## Experimental Evidence of Disorder Effects in Hydrodynamic Dispersion

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Thanks to an acoustic technique, we have measured the longitudinal dispersion of miscible fluids in three disordered porous media over a wide range of Péclet number. The hydrodynamic dispersion is always Gaussian whatever the flow rate, the sample, or the length of the sample is. The low-flow-rate regime gives the tortuosity of the medium. At higher flow rate, two media having the same permeability and tortuosity but very different pore-size distributions exhibit linear (glass beads) and nonlinear dispersion (foamlike fireproof brick) in accordance with recent numerical simulations.

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The mixing of a dye or a fluid going along with the flow in a porous medium is of great applied importance in secondary oil recovery, chemical engineering, and dispersion of either chemical or nuclear pollutant. From the fundamental point of view, recent efforts have been paid<sup>1-5</sup> to understand this mixing as occurring through the subtle interplay between the two basic phenomena which account for hydrodynamic dispersion, namely molecular diffusion and convection. If convection has been explored thirty years ago in the pioneer works of Taylor<sup>6</sup> and Saffman,<sup>7</sup> it is only recently that new methodologies<sup>4,5</sup> have been developed in order to show how the disorder of porous media, such as pore-size distribution, affects hydrodynamic dispersion. From the experimental point of view the greater part has been per-formed on unconsolidated sediments<sup>8-10</sup> and some in either natural<sup>11</sup> or sintered<sup>12</sup> consolidated media. But these experiments are carried out through effluent techniques, which totally miss the spatial aspect of dispersion or require similar samples of different lengths. In the experiment that we report here, we use an acoustic technique<sup>10,13</sup> which allows us to determine precisely the fluid concentration all along the cores of large porous media. We examine three different porous media and we cover four decades in flow rates. Two samples have the same tortuosity and permeability but very different pore-size distributions; the third one has a very different tortuosity. Our data support strong evidence of disorder effects on hydrodynamic dispersion.

Dispersion in a disordered, multiconnected, statistically homogeneous porous medium is classically described by a macroscopic convection-diffusion equation  $^{1-7}$ :

$$\partial C/\partial t + U \,\partial C/\partial x = D_{\parallel} \,\partial^2 C/\partial x^2 + D_{\perp} \nabla_{\perp}^2 C, \qquad (1)$$

where C(x,t) is the space- and time-dependent fluid concentration; U, the average Darcy fluid velocity in the x direction; and  $\nabla_{\perp}^2$ , the Laplacian in transverse direction. The coefficients  $D_{\parallel}$  and  $D_{\perp}$  characterize the efficiency of dispersion in longitudinal and transverse directions relative to the flow, and may depend on U. In this paper we focus on longitudinal dispersion. The basic problem is to find out the flow-velocity dependence of  $D_{\parallel}$ .

(i) As U tends towards zero, molecular diffusion dominates and  $D_{\parallel}$  becomes constant:

$$D_{\parallel} = D_m / \alpha, \tag{2}$$

where  $D_m$  is the molecular diffusion coefficient and  $\alpha$  the tortuosity.  $\alpha$  is a reducing factor ( $\alpha > 1$ ) accounting for the tortuous nature of the porous medium; molecules spend a lot of time to diffuse in the tortuous branches of the medium. This tortuosity  $\alpha$  is also involved in conductivity measurements<sup>10</sup> through the formation factor F of the medium ( $\alpha = F\phi$ ;  $\phi$  porosity); F is the ratio of the conductivity of a brine to the conductivity of the insulating porous medium saturated with the same brine.

(ii) At large flow rate U, i.e., at large Péclet number  $(N_{Pe} = Ul/D_m)$ , mechanical mixing dominates; fluid particles are convected along different streamlines, sometimes particles move faster than the average flow U, sometimes slower; mechanical dispersion is due to the different transit times along these flow passages. Basically, the transit time  $\tau$  through a pore of length l is of the order l/U; the corresponding dispersion coefficient  $D_{\parallel}$  is then of the order  $l^2/\tau$  leading to  $D_{\parallel} \sim Ul$ . In randomnetwork models of porous media it was first pointed out by Saffman<sup>7</sup> and De Jong<sup>14</sup> that stagnation effects lead to logarithmic correction to  $D_{\parallel}$ . These stagnation effects have been recently reconsidered as well as boundary layers<sup>3</sup> and recirculation zones.<sup>15</sup> All these effects lead to small correction to  $D_{\parallel} \sim Ul$ .

