**Abstract.** The stability of miscible displacements of Newtonian and shear-thinning fluids of slightly different densities (\(\Delta \rho/\rho \approx 3 \times 10^{-4}\)) with a mean flow velocity \(U\) is investigated in a 2D transparent network of channels (average width = 0.33 mm). Concentration maps providing information at both the global and local scale are obtained through optical absorption measurements and compared in gravitationally stable and unstable vertical flow configurations; the influence of buoyant flows of typical velocity \(U_g\) is characterized by the gravity number \(N_g = U_g/|U|\). For \(N_g < 0.2\), the spreading of the mean relative concentration profile is diffusive for both types of rheologies and characterized by a single dispersivity value \(l_d = D/U\). For the Newtonian water-glycerol solution, \(l_d\) is only the same in the stable and unstable configurations for \(|N_g| < 0.01\). For \(0.01 < N_g < 0.2\), \(l_d\) is increased by buoyancy in the unstable configuration and increasingly large front structures are observed on the concentration maps; for \(N_g > 0.2\), front spreading is not diffusive any more. In the stable configuration, in contrast, the front is flattened by buoyancy for \(N_g < -0.01\) and \(l_d\) reaches values of the order of the length of individual channels. For the shear thinning water-polymer solution, both the concentration maps and the value of \(l_d\) are the same in the stable and unstable configurations over the full range of \(U\) values investigated: this stabilization is explained by their high effective viscosity at low shear rates keeping \(N_g\) below the instability threshold even at the lowest velocities.

1. Introduction

Miscible fluid displacements in porous media are found in many environmental, water supply and industrial processes [1, 2, 3]. The characteristics of these processes, such as the width and the geometry of the mixing front, are often influenced by contrasts between properties of the displacing and displaced fluids such as their density. For unstable density contrast configurations, gravity driven instabilities may appear and broaden the displacement front leading to an early breakthrough of the displacing fluid: this is the case, for instance, when a volume of dense pollutant percolates into a saturated porous medium from its upper surface [4, 5, 6, 7, 8].

The appearance of these instabilities may have harmful consequences, both by enhancing the spreading of pollutants and by reducing the efficiency of industrial processes. The objective of the present work is to demonstrate how such instabilities may be blocked by using shear thinning fluids while, for a same mean given flow velocity, they appear (and may even control the displacement process) for Newtonian fluids.
A key parameter is the characteristic velocity $U_g$ of the buoyant flow components induced by the difference between the hydrostatic pressure gradients in the two fluids which can be estimated from Darcy’s law:

$$U_g = -k \frac{\Delta \rho g}{\mu}, \tag{1}$$

where $\Delta \rho$ is the difference between the densities of the upper and lower fluid, $k$ is the permeability of the medium and $\mu$ the viscosity: with this definition, $U_g$ is positive if oriented upward. When a natural or externally induced mean flow of velocity $U$ is present, the relative magnitude of $U_g$ and $U$ is a key characteristic of the tracer transport process. In the present problem, it is characterized by the gravity number:

$$N_g = \frac{U_g}{U} = -\frac{\Delta \rho g k}{\mu U} \tag{2}$$

In the high velocity limit ($|N_g| << 1$), tracer transport is controlled by hydrodynamical dispersion [1]. If $N_g > 1$ (low velocity and unstable large density contrast configuration), large fingers corresponding to Rayleigh-Darcy instabilities appear in the displacement front. Finally, for $N_g < -1$, the stabilizing density contrast reduces the dispersion.

The present paper is focused on the domain $0 < |N_g| < 0.2$ including the intermediate case of moderate buoyancy effects. Practically, this corresponds to the release of a small amount of pollutant in a porous medium initially saturated by a clean fluid and inducing only small density contrasts (here $\Delta \rho / \rho \approx 3 \times 10^{-4}$); in this case $U_g$ is always small compared to $U$ (so that $|N_g| < 1$). We have studied particularly the influence of buoyancy driven flows on the structure and development of the mixing zone: an important issue is whether the variation of the front width $\Delta x$ remains diffusive ($\Delta x = 2\sqrt{D/t}$ in which the dispersion coefficient $D$ characterizes the mixing process) or whether it is convective ($\Delta x \propto t$).

