Reversible and irreversible tracer dispersion in porous media

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Abstract. We present experimental studies of the heterogeneities of porous media by tracer dispersion performed by using either a classical transmission technique or an echo technique; in the latter, the tracer is first injected into the sample and then pumped back. When large channel- or strata-like heterogeneities are present, measuring the degree of reversibility allows one to estimate the size of the heterogeneities and the permeability variations. In homogeneous media such as sphere packings, dispersion becomes irreversible for penetration depths larger than a few diameters. An electrochemical technique is described and applied to dispersion measurements with a resolution better than one grain size.

1. Introduction

From a practical point of view, porous media are of permanent interest in the fields of petroleum and chemical engineering, hydrogeology, soil and environmental sciences. From a fundamental point of view, they represent an archetype of a spatially disordered system. A key question is the evaluation of the heterogeneity of these media which will determine for instance the efficiency of a chemical reactor or of an oil recovery process from a given reservoir field. We shall describe here the analysis of these heterogeneities from non-local tracer dispersion measurements. Another alternative approach is the study of anomalous diffusion effects expected, for instance in the diffusion of pressure waves through poorly connected media [1].

2. Tracer dispersion in porous media

Tracer dispersion is the spreading of a tracer (dye, ionic salt or radioactive materials) in a fluid flowing through a porous medium. In homogeneous media, the tracer concentration $C(r, t)$ verifies [2] the 'Gaussian' convection diffusion equation

$$\partial C(r, t)/\partial t + (U \cdot \nabla)C(r, t) = D_{\|} \partial^2 C(r, t)/\partial x_{\|}^2 + D_{\perp}\Delta_{\perp} C(r, t) \qquad (1)$$

where U is the mean velocity of the fluid. $D_{\|}$ and $D_{\perp}$ are the longitudinal and transverse

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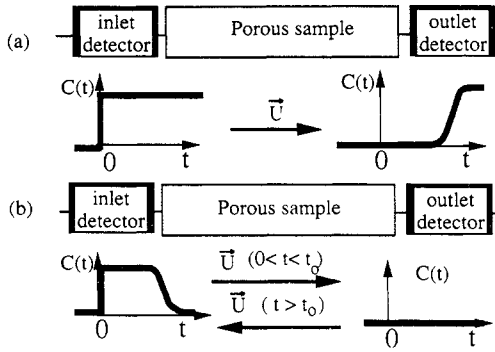‖ Present address: Département de Physique, Ecole Normale Supérieure de Lyon, 46 allée d'Italie, 69007 Lyon, France.

Figure 1. Schematic view of tracer dispersion experiments in a porous medium. (a) Classical transmission experiment: the spreading of the tracer concentration front is measured at the outlet of the sample. We assumed a step-like injection of the tracer. (b) Echo dispersion experiment: the concentration variation front first moves into the sample. It is then pumped back through a detector located at the inlet section.

dispersion coefficients and, except at very low flow velocities, are much larger than the molecular diffusion coefficient D_m; the relative magnitude of the convective and diffusive effects is given by the Péclet number Pe $= Ud/D_m$ (d is a characteristic microscopic length scale). Practically, we determine $D_\parallel$ by injecting tracer uniformly at the inlet of the sample (or by inducing a concentration variation) and measuring the variation $C(t)$ at the outlet (figure 1(a)). In heterogeneous samples, equation (1) is not verified [3, 4] and one computes a value of $D_\parallel$ from the width of the tracer spreading [5] ('non-Gaussian dispersion').

At flow rates such that Pe $\gg 1$, the dominant dispersion mechanism is the random variations in the fluid velocity from one pore to another. Dispersion appears as a random walk of tracer particles from pore to pore superimposed over the mean drift motion due to the global flow. $D_\parallel$ is in this case proportional to U [2, 6] with

$$D_\parallel = Ul_d. \tag{2}$$

l_d is the 'dispersion length' or 'dispersivity'; it represents the length of one step of the random walk and is equal to the Lagrangian correlation length of the fluid velocity [7]; it may be large in a heterogeneous medium where a particle keeps a significant deviation from the mean velocity while crossing a block of given permeability. In a homogeneous non-consolidated medium, on the other hand, l_d is of the order of the grain size [8]).