If Saffman's approach demonstrates the relevance of the statistical method, the investigated disorder in his network of capillary tubes of equal length and radius is only an orientation disorder relative to the flow direction. Recent new methods<sup>4,5</sup> of calculation allow us to investigate another type of disorder: network models of porous media consisting of a lattice of tubes of equal length *l* but with a size distribution of diameters. The width of the size distribution characterizes the disorder of the system; whatever the technique is the diffusion regime is recovered  $[N_{Pe} \ll 1; Eq. (2)]$ . At high flow rate ( $N_{Pe}$   $\gg$ 1), mechanical dispersion dominates and

$$D_{\parallel} = U I_D, \tag{3}$$

where  $l_D$  is a dispersion length; the larger the disorder, the larger  $l_D$  compared to l.

Between these two regimes, the crossover region gives rise, depending on the disorder importance, to a dispersion coefficient increasing faster than linearly with U.

As the solution C(x,t) of Eq. (1) is space and time dependent, we use an acoustic technique to determine C(x,t) in our porous samples of size  $4 \times 4 \times 30$  cm<sup>3</sup>. Concentration measurements are derived from the velocity variation of a sound wave in ten cross sections of the sample (ten x values). The spatial resolution reaches 3 mm and the ten positions are scanned in less than 1 min through automatic procedure; this allows us to measure dispersion over a wide range of flow rates (at least four decades for each sample) from 0.01 cm<sup>3</sup>/h to 5000  $cm^{3}/h$ . With temperature stabilization taken care of (better than 0.2 °C), our calibration curve of velocity variations with the mixture concentration C provides an overall precision better than 0.1%.<sup>10,13</sup> We have investigated three porous media: an unconsolidated pack of 200- $\mu$ m glass beads, a fireproof brick, and a mill sandstone. Table I summarizes our measurements of porosity  $\phi$ , permeability k, and tortuosity  $\alpha$ , deduced from conductivity measurements at 1 kHz. In order to compare these three media we use the characteristic pore size  $l_c$ , which has been derived recently by different authors<sup>16,17</sup> from permeability (k) and conductivity  $(\alpha)$  measurements,

$$l_c^2 = a\alpha k/\phi,\tag{4}$$

where the constant a=226 for a percolationlike broad distribution of pore ratio. In fact, a more precise description of our porous media requires the pore-size distribution, the width of which is characterized by a small (r) and a large (R) pore size. Glass beads and our brick sample are very similar from k and  $\alpha$  measurements, but they look very differently from direct microscopic observation. The beads are quasimonodisperse (R/r=1). In contrast to this sample, the fireproof brick

TABLE I. Measured characteristics of our porous media: porosity  $\phi$ , permeability k (1 d=10<sup>-12</sup> m<sup>2</sup>), and tortuosity  $\alpha$ deduced from conductivity measurements (cond.) and dispersion measurements [diff., Eq. (2)].  $l_c$  is the characteristic size of the pore deduced from k and  $\alpha$  (diff.) through Eq. (4).  $l_D$  is the dispersion length measured in the experiment.

