Laboratory measurement of hydrodynamic saline dispersion within a micro-fracture network induced in granite

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Abstract

We report the first measurements of hydrodynamic dispersion in a microfractured granite using a combination of novel techniques. A fracture network was induced in a cylindrical plug of Ailsa Craig micro-granite by thermal stressing, to produce an isotropic network of fractures with an average aperture of ~0.3 μm, a density of approximately 4 × 10^4 fractures/mm^3 and a permeability of 5.5 × 10^-17 m^2. After saturating the cores with 0.01 M NaCl solution a step in the concentration profile to 1 M was advected into the plug at flow rates of 0.07 to 2.13 cm^3 h^-1. The longitudinal electrical impedance of the plug was measured continuously as the solute front advected through its length until the plug was saturated with the concentrated electrolyte. Analysis of the impedance versus time relationships allows the derivation of the longitudinal dispersion coefficient, D_L, and hydrodynamic retardation, R_H. The Peclet number−dispersion relationship for the micro-fracture network is very similar to that predicted for other, radically different, fracture networks. Thus dispersion may be more dependent on fracture connectivity and length than fracture density and display a relationship similar to that shown by particle beds and clastic sandstones. The high retardation values observed (2.2–4.9) reflect flow behaviour within a fracture network with a proportion of ‘blind’ sections, and demonstrates how such networks can slow the advance of conservative solute components.

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1. Introduction

The movement and dispersion of contaminants in groundwater systems has been the focus of intensive investigation for more than 30 years (see Dagan and Neuman, 1997; Zimmermann and Main, 2004 for reviews). In an effort to maximise exploitation and preservation of groundwater resources much of this effort has concentrated on the description, quantification and
modelling of contaminant transport in shallow aquifers consisting mainly of highly porous sediments. However, with recent proposals to site radioactive and toxic waste repositories in crystalline rock formations (e.g. Bullen and McCormick, 1998), there has been increasing interest in investigating the hydraulic properties of rocks where permeability is controlled by fracture networks rather than flow within a porous matrix. To date, research into these areas has concentrated on numerical modelling with relatively few laboratory scale investigations on fracture networks in impermeable matrices. Grisak et al. (1980) measured ion breakthrough curves in large samples of fractured glacial till containing two orthogonal fracture sets. They found non-Fickian dispersion and attributed this to exchange and adsorption of ions within the porous matrix of the till. Permeability related to pervasive damage induced by thermal stressing has been used in a series of experiments by Darot and co-workers e.g. Darot et al. (1992).

Most laboratory studies have concentrated on flow in a single fracture. For example, Durham and Bonner (1994) have investigated flow in a single fracture under load, and Neretnieks et al. (1982) examined tracer movement in a single natural fissure in a granitic sample and found non-Fickian dispersion in non-sorbing tracers, ascribed to preferential channelling within the fracture plane. Moreno et al. (1985) also examined dispersion in a single fracture in granite in an experiment very similar to Neretnieks et al. (1982) and concluded that non-sorbing tracers can be modelled well by both hydrodynamic and channelling diffusive mechanisms. Despite these efforts to develop realistic theoretical models for flow in fracture networks, predictive capabilities remain severely limited as noted by Berkowitz and Scher (1997) and for full review, Berkowitz (2002). There is, therefore, a great need for laboratory scale studies both to establish the hydraulic characteristics of fracture networks, and to provide data sets with which to test the efficacy of theoretical models.

There are three main reasons why there is a paucity of laboratory tests dealing with fracture networks.

First, samples of competent crystalline rock that contain homogeneously distributed fracture networks at laboratory sample scale are rare. Where naturally fractured crystalline rocks do occur, the scale of the fracture systems is such that unmanageably large samples are required to achieve a statistically homogeneous domain. In addition, it is unlikely that the process of sample collection, such as coring, will leave the fracture network unmodified either by relaxation of the network or blockage of fractures by debris from the collection procedure.

Second, the measurement of dispersion profiles requires well-characterised experimental boundary conditions, which include true linear plug flow within the sample volume, and a well-defined initial condition, such as an instantaneous initial step change in fluid composition across the entire up-stream face of the sample. It is easy to demonstrate that the standard method of introducing fluid into a sample and collecting the advected pore fluid via central axial ports, with or without feeder channels on platen faces, will not result in radially symmetric plug flow along the sample axis. For this generally used experimental configuration, path length differences and slight pressure variations ensure that axial flow directly from central platen inlet to outlet dominates over flow down the periphery of the sample plug, producing dispersion effects unrelated to, and potentially masking, dispersion arising from within the sample fracture network.

