

● *Short Communication*

CHARACTERISATION OF FLUID FLOW THROUGH POROUS MEDIA USING THREE-DIMENSIONAL MICROIMAGING AND PULSED GRADIENT STIMULATED ECHO NMR

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Pulsed gradient stimulated echo (PGSTE) and microimaging nuclear magnetic resonance (NMR) are used to probe correlations between structure and flow in the void space of a model porous system formed from a packing of 1-mm diameter glass spheres. The pulsed gradient stimulated echo data determine the average propagator and permit the dispersion of the flow to be studied as a function of delay time. Microimaging yields structural information and, specifically, a reduced radial distribution function (rdf) for the structure of the void space. Transition to fully developed dispersive flow is shown to occur on a scale size for which no further correlations in the structure of the void space are observed. © 1998 Elsevier Science Inc.

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INTRODUCTION

The pulsed gradient stimulated echo (PGSTE) nuclear magnetic resonance (NMR) technique probes molecular motion by measuring directly the average displacement propagator, $\bar{P}_s(R, \Delta)$ (i.e., the probability that a molecule at any initial position is displaced by a distance R in a time Δ). This is achieved by encoding the position of a molecule using a pulsed magnetic field gradient of strength g and duration δ . Molecules moving a distance R during a time Δ suffer a phase shift $\phi = \gamma \delta g R$, where γ is the gyromagnetic moment of the nucleus under investigation. A second pulsed gradient applied after the time Δ produces a signal that is attenuated by an amount given by the echo attenuation function $E(q, \Delta)$:

$$E(q, \Delta) = \int \bar{P}_s(R, \Delta) \exp(i2\pi q R) dR,$$

where $q = (2\pi)^{-1} \gamma \delta g$ and an inverse Fourier transform then gives the average propagator. For short gradient pulses $\delta \ll \Delta$, the amplitude of the echo attenuation function can be approximated by:

$$|E(q, \Delta)| = \exp(-4\pi q^2 \Delta R D^*),$$

where $D^*(\Delta)$ is an effective dispersion coefficient. At long Δ , D^* tends toward the zero-frequency dispersion coefficient.²

The average displacement propagator is an ensemble average over all molecular motion in the sample; the information it provides is complimentary to magnetic resonance imaging of the velocity field that gives the spatial distribution of the mean fluid flow. Microimaging of the void space in which the flow is occurring enables all aspects of the flow to be related to the structure of the porous medium.

MATERIALS AND METHODS

The model porous medium investigated in this work was an unconsolidated packing of 1-mm diameter glass spheres packed within a 10-mm internal diameter glass tube. The packing was prepared by letting the glass spheres settle within the tube initially filled with deionised water. Flow through the packing was achieved using a constant pressure drop main-

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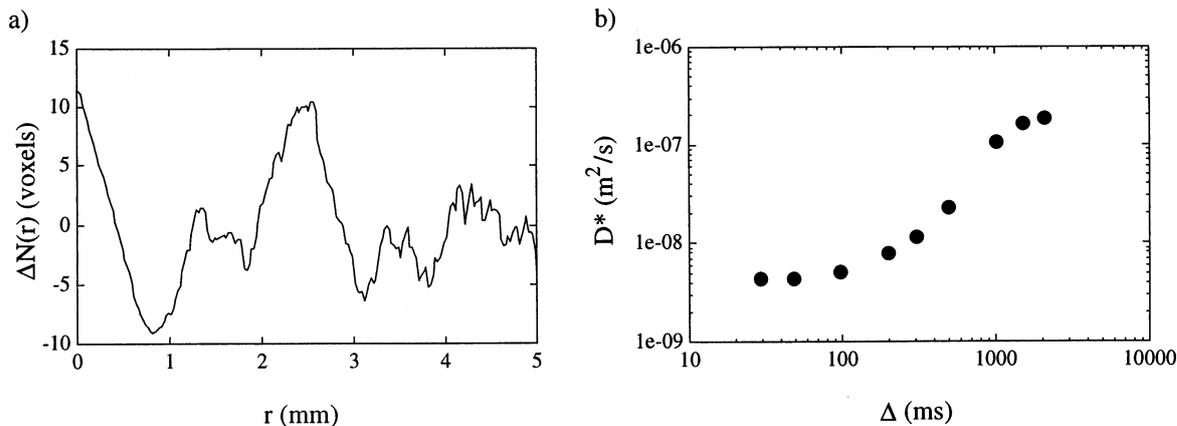