In previous work [9], we showed that, for sintered glass beads samples, l_d increases by a very large factor (50) with the degree of sintering but is constant with Pe. We believe that this is due to inhomogeneities in the sintering process and not to features of the microscopic structure of the medium. We also observed dispersion lengths l_d much larger than the grain size in natural Berea and Massilon sandstone samples (l_d = 2 mm and 1 cm for grain sizes of 50 μm and 100 μm). Similar results have been reported by other workers [2, 4, 5]. Let us now study the reversibility of the dispersion process.

3. Echo tracer dispersion

In the tracer dispersion experiment in figure 1(a), the concentration variation front moves all the way through the sample and is detected at the outlet. In echo experiments [10], it penetrates by a certain distance into the sample; then the flow direction is reversed and an echo concentration front is detected by a detector place at the inlet end (figure 1(b)). At each penetration depth, one determines an effective echo dispersion coefficient
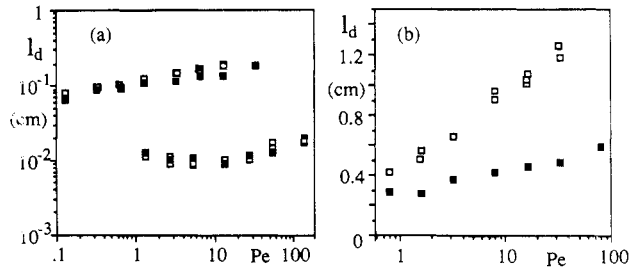
Figure 2. Dispersivity variations with the Péclet number in transmission (□) and echo (■) dispersion experiments. (a) Homogeneous samples giving Gaussian dispersion curves; the upper curve corresponds to a Berea sandstone sample and the lower curve to non-consolidated beads. (b) Heterogeneous Massilon sandstone sample giving non-Gaussian transmission dispersion curves that do not verify equation (1).

D_{ec} from the ratio of the mean square spreading and the mean transit time. Figure 2 displays variations with the Peclet number Pe of the transmission dispersion length l_d and the echo dispersion length $l_{dec} = D_{ec}/U$. For Gaussian samples where the transmission data verify equation (1), l_d and l_{dec} are equal over the whole range of Péclet numbers investigated (figure 2(a)). For more heterogeneous samples such as Massilon sandstone (figure 2(b)), l_{dec} is smaller than l_d by a factor of 2 or 3 and the relative difference increases with increasing Péclet number; however, l_{dec} is still much larger than the microscopic length scale. In addition, in the latter case, the echo dispersion curves are Gaussian although the transmission curve does not follow equation (1). One likely interpretation is that the heterogeneous sample contained large-size inhomogeneities such as high-permeability channels or strata. If, during the flow into the sample, transverse dispersion is sufficient for a tracer particle to move out of the channel, it retains the same velocity difference while flowing in the reverse direction and the spreading effect is cancelled. l_{dec} is then much smaller than l_d. Stratified media are a particularly useful model to examine such effects because they have two well defined length scales [11, 12]: the grain size and the layer thickness.

4. Dispersion in stratified media

We have studied a system made of five alternate layers of large (450–500 μm diameter) and small (355–400 μm diameter) glass beads with a total thickness of 3 cm [12]. The fluid flow is parallel to the plane of the strata. At high Péclet numbers, one observes clearly two distinct fronts in the transmission curves corresponding to the two flow velocities; the transverse mixing effect is weak in this case. The corresponding echo curve is much sharper; the tracer reaches back the origin at about the same time in both strata. At a very low Péclet number Pe = 0.5, transverse diffusion is sufficient to homogenize the concentration between layers and the transmission curve is Gaussian.

Figure 3 displays the variations in l_d and l_{dec} with the flow velocity. At low Pe values, l_d and l_{dec} are equal; their value is determined (as in Taylor dispersion in a capillary tube [13]) by a compromise between longitudinal convective dispersion and transverse dispersion. At high Péclet numbers, l_{dec} tends towards a value corresponding to dispersion in a single layer while l_d tends towards an upper limit determined by the permeability contrast and the path length. These variations are, as expected, similar to
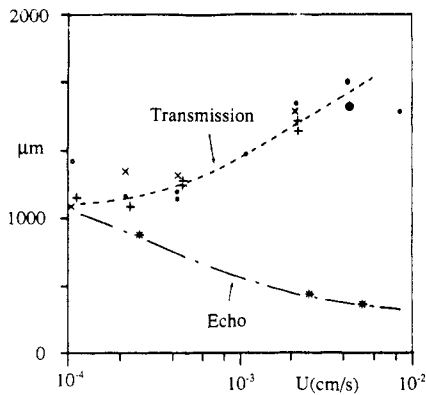
Figure 3. Variation in the characteristic length l_d with the Péclet number for echo and transmission dispersion measurements in stratified media made of five alternate layers of glass beads of small and large diameters (355–400 μm and 450–500 μm). The data points giving the largest values of l_d correspond to transmission experiments, and the smaller values correspond to echo curves.

