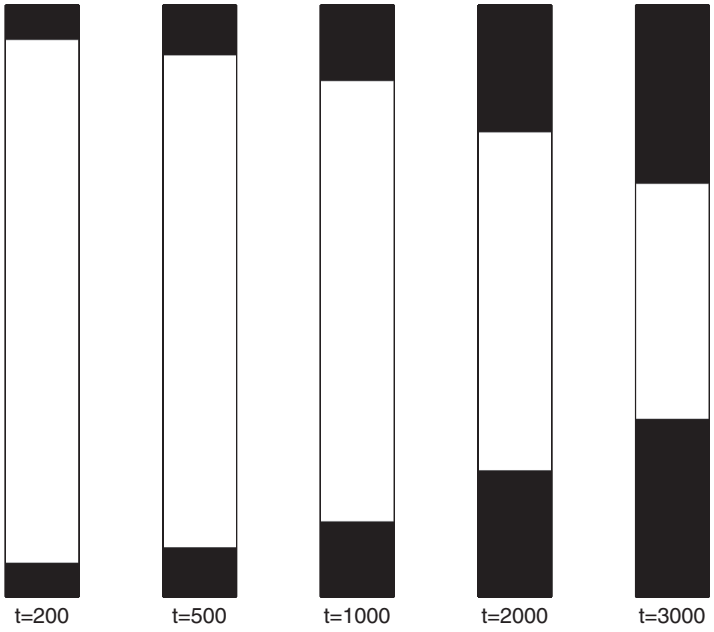

FIG. 10. Nonlinear dynamics of autocatalytic chemical fronts at successive time for $Ra = 0$, and a transverse magnetic field $M_R = 10$, $\alpha = 0.25$.

Rayleigh instability ($Ra = 0$), both up- and downward moving fronts remain planar even in presence of a transverse magnetic field (see Fig. 10).

VI. CONCLUSIONS

Simple magnetic bars are efficient tools to influence and control the dynamics of traveling autocatalytic fronts. We have here developed a theoretical model taking into account the influence of the related magnetic force on buoyancy-driven instabilities of fronts traveling in the gravity field. If the bar is oriented such that the magnetic force acts in the direction of propagation of the front, this force combines to gravity to yield convective flows deforming the front provided gradients of concentration transverse to the direction of propagation exist. A renormalized Rayleigh number $R = M_R\alpha + Ra$ depending upon the intensity M_R of the magnetic field and on the magnetic susceptibility α specific to the reaction at hand can be defined. As a consequence, depending on the values of parameters, the magnetic field is able to stabilize or on the contrary enhance a buoyancy-driven fingering instability depending whether it decreases or increases R. For the special case where $R = 0$, it can even suppress any convective motions. If the bar is oriented such as to exert a transverse force, drifting tilted fingers can be obtained as well. Our simple model recovers previously observed experimental results and paves the way to additional studies devoted to analyze how a magnetic field can control the speed and orientation of propagation of traveling waves. More generally, this simple model will be a convenient tool to be used for further studies on the influence of external forces on convective instabilities in reactive systems. Our results suggest that simple experiments to test our theoretical predictions can be made by using simple magnetic bars displaced along vertical Hele-Shaw cells in which buoyancy-driven instabilities of such fronts have recently been studied very much.[18,19]

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