Porous medium	ф (%)	k (darcy)	α		lc	
			Cond.	Diff.	(µm)	$I_D/I_c$
200- $\mu$ m glass beads	40	40	1.4	1.7	200	0.5
Fireproof brick	65.5	75	1.3	1.7	200	11
Mill sandstone	21	0.11	7	9	30	17

looks like a foam where pore bubbles are connected by small throats; pore size ranges over roughly a decade  $(R/r \approx 10)$ . This brick is indeed much more disordered than the pack of beads from the pore-size distribution point of view; it would provide a good test of the theory. The sandstone is much more tortuous but its microscopic structure is close to that of the pack of glass beads with irregular shape grains of different sizes. The fluids we use are pure water and a 10% ethanol-water mixture; their densities and viscosities at 30 °C are respectively 995, 975 kg m<sup>-3</sup> and  $8 \times 10^{-4}$ ,  $8.2 \times 10^{-4}$  SI units. At this temperature the molecular diffusion coefficient of ethanol is  $D_m = (1.2 \pm 0.1) \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ . In the experimental procedure, the sample is first saturated with water by a vacuum impregnation technique. Flows are driven vertically along the larger dimension of the sample at a constant flow rate; with such fluids, displacements are stable<sup>9</sup> (no viscous fingering instability) for the more viscous fluid displacing the less viscous one.

The five curves (out of the ten we record) in Fig. 1 correspond to the time-dependent concentration C(t) for five different pairs of transducers at x = 2, 4.1, 6.2, 8.3, and 10.4 cm from the inlet at a U = 0.20 cm/h flow rate in the brick. The small fluctuations in final concentrations are only due to fluctuations of the local porosity (less than 3%). The change of slope as we go away from the inlet is the signature of the dispersion effect. In order to determine the dispersion coefficient  $D_{\parallel}$ , we can remark that, as we neglect the transverse dispersion in the one-dimension convection diffusion equation (1), C(x,t) is a function of the single variable  $\xi = (x - Ut)/2(D_{\parallel}t)^{1/2}$  and for a step concentration at the entrance;

$$C(\xi) = \frac{1}{2} (1 - \text{erf}\xi),$$
 (5)

where erf is the error function. So for each sample and each flow rate U we plot  $C(\xi)$  vs  $\xi$  for the ten pairs of transducers (ten x values).  $D_{\parallel}$  is adjusted in order to give the best fit with Eq. (5). These fits are indeed very good (within better than 5%) and, as we use ten x val-



FIG. 1. Time dependence of the water-ethanol mixture concentration C(x,t) in five out of ten cross sections of the sample (from left to right, x = 2, 4.1, 6.2, 8.3, 10.4 cm from the inlet onwards) recorded during injection of water-ethanol mixture at constant flow rate U = 0.20 cm/h in a water saturated fireproof brick.

ues, give an accurate determination of  $D_{\parallel}$ . As fits by Eq. (5) work for the three samples and whatever the flow rate is, this means that Eq. (1) holds: that the dispersion is Gaussian for the packed glass beads, the brick, and the sandstone. We have to emphasize that the dispersion is Gaussian whatever the distance from the inlet is, i.e., the length of the sample from 2 to 30 cm; we then do not require any asymptotic extrapolation.<sup>11,12,18</sup> The corresponding dispersion coefficients  $D_{\parallel}$  are given in Fig. 2 in a universal representation  $D_{\parallel}/Ul_c$  versus a Péclet number  $N_{\rm Pe} = U l_c / D_m$  which allows a comparison between the three porous media and with theory.<sup>4</sup> Let us focus on the data of the fireproof brick (full circles) and the pack of glass beads (open circles); these two media having similar tortuosity and permeability (Table I) behave completely differently for hydrodynamic dispersion. This is our central result which is well predicted by recent numerical simulations (Fig. 2 of Ref. 4).

Thanks to the wide range of flow rates we have used, we reach the three interesting regimes:

(i) At low flow rate  $(N_{Pe} \ll 1)$ ,  $D_{\parallel}$  becomes nearly constant (slope -1 in Fig. 2) and the tortuosity values  $[\alpha = D_m/D_{\parallel}; \text{Eq. (2)}]$  are 1.7 for the brick and the pack of beads (dashed line of Fig. 2) and 9 for the sandstone (dashed-dotted line). These values are 30% larger than the ones we deduce from conductivity measurements (Table I). This might be understood from the fact that our conductivity measurements are not performed at a low enough frequency.