Third, for a fractured sample to remain internally cohesive and competent, the volume percentage of the crack network must be low (e.g. <5%). This implies that, at the scale of typical laboratory test samples (~100 cm³) the volume of fluid expressing the dispersion profiles is very small. Dispersion effects being sought from within the sample are potentially masked by mixing and dispersion at the sample ends and within the fluid lines conveying the fluid to the point of analysis. Such effects can significantly degrade the true dispersion profile, and result in the final measured profile being a complex composite of dispersion from the sample and dispersion due to advection within the fluid lines.

Here, we report a novel series of techniques that circumvent the three problems outlined above. By applying impedance spectrometric measurement techniques to fluid flow within a specifically micro-fractured cylindrical sample of granite, we have determined chemical dispersion in the fracture network, and compared the results with theoretical models for real and model fracture networks.

2. Methodology

2.1. Test material and sample preparation

The material used in this study to form the matrix for the fracture network is Ailsa Craig microgranite (ACM) from the island of Ailsa Craig in the Firth of Clyde, Scotland. ACM has a porphyritic texture, with a fine-grained groundmass (mean grain size of 0.25 mm) and sparsely distributed microphenocrysts of alkali feldspar up to 1.5 mm. This material has been examined in detail
by Clint et al. (2001), who have shown it to have, in its natural state, no pre-existing microcracks observable by optical or scanning electron microscopy, either within or between grains. It has a primary porosity of 0.9%, mostly within the altered but unconnected feldspar microphenocrysts. The rock has virtually no secondary fracture porosity and as a consequence has an exceptionally low permeability \((1.5 \times 10^{-23} \text{ m}^2)\), and a very low degree of P and S wave velocity anisotropy (less than the measurement accuracy of 1%).

A microfracture network was induced in samples of ACM by thermal stressing, using techniques developed by Clint et al. (2001). Cores of ACM, 38.4 mm in diameter and 90 mm long, were heated in a tube furnace at 1 °C/min to 900 °C, whereupon the temperature was held for approximately 1 h, after which it was decreased at 1 °C/min to room temperature.

A scanning electron micrograph of a fracture surface of heat treated sample is shown in Fig. 1a. Analysis of a series of such images shows that the average aperture of the induced fracture network is 0.3 μm. The fracture pattern is characteristic of fracture sets formed by tensile failure. It is characterised by successive halving of volumes with each tension fracture nucleating approximately halfway along a previous fracture. This mechanism gives fractures with a majority of intersections of ‘T’ geometry (Fig. 1b). Clint et al. (2001) have shown that although the P and S wave velocities in this type of thermally stressed ACM decrease by an approximate factor of two, there is no resultant anisotropy in either velocity, implying there is no preferred orientation in the induced fracture network. This is confirmed by observation of the thermal fracture patterns (Fig. 1a). We assume, therefore, in the theoretical treatment to follow, that the fracture network is homogeneous and isotropic.

### 2.2. Experimental protocol

#### 2.2.1. Pressure vessel

For the current experiments a 38.4 mm diameter by 90 mm long cylindrical core of heat-treated Ailsa Craig microgranite was placed in a specially modified Hassler cell, shown diagrammatically in Fig. 2, and cyclically flushed at different flow rates as detailed below. The Hassler cell consists of a confinement vessel with a central tubular rubber jacket encasing the sample between the ends of a pair of load pistons. End load is applied to the sample via the load pistons by an external hydraulic ram within a straining frame, while a simultaneous radial confining pressure is applied by hydraulic compression of the central rubber tube within the pressure vessel. The experiments were carried out with

![Fig. 1. a. A scanning electron micrograph of heat treated Ailsa Craig microgranite showing the fracture network (scale bar at bottom of photograph). The equivalent net of fractures used in the calculation of excluded volume, \(V_{ex}\).

![Fig. 2. Schematic Hassler cell design modified to allow step changes in pore fluid and impedance measurements.](image_url)
the sample under a modest hydrostatic load of 2.5 MPa, sufficient to provide a seal between sample and confining jacket whilst not significantly collapsing the induced fracture network.