Fig. 1. (a) Reduced radial distribution function. (b) Variation of the effective dispersion coefficient with delay time Δ .

tained by two large reservoirs with a height difference of approximately 2 m. The flow rate was adjusted by means of a valve to give a volumetric flow rate of 70 mL h^{-1} corresponding to a mean interstitial velocity of 0.6 mm s^{-1} . NMR experiments were performed on a Bruker DMX 200 spectrometer with microimaging probe. The maximum field gradient achievable in the axial direction is 1.6 T m^{-1} . The packing was aligned with the direction of flow along the axis of the magnet. The altering PGSTE pulse sequence³ was used for the determination of the average propagators to minimise magnetic susceptibility effects. The pulse duration $\delta = 1 \text{ ms}$ was held constant during the experiments, and g was incremented in 128 equal steps between 0.0 and 1.6 T m^{-1} . Homospoil gradients were used to compensate for imperfect radiofrequency pulses. A three-dimensional image of the proton density in the void space was obtained using a standard three-dimensional imaging pulse sequence¹ yielding a cubic voxel of dimension 0.086 mm ; this volume image was then gated to produce a binary image representing void space and the glass beads.

RESULTS

To characterise the structure of the void space a radial distribution function (rdf) was calculated that gives the average density, $n(r)$, of void space as a function of radial distance r from any point in the void space. Figure 1a shows the reduced rdf $\Delta N(r) = 4\pi r^2(n(r) - n_0)$, where n_0 is the mean void space density. The negative correlation at about 0.8 mm corresponds to the transition from void space to solid material and therefore indicates the scale size for individual pores. There is a second strong positive correlation between radii of 2 and 3 mm ,

and structure in the reduced rdf has largely disappeared beyond 4 mm .

The PGSTE data show a number of features that can be related to the structure of the void space. Firstly, a peak in $E(q, \Delta)$ is seen at a value of q that corresponds to the negative feature at 0.8 mm in the rdf; the PGSTE experiment is therefore sensitive to the size of individual pores. The shape of the average propagator changes from exponential to a broad peak shape with increasing Δ in agreement with previous work.^{4,5} The characteristic time at which this transition occurs can be related to a scale size by considering the average interstitial velocity; this gives a scale size of about 1.5 mm , which corresponds to the featureless region of the reduced rdf. Figure 1b shows D^* as a function of Δ . A clear transition is seen yielding a plateau in D^* at large Δ ($\geq 2000 \text{ ms}$), which corresponds to a scale size greater than 4.2 mm . This scale size corresponds to the region of the rdf beyond which there is no structure.

CONCLUSION

The results presented in this paper demonstrate that PGSTE and microimaging are a powerful method of probing structure-flow correlations in porous media. In particular, it is shown that the transition to fully developed dispersive flow occurs on scales over which correlated structures in the void space disappear.

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REFERENCES

1. Callaghan, P.T. *Principles of Nuclear Magnetic Resonance Microscopy*. New York: Oxford University Press; 1991.
2. Seymour, J.D.; Callaghan, P.T. Flow-diffraction structural characterization and measurement of hydrodynamic dispersion in porous media by PGSE NMR. *J. Magn. Reson.* A122:90–93; 1996.
3. Cotts, R.M.; Hoch, M.J.R.; Sun, T.; Markert, J.T. Pulsed gradient stimulated echo measurements for improved NMR measurements in heterogeneous systems. *J. Magn. Reson.* A83:252–266; 1989.
4. Packer, K.J.; Tessier, J.J. The characterization of fluid transport in a porous solid by pulsed gradient stimulated echo NMR. *Mol. Phys.* 87:267–282; 1996.
5. Lebon, L.; Leblond, J.; Hulin, J.P. Experimental measurements of dispersion processes at short times using a pulsed field gradient NMR technique. *Phys. Fluids.* 9:481–490; 1997.