Numerical study of unstable reaction-diffusion fronts in porous media

D Müller¹, A D’Onofrio¹,² and M Cachile¹,²,³

(1) Grupo de Medios Porosos. Facultad de Ingeniería. Universidad de Buenos Aires. Paseo Colón 850 (1063). Buenos Aires. Argentina, (2) CONICET
E-mail: (3) mcachile@fi.uba.ar

Abstract. Numerical simulations using a lattice-gas method (FHP-III) were performed. The method was modified for simulating two miscible fluids into a rectangular cell with obstacles (porous medium) and without obstacles (Hele-Shaw cell). An external force can be applied to each fluid. A suitable combination of this force and initial conditions can develop an unstable interface front (Rayleigh-Taylor instability). A chemical reaction between the fluids was also introduced, with a parameter that controls the reaction rate. The influence of the parameters that control the force and the chemical reaction on the development of the instability was studied.

1. Introduction

Usually the numerical resolution of a fluid flow is based in discretizing the relevant equations in several ways (finite elements, spectral techniques, etc.). The lattice methods, such as lattice-gas, use a different approach: the key idea is to use pseudo particles moving in a discrete lattice with a small number of possible velocities, constrained by the geometry of the lattice itself. In many real systems, the behaviour at macroscopic scales does not depend on the details of the dynamics at the microscopic scales. In these cases, it appears as a feasible option to approximate the macroscopic phenomena by using simplified rules applied at a much smaller scales.

Although at microscopic (lattice) scales the interparticle dynamics is extremely simplified, the method reproduces the correct behavior of a fluid at larger scales. Under some hypothesis, this method satisfies the incompressible Navier-Stokes equation[1, 2].

The main advantage in using this method is that it is not required the use of the macroscopic equations of the problem, since only elementary microscopic interactions between particles are involved.

At the same time, it is relatively easy to define rather complex boundaries and contours, which makes this method suitable to study flows in porous media [2, 3].

The lattice-gas method can be applied in problems with two fluids, with different mass densities. If the density gradient through the interface is opposite to the gravity, there will appear density-driven instabilities [4, 5]. It has been shown that this situation can be simulated by the lattice-gas method by applying a different force (weight) for each species [6]. This scheme can be applied not only to a difference in density, but to any other problem in which each species is under the influence of a different force.
In literature, the porous media used in this kind of simulations are usually composed of one-node obstacles, known as *scattering nodes* [7]. These obstacles reproduce quite well the effect of the porous media on the flow, but with a different permeability - porosity relationship of real porous media.

In addition, the model was modified to simulate a chemical reaction, where one species is transformed into the other. It was done in a way that reproduces an autocatalytic reaction, previously studied numerically [8] as well as by experimental methods [9, 10].

The main goal of this work is to analyze the influence of the external force and chemical reaction on the development of the unstable front between the two fluids, inside Hele-Shaw cells and porous media.

### 2. Model Set Up

A variation of the basic FHP-III model of seven velocities [2] has been set up, modified in order to simulate two fluids.

Particles of both species are identical, except in a scalar magnitude we called *colour* used to distinguish each species. The colours were chosen to be red and green, from RGB colour model of images.

#### 2.1. Species assignment

During the collisions, the particles are treated in the same way regardless their species, which implies the need of a redistribution after each collision. A random redistribution, uncorrelated with the initial condition was used, which ensures that both species behave as miscible.

#### 2.2. Initial and boundary conditions

On the solid boundaries, including the outer walls as well as the obstacles which form the porous media, we used viscous collisions, *i.e.* when a particle collide with an obstacle, its velocity is reversed. In this way, in the neighbourhood of solid boundaries, as in a real viscous fluids with no-slip condition, the fluid has essentially null velocity.

Two kind of boundaries were used: closed cells, used only in diffusion simulations, where the four walls are solid and periodic cells, where left and right walls (perpendicular to the force) are solid, while top and bottom walls are open.

The simulations were performed with an initial condition that consists in a uniform distribution of particles in the cell. The green particles are placed at the left half of the cell, while the red ones are in the other half of the cell. The concentration of both species is identical. In figure 1 is shown this initial condition.

The particle concentration was controlled by the average filling fraction per node, which is kept constant in each simulation. Concentration $c = 1$ is obtained with 7 particles per node,
values of $c$ ranging from 0.15 up to 0.99 were investigated.