High resolution optical measurements have been performed in a vertical transparent 2D network of channels of random widths [9] with dye added to one of the fluids. The experiments combine visualizations at the pore scale providing a local description of the mixing process and measurements of the average of the relative concentration of the fluids across the width of the model.

Experiments are performed at different mean displacement velocities $U$ in both stable and unstable density contrast configurations. Comparing displacement processes in the two configurations at a same velocity allows one to detect and characterize the development and influence of buoyancy driven flows. The present experiments will demonstrate that an “ideal” tracer situation (i.e. negligible buoyancy effects with $U_g << U$ [10]) may be reached by adding a polymer to the solutions: this increases indeed very much their effective viscosity, particularly at low velocities $U$.

2. Experimental set-up and procedure

The experimental system and the technique for analyzing the data have already been described in reference [10] and the setup is shown in Fig. 1. The model is a network of $140 \times 140$ channels with a mesh size $d = 0.67$ mm and a depth equal to 0.5 mm. The width $a$ of the channels takes 7 values between 0.1 and 0.6 mm with a log-normal distribution and a mean $\bar{a} = 0.33$ mm. The permeability of the network is $k = 3 \times 10^{-9}$ m$^2$. The model is vertical with its open sides horizontal; its lower end is initially slightly dipped into a bath of the initial fluid of density $\rho_1$ which is sucked uniformly at the top of the model by a syringe pump to saturate the medium. After the saturation, the bath surface is lowered and the initial fluid is replaced by the second one of density $\rho_2$ without perturbing the fluid inside the model. The bath surface is then raised and fluid is pumped again at the top of the model to induce the miscible displacement. This allows one to obtain a front of the second fluid which is initially perfectly flat.
Here, the absolute value $|\Delta \rho|$ of the density difference between the two fluids is kept constant with $|\Delta \rho| = |\rho_1 - \rho_2| = 0.3 \text{ kg.m}^{-3}$ and is due to the small amount of Water Blue dye added to one of the fluid. Since the density varies linearly with the concentration of dye, measuring this concentration optically also provides the value of the local density. For each flow rate and pair of fluids used, both stable ($\Delta \rho > 0$ and $N_g < 0$) and unstable ($\Delta \rho < 0$ and $N_g > 0$) density contrast configurations are studied by swapping fluids 1 and 2. The mean flow velocity ranges from $5 \times 10^{-3}$ to $2.5 \text{ mm.s}^{-1}$.

The fluids used in the experiments are either Newtonian water-glycerol mixtures or shear thinning water-polymer solutions. The Newtonian solution is obtained by mixing 60% in weight of glycerol in pure water. Its viscosity is equal to $\mu = 10^{-2} \text{ Pa.s}$ at 20 °C and the buoyancy induced velocity $|U_g|$ is $10^{-3} \text{ mm.s}^{-1}$. At the lowest imposed flow velocity ($U = 5 \times 10^{-3} \text{ mm.s}^{-1}$ so that $N_g = U_g/U = 0.2$), the time necessary to saturate completely the network is $t \simeq 3 \times 10^4 \text{ s}$. During this time lapse, the characteristic length $U_g t$ characterizing the size of the gravitational instabilities is 28 mm for the water glycerol mixture: this is much larger than the length $d$ of the individual channels of the network and a noticeable influence of gravity on the structure of the displacement fronts is thus expected.

In an effort to reduce the gravitational instabilities, glycerol was replaced in the solutions by 1000 ppm of scleroglucan (high molecular weight polymer from Sanofi Bioindustries). At low shear rates, these solutions reach a constant high viscosity ($\mu \approx 4.5 \text{ Pa.s}$) and the corresponding velocity of buoyancy driven flows (see Eq.2) is $|U_g| = 2 \times 10^{-6} \text{ mm.s}^{-1}$. At the lowest imposed flow velocity used here ($U = 5 \times 10^{-3} \text{ mm.s}^{-1}$), one has then $N_g = 4 \times 10^{-4}$ and the characteristic size of the gravitational instabilities is less than 0.6 mm, i.e. smaller than the pore size $d$; one expects therefore that, for this solution, gravitational instabilities will be damped and dye will behave like an “ideal” tracer.