(ii) At high flow rate  $(N_{Pe} \gg 1)$ ,  $D_{\parallel}/Ul_c$  becomes nearly constant for the three media but with very different values for the corresponding dispersion length  $l_D$  [Eq. (3)]:  $l_D = 0.5l_c$  for the pack of glass beads in accordance with previous results in sediments,<sup>8,10,12</sup>  $l_D = 11l_c$  for the fireproof brick, and  $l_D = 17l_c$  for the



FIG. 2. Universal log-log plot of the reduced dispersion coefficient  $D_{\parallel}/Ul_C$  vs the Péclet number  $N_{Pe} = Ul_c/D_m$ .  $D_{\parallel}$  is the dispersion coefficient, U the flow rate,  $D_m$  the molecular diffusion coefficient,  $l_c$  the characteristic pore size of the porous media. 200- $\mu$ m glass beads (open circles), fireproof brick (filled circles), sandstone (plusses). The dashed lines of slope -1 correspond to the diffusion regime and lead to the tortuosity  $\alpha$  [Eq. (2)] of the three media.

sandstone. Such large  $l_D$  values, compared to  $l_c$ , have been previously reported, <sup>11,12</sup> but in samples in which anomalous, i.e., non-Gaussian, dispersion occurs.

(iii) In the crossover region the pack of beads and the sandstone change monotonically while the fireproof brick exhibits a bump, a deep hollow corresponding to a dispersion coefficient  $D_{\parallel}$  increasing faster than linearly with U. If we want to state this enhancement with a power law  $D_{\parallel} \sim U^{\beta}$ , we find  $\beta = 1.50 \pm 0.05$ . This is, in our opinion, the first time that such an important disorder effect is observed.

In order to attempt to relate these experimental results with theoretical predictions, we must remind ourselves of the main characteristics of the three samples.

(a) The brick and the beads nearly have the same tortuosity and permeability and consequently the same typical length  $l_c$ , but their pore-size distributions are very different (cf. porous media description). As stated before, our results for the brick and the beads look similar to numerical simulations<sup>4,5</sup> on network models with uniform pore-size distributions between a small (r) and a large (R) values. By chance, the constant tortuosity in this model is close to our value 1.7. Recent calculations of Charlaix et al.<sup>5</sup> provide a complete set of curves with various disorder characterized by the ratio R/r. For  $R/r \simeq 1$  they recover a curve similar to the glass-beads one  $(l_D \approx 0.5 l_c)$ . For  $R/r \approx 10$ ,  $l_D \approx 10 l_c$ , and the deepness of the hollow is roughly in agreement with our brick data; so is it for the  $\beta = 1.50$  slope in the crossover region.

(b) The sandstone and the beads correspond to narrower, especially for the beads, grain-size distributions, at different  $l_c$  length scales; they have very different tortuosities. The absence of deep hollow in the crossover region on both samples is in favor of narrow-poresize-distribution interpretation, but previous predictions<sup>4,5</sup> account for neither tortuosity effect nor such a large  $l_D/l_c$  in sandstone. Without any theoretical support, if for universality's sake we want to put the three samples in a series of curves with various disorder but same tortuosity, we have to change  $D_m$  to  $D_m/\alpha$  and  $l_c$  to  $\alpha l_c$ ; the brick and the beads behave similarly as in Fig. 2 and the sandstone comes in between the brick and the beads. Indeed such a construction would mean that the dispersion length involves  $\alpha l_c$ , rather than  $l_c$ , and the disorder.

In summary, by means of an acoustic technique, we have measured the hydrodynamic dispersion all along the cores of three different samples. The dispersion is Gaussian independent of sample, flow rate, and length of the sample. At low flow rate the dispersion coefficients tend to a constant which gives the tortuosity of each medium. At higher flow rate, the monodisperse beads sample and the foamlike brick, having the same tortuosity and the same permeability, give fundamentally different behavior for hydrodynamic dispersion. These features are compared with recent numerical simulations on network models of porous media having a pore-size distribution disorder. As a last comment, hydrodynamic dispersion appears as a suitable tool to differentiate porous media.

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