2.2.2. Inflow piston design

The inflow (lower) piston design, shown in Fig. 3a, incorporated three elements to facilitate true plug flow through the sample, and enable an ‘instantaneous’ step change in inlet fluid composition to provide robust boundary conditions for the experiments. The piston body was machined with two fluid ports, a central axial fluid access port and a second, peripheral drain conduit. Fluid movement between these ports was controlled by an overlying shallow distribution plate that allows the fluid to flow from the central port, across the face of the sample, and exit, via a peripheral circular channel, down the off-axis drain port in the piston body. To allow the fluid to sweep efficiently across the front of the core the distribution plate is separated from the core sample by a thin layer of stainless steel woven mesh. As this layer is highly permeable (porosity $\sim 50\%$) but of very low volume, a relatively small flow will rapidly sweep the front face of the core. The high porosity of the mesh also facilitates access of the fluid to all of the core face, promoting plug flow within the fracture network. Being the same composition as the contiguous piston, the gauze is electrically its continuation, and piston and gauze can be considered a single functional unit. This stainless steel woven mesh layer can only be used at low sample end loads.

2.2.3. Outflow piston design

The modified outflow (upper) piston is shown in Fig. 3b. The piston has a single, axial fluid exit port, and is separated from the core sample by a layer of fine stainless steel woven mesh to allow free exit of pore fluid from the core end. This minimises any tendency for flow focusing in the core adjacent to the exit port in the upper piston. To allow electrical measurements of the saline saturated ACM cores, the upper piston was insulated from the body of the steel pressure cell body by a plastic sleeve, and from the load frame by a plastic disc in the load stack. Thus impedance measurement between the inflow and outflow piston units measured solely the electrical response of the fluid-saturated test core.

2.2.4. Pore fluid delivery

The accurate measurement of dispersion profiles in porous media requires that variations in the fluid flow rate be kept as low as possible. This constraint becomes increasingly important as pore volume decreases. To minimise variations in the flow rate, fluid was supplied to the rig by an ISCO LC5000 syringe pump, actuated by a stepper motor giving a flow variation of $\ll \pm 1\%$.

2.3. Impedance spectroscopy measurements

Impedance spectroscopy is a powerful method of examining the distribution of electrically conductive media within a rock sample. Particularly, it provides information on connectivity and tortuosity, as demonstrated, for example, in the study by Garrouch et al. (2001) on tortuosity in porous media.
For a solution of moderate salt concentration, electrical conductance is proportional to salinity, and hence the electrical resistance of a bulk saline sample is proportional to the sample length, and inversely proportional to cross-sectional area and fluid salinity. For the microfractured granite of this investigation, flooded with saline solution, the overall conductivity of a sufficiently large sample can be represented by a random network of conductors corresponding to the conductive contribution of each connected fluid-filled fracture within the network. The conductivity of the granite matrix itself is many orders of magnitude lower than that of dilute NaCl solutions, and its contribution to overall conductivity can, therefore, be neglected.

For electrical measurement systems, it is convenient to consider electrical resistance or impedance, rather than its inverse, conductance. The saline-flooded fracture systems of the present samples can thus be represented by a hypothetical resistor network with values related to local fracture aperture and fluid salinity. Unfortunately, the DC resistivity of an ionic fluid cannot be measured directly because ion drift toward each electrode generates a blocking counter-voltage. Rather, an AC measurement is required, where an alternating voltage is applied at the sample ends, and the resultant current through the sample is measured. For a simple resistor the drive voltage and resultant current would be exactly in phase, but real samples of the type discussed show a small phase lag between voltage and current, arising from a small capacitative contribution within the fluid–rock–electrode system. The true resistive component can be calculated by examining the frequency dependence of the in and out-of-phase components of impedance, as discussed in Macdonald (1987), and calculating the functionally equivalent simple resistor–capacitor (RC) network.