The model is illuminated from the back by means of a fluorescent light panel and images are acquired by a 12 bits high stability digital camera with a 1030 × 1300 pixels resolution (pixel size = 0.16 mm). Typically, 100 images are recorded for each experimental run at time intervals between 2.5 and 700 s. The images are then translated into maps of the relative concentration distribution of the two fluids in the model using a calibration procedure already described in

**Figure 1.** Experimental setup for miscible displacement measurements in 2D micromodels. Flow is vertical and parallel to the $x$ axis. The width of the channel network is horizontal and parallel to the $y$ axis.
3. Experimental results

3.1. Qualitative observations.

Figure 2 displays relative concentration maps obtained using these different fluid couples in both stable and unstable density contrast configurations. The water-glycerol experiments displayed in Figures (a,c) and (b,d) correspond respectively to unstable ($N_g > 0$) and stable ($N_g < 0$) density contrast configurations.

Fig. 2a and b correspond to a gravity number $|N_g| = 0.04$. Even though this value is significantly lower than 1 and buoyancy might be expected to play a minor role, its effect on the mixing fronts (yellow contours in the figures) is clearly visible. In the unstable configuration (case a), these contours display large fingers increasing the width of the front in the direction parallel to the mean flow. As will be discussed in the next section, the dispersion of the dye tracer is enhanced although it can still be described by the classical convection-dispersion equation.

In the gravitationally stable case (Fig. 2b), gravity flattens the mixing front resulting in a reduced dispersion. Similar pictures obtained during experiments using water-scleroglucan solution are also displayed in Figure 2. Although the velocity $U$ is similar to that corresponding to Figs. 2a and b, no instability or flattening of the front due to buoyancy appears in either the stable and unstable configuration (cases (c) and (d)).

Globally, these results agree qualitatively with macroscopic dispersion measurements performed on 3D packings of glass beads [14] using conductivity tracers detected at the outlet of the samples. In these experiments, buoyancy driven instabilities are also observed at low velocities for Newtonian water-glycerol solutions in a gravitationally unstable configuration but do not appear for water-scleroglucan solutions. With respect to this latter work, the present optical measurements provide additional information on the variation with the control parameters and with time of the width of the mixing zone and of the different geometrical features of the front. We examine first now the variation of the global dispersion characteristics as a function of the experimental parameters and of the configuration of the fluids.
3.2. Quantitative global dispersion measurement procedure

![Figure 3](image-url)  

**Figure 3.** Variation of the normalized mean concentration $C(x, t)$ as a function of the normalized time $(t/\bar{T})$ for $x = 104$ mm and for a displacement experiment using the water-glycerol mixture in a gravitationally unstable configuration ($U = 0.025$ mm$^2.s^{-1}$, $N_g = 0.04$). Continuous line: fit by Equation (3) with $\bar{T} = 4350$ s and $D_{||}/U^2 = 110$ s. Inset: variation of $D_{||}/U^2$ obtained by fitting the mean concentration variation $C(t)$ by Eq. (3) as a function of the distance $x$ from the inlet.

The procedure for determining a global dispersion coefficient from the concentration maps is described in detail in reference [10]. A mean value $C(x, t)$ of the relative local concentration $c(x, y, t)$ of the heavy (dyed) fluid at a distance $x$ from the inlet is first determined by averaging the value of $c(x, y, t)$ for individual pixels over a window of width $\Delta y = 140$ mm perpendicular to the flow and located in the central part of the model (only pixels located inside the pore volume are included in the average).

Figure 3 displays the variations with time of $C(x, t)$ for $x = 104$ mm from the inlet. This variation is well fitted (continuous line) by the following solution of the convection diffusion equation when an initial step-like concentration variation and a constant concentration in the direction perpendicular to the mean flow are assumed:

$$C(x, t) = \frac{1}{2} \left[ 1 - \frac{N_g}{|N_g|} \text{erf} \left( \frac{t - \bar{T}}{\sqrt{\frac{4D_{||}}{U^2}t}} \right) \right]$$

(3)

Adding the factor $N_g/|N_g| = \pm 1$ makes the equation usable in both the gravitationally stable and unstable density contrast configurations (respectively $N_g < 0$ and $N_g > 0$). This fit provides the values of the mean transit time $\bar{T}$ and of the ratio $D_{||}/U^2 = \Delta t^2/(2\bar{T})$ in which $\Delta t^2$ is the centered second moment.