those in figure 2(b). These results show that, from dispersion experiments on stratified materials, one can get information on the structure of individual layers, on the transverse dispersion and on the permeability contrast between layers. Comparing echo and transmission dispersion allows one to detect large-scale heterogeneities of a medium; it has been applied to the evaluation of the optimal resolution of gel chromatography columns.

5. Transition to irreversibility in homogeneous samples

The results obtained on the homogeneous Gaussian samples (figure 2(b)) seem to contradict the principle of the reversibility of flows at low Reynolds numbers; one explanation is that reversibility would appear only at small distances while, in the previous experiments, the path lengths are equal to several hundred diameters. We have therefore designed a system with a very low dead volume, allowing measurements for path lengths below one grain diameter [14]. The tracer is Fe^{3+} ions produced by an electrochemical reaction [15, 16] on a plane electrode grid stretched across the inlet section of a packing of glass beads with diameter $d = 4$ and 2 mm; initially, the sample is saturated with a solution containing only Fe^{2+} ions. Practically, we establish an inward flow in the sample, apply a short pulse of current to the electrode (creating a planar-like distribution of tracer ions) and let the tracer penetrate over a chosen distance L into the sample. At the time $T_{inv} \propto L$, the flow direction is reversed and the variations in tracer concentration with time on the grid are monitored by measuring the current necessary to reduce the Fe^{3+} ions back to Fe^{3+} ions. We obtain an 'echo' curve for each T_{inv}-value and compute an effective echo dispersion coefficient D_{eff} from the variance of the transit time.

Figure 4 displays the variation in $l_{dec} = D_{eff}/U$ with the ratio L/d. The data points correspond to three different Péclet numbers $Pe = Ud/D_m$ varying from 390 to 1570 and to $d = 4$ mm. Most of the variation in l_{dec} occurs over a range of penetration distances of $15d$ or so; this explains why these effects are not observed in the experiments in figure 2. The asymptotic dispersivity l_{dec}^{as} is equal to $(0.6$–$0.7)d$ for all Péclet numbers used in figure 4 and for both $d = 4$ mm and $d = 2$ mm. This value is of the order of that obtained through transmission measurements at the same velocities; dispersion is therefore totally irreversible at large distances but partly reversible for L-values below a few diameters.
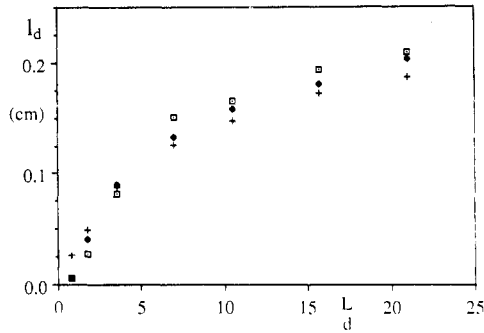
Figure 4. Variation of the dispersivity l_d as a function of the mean penetration distance L (normalized by the bead diameter d) in a homogeneous 4 mm glass bead packing: $\square$, Pe = 1570, Re = 1.13; $\blacklozenge$, Pe = 785, Re = 0.57; +, Pe = 390, Re = 0.28.

Because of the weak dependence of the curves of (figure 4) on Pe, molecular diffusion cannot be the only irreversibility mechanism. We suggest that it is 'enhanced' by the disorder of the flow in the medium which divides the tracer filaments along their path and makes them very sensitive to any kind of perturbation; in this case, one expects a logarithmic dependence with Pe of the L-value corresponding to irreversibility [14].

6. Conclusion

In conclusion, we have shown that tracer spreading is irreversible in homogeneous porous media as soon as the path length L exceeds a few grain diameters. In heterogeneous media, variations in the effective echo dispersion coefficient with L provide information on the transverse size of the heterogeneities. In media with strata or channels, dispersion is largely reversible when the transverse dispersion is sufficiently low that the tracer remains inside one strata or channel during a large fraction of its path. These processes are analogous to turbulent mixing; flow in porous media can indeed be considered as a frozen turbulence where only spatial disorder is present.

Acknowledgments

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