AC impedance measurements were made using a Solartron SI 1260 impedance/gain phase analyser connected across the sample via the upper and lower pistons as discussed above. For each impedance measurement the computer-controlled analyser applied a logarithmic sequence of AC voltages from 1 MHz to 1 Hz sampling 10 points per decade. At each frequency the mean absolute values of the impedance $|Z|$ and the phase angles $\phi$ between voltage and current were recorded over a one second period. Impedance data were recorded with the Solartron Impedance Measurement Software (Version 1.3) and analysed using the software package ZView2. A typical frequency scan is shown in Fig. 4, with the calculated equivalent RC network values. In the following discussion we reserve the term ‘impedance’ for electrical impedance values measured by the Solartron 1260, and ‘resistance’ for the equivalent RC electrical resistances calculated from impedances via ZView2.

2.4. Experimental procedure

To measure the dispersion–time relationship for a given flow rate the following procedure was followed. The core was first saturated with degassed 0.01 M NaCl solution and its impedance measured: flooding with low salinity fluid as an initial condition avoids the mathematical problems and measurement errors which would arise from the extremely high electrical resistance of pure water. A very slow reverse flow (1 cm$^3$/h) of the 0.01 M NaCl fluid downward in the core was then begun, to block possible upflow into the sample. The bottom piston drain was then opened and 1 M NaCl solution pumped rapidly across the bottom face of the core to give a near-instantaneous change in the composition of fluid at the inflow end of the core. The 1 M NaCl pump rate was then reduced to the flow rate required for the experiment, the bottom piston drain valve closed, and the upper piston drain opened, allowing the 1 M NaCl fluid to begin advecting into the core. Impedance measurements were then made at suitable intervals to achieve some 200 measurements during the advection of the concentrated salt front upward within the core. As the 1 M NaCl front advected up the core, replacing lower-salinity fluid, the impedance of the core dropped, and the run was ended when a constant impedance value showed 1 M NaCl solution had completely flooded the core. During the run the fluid expressed from the core was collected to monitor total fluid flux. At the end of each run the sample was flushed continuously with the original 0.01 M NaCl.
solution for a minimum of 24 h, at which point a steady high impedance value had been re-attained.

3. Results

3.1. Mean cross sectional area of the fracture network and impedance–flow relationships

When loaded to 2.5 MPa confining pressure and saturated with a 1 M NaCl solution the core has a resistance of 700 Ohms. This is equivalent to that of a cylinder of 1 M NaCl solution 90 mm in length and 4.6 mm in diameter. The average total aperture of the connected fracture network is, therefore, 16.6 mm², or 1.4% of the cross-sectional area of the core. A true sample porosity of 5.3% was determined by weight loss on drying after careful water saturation. Sample permeability was measured as $5.5 \times 10^{-17}$ m².

Fig. 5 shows the resistance–time traces normalised to an arbitrary range between zero and one, obtained when IM NaCl fluid is introduced into the core at flow rates varying from 0.067 to 2.132 cm³ h⁻¹ corresponding to average fluid velocities of $1.1 \times 10^{-4}$ and $3.5 \times 10^{-3}$ m s⁻¹ respectively, relative to the electrically active cross-sectional area, with no tortuosity correction (see Table 1). The curves are clearly dissimilar to the typical sigmoid curve of a compositional breakthrough curve and consist of an initial approximately linear fall followed by an asymptotic section where the resistance decreases less rapidly towards a steady state value. In the runs at high flow rate the initial drop in resistance is more rapid and the asymptotic section shorter than those of slower flow rates. The minor fluctuations in slope during the initial resistance drop at the slowest flow rates are attributed to small variations in pump delivery rates at these low flow rates.

3.2. Interpretation of impedance time curves: a model for dispersion in the micro-fracture network

The shapes of the resistance–time curves are very different to the classic composition–time ‘breakthrough’