The parameter $\bar{T}$ is always observed to vary linearly with time, implying that the mixing front moves at a constant velocity. Moreover, but for $N_g < 0.2$, linear regressions on the data indicate that the front velocity is (within $\pm 3\%$) equal to the value estimated from the flow rate. The inset of Figure 3 displays the variation of the second parameter $D_{||}/U^2$ as function of the distance $x$. $D_{||}/U^2$ increases slightly at first with $x$ and levels off for $x \sim 50$ mm; for $x > 50$ mm,
it fluctuates around a constant value which is referred to in the following as $D/U^2$. In this "plateau" regime and at all velocities, the standard deviation of the second fitting parameter $D_y/U^2$ is always less than 2%. These results prove that, except for $N_g = 0.2$, front spreading is diffusive with an effective width increasing as the square root of time: this is in agreement with previous studies [17, 6] in which a transition between dispersive and convective spreading was reported for $N_g \simeq 0.30$. One may therefore assume that $D/U^2$ characterizes globally the spreading of the front parallel to the mean flow.

3.3. Dispersion measurement results

![Figure 4. Variation of the dispersivity $D/U$, as a function of the Péclet number $Pe = Ua/D_m$ for two fluids of different rheologies: ($\circ$, $\bullet$) water-glycerol mixture; ($\square$, $\blacksquare$) 1000 ppm polymer solution in gravitationally stable ($\circ$, $\square$, $N_g < 0$) and unstable ($\bullet$, $\blacksquare$, $N_g > 0$) configurations](image)

The present section is devoted to the variation of $D$ (or rather of the normalized dispersivity $l_d/a = D/(Ua)$) as a function of the Péclet number $Pe = Ua/D_m$ in the stable and unstable configurations: these variations are displayed in Figure 4 for the different solutions investigated.

For water-glycerol solutions and at low $Pe$’s, the values of $l_d/a$ in the gravitationally unstable configuration ($\bullet$) are significantly larger than in the stable one ($\circ$). This difference reflects the increase of the global front thickness parallel to the flow due to the development of finger-like structures at low velocities in the first case (Fig. 2a) while the front is flatter in the stable case (Fig. 2b). At the lowest $Pe$ value ($Pe \simeq 25$) for which the dispersivity can be measured in both configurations, there is nearly a factor of 10 between the values of $l_d/a$ for $N_g > 0$ and $N_g < 0$. For $N_g < 0$ and at low $Pe$’s, the normalized dispersivity $l_d/a$ reaches a limiting value (1 mm) of about twice the mesh size of the lattice (horizontal dashed line in Fig. 4).

For displacements in the same range of $Pe$ values but using a 1000 ppm water-polymer solution, the variations of $l_d/a$ with $Pe$ are instead the same for both $N_g > 0$ and $N_g < 0$: this could be expected in view of the very similar maps displayed in Figs. 2c-d for the two configurations. This coincidence confirms that the high viscosity of these shear thinning solutions at low shear rates prevents the development of fingering instabilities in the gravitationally unstable configuration. Quantitatively, this is consistent with the low value $N_g \simeq 5 \times 10^{-4}$ estimated for the lowest experimental velocities by taking the viscosity of the solutions equal to...
the low shear rate limit \( \mu \sim 4.5 \text{ Pa.s} \). Similarly, in the stable configuration, \( l_d/\bar{u} \) is not reduced and the front is not flattened by buoyancy, unlike for the water-glycerol solutions.

At higher velocities \((U > 0.1 \text{ mm.s}^{-1} \text{ or } Pe > 630)\), \( l_d \) takes similar values for both \( N_g > 0 \) and \(< 0 \) for all fluid pairs and increases significantly with \( Pe \). For the polymer solution, \( l_d/\bar{u} \) varies as \( Pe^{0.35} \) (like at lower Péclét numbers) and, for the water-glycerol solution, it increases as \( Pe^{0.5} \). Both behaviors differ from the slow increase observed in such 3D media as homogeneous monodisperse grain packings. Such variations have been studied experimentally in ref. [10] and numerically in ref. [11]: they have been shown to reflect the combined effects of geometrical dispersion due to the disorder of the velocity field and of Taylor dispersion due to the velocity profiles in individual channels. The latter becomes important at high Péclét numbers due to reduced mixing by transverse molecular diffusion at the junctions [12]: as a result, the correlation lengths of the velocity of the tracer particles along their trajectories (and, therefore, the dispersivity \( l_d \)) become larger.