2.3. External Force
The method used by Kadanoff [6] has been implemented. The particle velocities after a collision are reoriented towards the direction of the force. The only possible velocity changes are those parallel to the force ($+x$ in this case) occurring with a probability $u_f$. This value controls the force strength. Values of $u_f$ ranging from $10^{-4}$ to $10^{-2}$ were studied. This force is applied only to the green particles (initially placed at the left) and is pointing to the right, favoring the appearance of density-driven instabilities.

2.4. Obstacles
Two kinds of configurations were investigated: Hele-Shaw cells (without obstacles) and porous media (with obstacles). The obstacles which form the porous media are obtained by placing, at random positions circles of constant diameter $d_c$, with the condition that the minimum distance between any pair of centers is not smaller than $1.5d_c$. We investigated values of $d_c$ between 1 and 10 lattice units. The number of circles has also been varied, ranging from 700 to 3000 circles per $10^6$ nodes.

2.5. Chemical Reaction
With the aim of simulating the behaviour of an autocatalytic reaction between the species, a transformation of one species into the other was implemented through a random change of the color (red to green) of one particle at each node. This is done with a probability of the net reaction rate $p_q$ determined by the product $p_q = Q c_r c_v^2$, where $Q$ is a chemical constant, and $c_r$ and $c_v$ are the number of red and green particles at the node. The values of $Q$ used were between $10^{-5}$ and $10^{-2}$.

3. Results
3.1. Diffusion
The governing equation, with no external forces is [8]:

$$\frac{\partial c_i(x, t)}{\partial t} + \mathbf{u}_i \cdot \nabla c_i(x, t) = D \nabla^2 c_i(x, t) + q_i c_i(x, t)$$

(1)

Where $x, y, t$ are the position and time, $D$ is the diffusion coefficient, $q_i$ is the reaction rate and $c_i$ is the concentration of the species $i$. The solution of equation (1), without chemical reaction, and with a concentration step of $c_0$ placed at $x_0$ as initial condition, is given, for all $x$ by the error function, $erf$ [11]:

$$c(x, t) = \frac{c_0}{2} \left(1 - erf \left(\frac{x-x_0}{2\sqrt{Dt}}\right)\right)$$

(2)

This study of the diffusion has been done in matrices of $1000 \times 2000$ nodes, the average particle concentration, kept constant in each simulation, ranged from 0.15 to 0.99.

The concentration profiles for each species, averaged by column were fitted using equation (2) and then $D$ was found for each concentration.

The same study was done using porous media, in order to analyze its influence on the diffusion coefficient. Porous media formed by circles of constant radius (between 1 and 10 nodes), different obstacle concentration (between 700 and 3000 circles per $10^6$ nodes) and particle concentration between 0.15 an 0.99 were investigated. These results are shown in figure 2.
Figure 2. Diffusion coefficient as a function of particle concentration. Markers represent different porous media.

3.2. Chemical Reaction

The chemical reaction used in the code transforms red particles into green ones. Since in the initial condition green particles are at the left, the front advances to the right. Without forces, this movement is expected to be stable, i.e., the front should remain straight. This behavior has been observed in our simulations.

The position of the front can be determined by plotting the profile along the horizontal axis and then finding the mean-concentration point \( x_f \) where \( c(x_f, t) = c_0/2 \). Plotting \( x_f \) as a function of time, the front velocity is found. After a short transient, the velocity attains a constant value, \( V_f \), shown in figure 3.

4. Instabilities

Using the same initial conditions of the previous study, simulations with the addition of a uniform force, applied to the particles at the left (green) were run.

A short time Fourier analysis of the interface between the two species was developed to study the growth of the instabilities as a function of the wavelength. This study has shown that there exist a range of wavelengths where instabilities can grow, as shown in figure 7.

Porous media were also added, and the radius as well as the concentration of the obstacles were varied.

In figure 8 is shown the behavior of the growth rate (in iterations\(^{-1}\)) when the diameter of the obstacles is changed, while its number is kept constant in 1500 circles /10^6 nodes.
Figure 3. Reaction front velocity $V_f$ as a function of the chemical parameter $Q$ and different particle concentrations.

In figures 4 and 5 examples of this instability are shown for different configurations with and without porous media.

When the instability occurs under a chemical reaction, a change in the shape of the interface is observed, coherent with experimental works [10] and other numerical simulations [8] as shown in figure 6. An influence of the chemical parameter $Q$ on the development of the instability has been observed [8, 13]. As $Q$ increases, the instability becomes smaller, as shown in figure 9.