<table>
<thead>
<tr>
<th>Test</th>
<th>Flow rate (cc/hr)</th>
<th>Average linear velocity (cm/s)</th>
<th>Calculated linear velocity (cm/s)</th>
<th>Dispersivity $D_L/D_d$ Retardation precision Pe=λ(x)L/D</th>
<th>$D_L/D_d$ Retardation</th>
</tr>
</thead>
<tbody>
<tr>
<td>B42</td>
<td>0.067</td>
<td>$1.093 \times 10^{-4}$</td>
<td>$2.25 \times 10^{-5}$</td>
<td>$3.0 \times 10^{-6}$</td>
<td>$5.0 \times 10^{-8}$</td>
</tr>
<tr>
<td>B43</td>
<td>0.085</td>
<td>$1.391 \times 10^{-4}$</td>
<td>$3.30 \times 10^{-5}$</td>
<td>$5.2 \times 10^{-6}$</td>
<td>$1.3 \times 10^{-6}$</td>
</tr>
<tr>
<td>B45</td>
<td>0.129</td>
<td>$2.101 \times 10^{-4}$</td>
<td>$5.35 \times 10^{-5}$</td>
<td>$5.0 \times 10^{-6}$</td>
<td>$1.0 \times 10^{-6}$</td>
</tr>
<tr>
<td>B49</td>
<td>0.180</td>
<td>$2.941 \times 10^{-4}$</td>
<td>$8.00 \times 10^{-5}$</td>
<td>$2.75 \times 10^{-5}$</td>
<td>$2.0 \times 10^{-7}$</td>
</tr>
<tr>
<td>B47</td>
<td>0.282</td>
<td>$4.600 \times 10^{-4}$</td>
<td>$2.10 \times 10^{-4}$</td>
<td>$3.0 \times 10^{-5}$</td>
<td>$1.0 \times 10^{-5}$</td>
</tr>
<tr>
<td>B48</td>
<td>0.757</td>
<td>$1.237 \times 10^{-3}$</td>
<td>$4.20 \times 10^{-4}$</td>
<td>$5.5 \times 10^{-5}$</td>
<td>$1.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>B51</td>
<td>2.132</td>
<td>$3.483 \times 10^{-3}$</td>
<td>$1.10 \times 10^{-3}$</td>
<td>$1.0 \times 10^{-5}$</td>
<td>$2.0 \times 10^{-5}$</td>
</tr>
</tbody>
</table>
Curves, measuring fluid composition as it exits the sample. Impedance spectroscopy records the integrated electrical contribution of the entire sample. Thus, to interpret the resistance–time curves in terms of advection and breakthrough of a solute front in the core, we develop a new model for the interpretation of the resistive response to advection. The electrical resistance of a salt solution, at the concentrations used, is inversely proportional to concentration, and the fracture network containing the fluid is spatially homogeneous and isotropic at the measurement scale used. Therefore, the resistance response of the saline-saturated micro-fractured ACM can be modelled by a simple series resistor model, and the effective total resistance will be the sum of concentration dependent contributions along the length of the core.

At any point \((x)\) along the sample, the electrical resistance \((R_x)\) will be related to fluid saline concentration \((C_x)\) through

\[
R_x \propto \frac{1}{C_x}
\]  

(1)

It follows, therefore, that the bulk resistance of a sample of length \(l\) will be the integral of incremental resistances along its length:

\[
R_e = \int_0^l R_x \, dx \propto \int_0^l \frac{dx}{C_x}
\]  

(2)

Thus, if the concentration profile can be represented by an analytical relationship between concentration, time and distance, the impedance time response of a sample can be predicted and compared against measurements. Many studies of dispersion have identified complex, non-Fickian dispersion behaviour in fluids advecting through fracture networks. However, in this first study of impedance response to dispersion we test our observations against the assumption that the longitudinal dispersion of a solute front in the fracture network obeys the simple advection-dispersion equation (ADE, Freeze and Cherry, 1979).

The effective resistance at time \((t)\), therefore, will be related to the integral of the solution to the ADE for the given boundary conditions along the core with respect to length:

\[
R_e = K \int_0^l \frac{dx}{C_x} \left[ \text{erfc} \left( \frac{x-v_x t/R_x}{\sqrt{l D_L/R}} \right) + \exp \left( \frac{v_x t}{l^2} \right) \text{erfc} \left( \frac{x+v_x t/R_x}{\sqrt{l D_L/R}} \right) \right]
\]  

(3)

with the initial and boundary conditions

\[
C(x, 0) = 0, \quad C(0, t) = C_0, \quad t > 0, \quad 0 \leq x \leq l
\]

where \(C_x\) is the concentration at \(x\) \((x \leq l)\), \(C_0\) is the initial concentration, \(l\) is the length of the sample, \(v_x\) is the average longitudinal velocity. \(R_0\) is the retardation (hydrodynamic), \(D_L\) is the longitudinal dispersion coefficient and \(t\) time. \(K\) is the relevant constant of proportionality.