In all cases, the dispersivity variations reported above combine several types of effects. A first one is the local spreading of the concentration variation front which may be deduced from concentration profiles along a single line of pixels parallel to \( x \). A second one is the variations of the geometry of the front resulting from variations of the mean transit time between neighbouring flow paths. These latter variations are now investigated in the present system down to small length scales by taking advantage of the 2D geometry of the model and of the high resolution of the imaging system.

### 4. Spatial structure of the displacement fronts for unstable flows

In porous media, the front structures associated to heterogeneities have been shown experimentally to be strongly amplified in unstable density contrast configurations [16]. This latter effect is studied here by comparing front geometries observed for different values of \( N_g \).

At a time \( t \), the front is assumed to correspond in the relative concentration map \( c(x, y, t) \) to the set of points (or iso-concentration contour) for which \( c(x, y, t) = 0.5 \); as mentioned above, \( c \) is the local relative concentration on the pixel of coordinates \((x, y)\). Examples of such contours determined at four gravity numbers \( 0.02 \leq N_g \leq 0.2 \) are displayed in Fig. 5 for both solutions used in the present work at low and high velocities. For the water-glycerol solution and for \( N_g = 0.2 \) which corresponds to the lowest flow velocity investigated, front spreading, as mentioned above, is not dispersive. In this latter case, \( U_g \sim U \) so that the displacement is dominantly controlled by buoyancy driven flows: they are localized in a few fingers soaring up in the model while the front displacement is almost zero in other regions (Fig. 5a). At higher mean flow velocities (\( N_g < 0.2 \)), the global spreading of the concentration profile is again dispersive (see Sec. 3.3): the large rugosities of the front disappear as the velocity increases as can be seen Fig. 5b. For the shear-thinning water-scleroglucan solution, no instability is visible, even at the lowest \( U \) value which, in Fig. 5c is of the same order of magnitude as in Fig. 5a: as mentioned above, this results from the high effective viscosity at these velocities which leads to a much lower value of \( N_g \approx 0.00045 \). For this solution, increasing the velocity leads to similar large scale structures of the front as shown in Fig. 5d; its local roughness is however of larger amplitude which reflects an enhancement of local spatial velocity contrasts [10].

### 5. Discussion and conclusion

The present experiments on have demonstrated that buoyancy driven instabilities of the vertical miscible displacement of two fluids of slightly different densities could be prevented by using shear thinning fluids: the stabilization results from the high viscosity of these latter solutions at low shear rates. These results agree qualitatively with those of previous investigations on a 3D bead packing which used only tracer concentration measurements at the outlet of the sample [14].
Figure 5. Isoconcentration contours $c(x, y, t) = 0.5$ for mixing fronts of Newtonian and shear thinning fluids in a gravitationally unstable flow configuration (injected volume of fluid = half the pore volume of the model). a), b): water-glycerol solution. c), d): water-scleroglucan solution. (a) $U = 0.005$ mm.s$^{-1}$ ($N_g = 0.2$), (b) $U = 0.05$ mm.s$^{-1}$ ($N_g = 0.02$), (c) $U = 0.005$ mm.s$^{-1}$ ($N_g = 0.00045$)(d) $U = 0.125$ mm.s$^{-1}$. Flow is upward on the figure and $\vec{g}$ is oriented downward.

In contrast, for Newtonian water-glycerol solutions in a gravitationally unstable density configuration, fingering instabilities are observed even for low velocities and small $\Delta \rho$'s. In a stable situation, the effect of buoyancy is, instead, a clear flattening of the front at low flow velocities.