5. Analysis and discussion

Without forces, nor chemical reaction, the system evolves diffusively, i.e. when the initial condition is a concentration step, each species has a concentration profile that follows equation 2 not only in space, but also has the time variation given by the same equation.

The study on the diffusion has shown that the diffusion coefficient varies as $1/c$, coherently with results found in gaseous systems, in an important range of concentrations as shown in figure 2. This result is not contradictory with the previous one, since local concentration does not apart significantly from the average value.

It has been found that the diffusion coefficient is not modified by the presence of porous media, as shown in figure 2. This fact makes it independent on the presence of obstacles in the integration domain.

The dispersion relation of the instability at short times shows an interval of wavelengths in which instabilities can grow. It can be seen that the wavelengths in the center of that interval...
Figure 4. Unstable evolution. 10,000 iterations, no porous medium, concentration 0.7, force parameter: $8 \times 10^{-4}$.

Figure 5. Identical conditions as in figure 4, but with a porous medium of 1500 obstacles /10$^6$ nodes.

grow faster, as in the case of Rayleigh-Taylor instability [4, 5]. This fact shows that the model reproduces the essential characteristics of that instability in Hele-Shaw cells. Similar behaviour has been obtained when a porous medium is included in the cell.

With the addition of porous media, instabilities are dimmed, and also some shorter wavelengths become stable. Although this study is not completed, the tendency observed on the influence that the porous media have on the instabilities is very close to the observed in experiments in Hele-Shaw cells [13].

In absence of external forces and with the addition of a chemical reaction, a stable front which moves at constant velocity has been obtained. This velocity depends on the chemical parameter
Figure 6. Identical conditions as in figure 4, but with the inclusion of a chemical parameter $Q = 3 \times 10^{-4}$.

Figure 7. Dispersion relation for the Hele-Shaw cell. The full line is to guide the eye. Markers represent different runs.
Figure 8. Growth rate dependence as a function of the diameter of the obstacles. for porous media with 1500 obstacles /10^6 nodes. The force parameter is, from up to down: 8, 4 and 2 x10^{-4}, wavenumber q = 0.03 nodes^{-1}.

as V_f \propto \sqrt{Q}, coherently with experimental results from Horvath [9].

By adding a chemical reaction, the morphology of the instability is clearly modified, as shown in figure 9. This results together with the reduction in the amplitude of the instability because of the presence of a chemical reaction, are coherent with previous experimental works [10, 13].

The results of these simulations show that instabilities grow slower when: a) the diameter of the obstacles (of the porous medium) is kept constant and more obstacles are included, b) the number of obstacles is kept constant and the diameter is increased and c) when chemical parameter Q is increased.

This preliminary study shows that the Lattice gas method with two species is a suitable computational method for the study of the flow in porous media when studying two miscible fluids and under a chemical reaction.

Acknowledgments
This work has been financed by I062 project of the University of Buenos Aires.
**Figure 9.** Growth rate $\sigma$, corresponding to $\lambda = 800$ nodes, as a function of the chemical parameter $Q$. The straight line is the linear regression.
References

[1] Frisch U, Hasslacher B and Pomeau Y 1986 Phys. Rev. Lett. 56 1505
[2] Rothman D and Zaleski S 1997 Lattice Gas automata: simple models of complex hydrodynamics (Cambridge: Cambridge University Press)
[3] Rivet J P and Boon J P 2001 Lattice gas hydrodynamics (Cambridge: Cambridge University Press)
[4] Chandrasekhar S 1961 Hydrodynamic and Hydromagnetic Stability (Oxford: Oxford)
[5] Manickam O and Homsy G M 1995 J. Fluid Mech. 288 75
[6] Kadanoff L McNamara G and Zanetti G 1987 Complex Systems I 791
[7] Kohring G A 1991 J. Stat Phys. 63 411
[8] Yang J D’Onofrio A Kalliadasis S and De Wit A 2002 J. Chem. Phys. 117 20
[9] Toth A Horvath D and Siska A 1997 J. Chem. Soc., Faraday Trans. 93 73
[10] Horvath D Toth A 1998 J. Chem. Phys. 108 1447
[11] Jost W 1960 Diffusion in Solids, Liquids, Gases (New York: Academic Press)
[12] Manickam O HomsyG M 1995 J. Fluid Mech. 288 75
[13] Garcia Casado G Tofaletti L Müller D and D’Onofrio A 2007 J. Chem. Phys 126 114502