Quantitatively, the variation with time and with distance of the mean concentration is, almost always, well fitted by solutions of a classical convection-dispersion equation. For the water-glycerol solutions, the corresponding dispersivity $l_d = D/U$ in the unstable configuration becomes larger than in the stable one below a threshold velocity; the difference increases at still lower velocities for which the amplitude of the instabilities is observed to be very large on the concentration maps (at the lowest velocity investigated, front spreading is not even dispersive). The values of the threshold velocities at which the dispersivities in the stable and unstable cases become different suggest that the ratio $N_g = U_g/U = -\Delta \rho g k/\mu U$ of the characteristic velocity $U_g$ of buoyancy driven motions and of the mean flow velocity $U$ is the suitable parameter for predicting the onset of the instabilities.

For $N_g < 0.2$, the present measurements demonstrate that, even when instabilities are present, front spreading is dispersive all along the length of the model parallel to the flow with a front width increasing as the square root of distance. Front spreading results here from the combined effects of the structure of the velocity field (sole mechanism in the passive tracer case) that controls the mixing of the fluids and of buoyancy driven flows; the latter either reduce or enhance dispersion depending on the stable or unstable configuration of the fluids.

At low Péclet numbers or equivalently for $N_g > 0.2$, the front displays large spikes and troughs...
and their dynamics cannot be analyzed from linear stability theories. An approach taking into account the combined influence of longitudinal buoyancy forces parallel to the mean flow and of the exchange of solute between the instability fingers and the surrounding fluid is needed to account for the dependence of the dispersivity on the Péclet number ([15]). For the shear-thinning water-polymer solutions, \( |N_g| \) is always less than \( 10^{-4} \): \( N_g \) is then always lower than the threshold value and the dispersivities measured in the stable and unstable configurations are indeed the same.

Preventing such instabilities may be relevant both to the control of the propagation of pollutants and to the improvement of the efficiency of displacement processes in petroleum or chemical engineering. Compared to achieving the same result by means of a Newtonian high viscosity fluids, shear thinning fluids allow one to avoid both an excessive energy dissipation at high shear rates (their effective viscosity is reduced) and to leave the molecular diffusion coefficient practically unchanged.

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References

[1] J. Bear, Dynamics of Fluids in Porous Media, Elsevier Publishing Co., New York (1972).
[2] F.A.L. Dullien, Porous Media, Fluid Transport and Pore Structure, 2nd edition, Academic Press, New York (1991).
[3] M. Sahimi, Flow and Transport in Porous Media and Fractured Rock, VCH, Weinheim, Chap. 12 (1995).
[4] R.A. Schincariol, F.W. Schwartz and C.A. Mendoza, Water Resour. Res. 33 (1), 31–41 (1997).
[5] M. Beinhorn, P. Dietrich and O. Kolditz, J. Contam. Hydrol. 81, 89–105 (2005).
[6] T. Menand and A.W. Woods, Water Resour. Res. 41, W05014 (2005).
[7] K. Johannsen, S. Oswald, R. Held and W. Kinzelbach, Adv. Water Resour. 29, 1690–1704 (2006).
[8] T. C. Flowers and J. R. Hunt, Water Resour. Res. 43, W01407 (2007).
[9] R. Lenormand, C. Zarcone and A. Sarr, J. Fluid Mech. 135, 337–353 (1983).
[10] V. D’Angelo, H. Auradou, C. Allain, J-P Hulin, Phys. Fluids 19, 033103 (2007).
[11] C. Bruderer and Y. Bernabé, Water Resour. Res. 37, 897–908 (2001).
[12] Y. Park, J.R. de Dreuzy, K. Lee and B. Berkowitz, Water Resour. Res. 37, 2493–2501 (2001).
[13] A. Boschan, H. Auradou, I. Ippolito, R. Chertoff and J.P. Hulin, Water Resour. Res. 43, W03438 (2007).
[14] M.A. Freytes, A. d’Onofrio, M. Rosen, C. Allain, J.P. Hulin, Physica A 290, 286–304 (2001).
[15] V. D’Angelo, H. Auradou, C. Allain, M. Rosen, J-P Hulin, Phys. Fluids 20 034107 (2008).
[16] V. Kretz, P. Berest, J.P. Hulin, D. Salin, Water Resour. Res. 39, 1032-1041 (2003).
[17] H.H. Liu and J.H. Dane, J. Contam. Hydrol. 23, 233–243 (1996).