No analytic solution is available for Eq. (3), making inversion for model parameters difficult. The resistance–time response for the experiments was, therefore, modelled in forward mode through numerical integration, and application of a suitable integration constant to normalise the resistance–time curves to unity at \(t=0\). As detailed below, multiple computations with varying initial parameters were run to determine the best fit to the observed resistance–time data and derive values for longitudinal dispersion in the micro-fractured ACM. The quality of the fit, therefore, indicates how well the ADE describes the dispersion caused by flow through this fracture network.

The form of the family of resistance–time curves obtained in Fig. 5 is explained in Fig. 6, which shows movement of electrically conductive fluid within a sample, flowing with velocity \(V\). A step profile in the fluid concentration is imposed at time \(t_0\) and advances from left to right undergoing dispersion obeying the ADE. Prior to solute breakthrough at the right hand core end, impedance drops linearly with time because the concentration profile is always symmetric about the advected initial position (Fig. 6b). Once the concentration profile ‘breaks out’ of the end of the core the response curve becomes nonlinear (Fig. 6c), with form dictated by the relative rates of advection and diffusion. Finally, the sample becomes saturated with the new fluid composition (Fig. 6d), and the resistance of the sample trends asymptotically to a constant value, from which the mean connected aperture can be determined with confidence.

3.3. Example of fitting model to curve — derivation of retardation, \(R_f\) and longitudinal dispersion coefficient \(D_L\) for experiment run B47 (flow rate 0.282 cm\(^2\) h\(^{-1}\))

Fig. 7 shows typical normalised impedance–time data, in this instance for run B47, with two curves derived by integration of the solution to the ADE using different values of \(D_L\). It is clear from these plots that the value for \(D_L\) for run B47 lies between \(2 \times 10^{-5}\) and \(4 \times 10^{-5}\) cm\(^2\) s\(^{-1}\), and that the critical information lies in the nonlinear sections of the data. To establish the best
fit value for $D_L$, the square root of the sum of squares of the residuals between the observed and calculated impedance–time curves is calculated for each trial value of $D_L$ for a particular value of the retardation $R_H$. An example is shown in Fig. 8a. The residuals are not randomly distributed about the best-fit solution but vary slightly and systematically with time for each fit. The best bestfit solution was determined by minimising the sum of squares of the residuals across the entire curve. Fig. 8b shows how the sum of squares of the residuals varies with trial values of $D_L$. The clear minimum in the curve allows the optimal value for $D_L$ to be estimated, and indicates the degree of precision for each fit. The range in $D_L$ corresponding to ±0.1% in sum of squares of residuals is quoted along with the minimum residual fit for each run in Table 1.

To obtain convergence between the observed impedance–time relationships and those derived from the model, substantial values of the retardation term ($R_H$) in the ADE solution are required. In the present context, the retardation term $R_H$ does not arise from chemical sorption/desorption processes, which is the common historical usage of $R$ in the classical solution of the ADE (Freeze and Cherry, 1979). Rather, it is a delay term presumed to arise from hydrodynamic flow patterns in the microfracture network. Clearly, it is conceptually similar to delays arising from chemical processes in other flow systems, but to clarify the mechanistic differences causing the retardation process in the experiments reported here, we use $R_H$ as the appropriate variable, signifying hydrodynamic retardation. Fig. 9 shows that the relationship between $R_H$ and fluid velocity required for convergence is not random. As the average linear velocity of the fluid rises, the implied retardation falls rapidly to a minimum and then rises more slowly towards higher fluid velocities, as discussed below.

4. Discussion

As far as we are aware, this is the first study to report measurements of longitudinal dispersion and retardation in a real fractured rock sample by impedance spectroscopy. Two recent studies have, however, modelled dispersion in
real and model fractures and relate the dispersion to scalar properties of the network. Gonzalez-Garcia et al. (2000) serially sectioned a naturally fractured granite block (0.06 m$^3$) to reconstruct the three-dimensional characteristics of a natural fracture network. Using random-walk simulations they predicted the dispersion that would occur as a result of flow through the fracture network in three mutually perpendicular directions. Huseby et al. (2001), in a more theoretical approach, also used the random walk technique to determine the dispersion characteristics of a monodisperse array of intersecting hexagons. Both studies examine the relationship between the dispersion coefficient and fluid velocity by defining a Peclet number ($Pe$) for the fracture system:

$$Pe = \frac{V_x L}{D_L}$$  

(4)

where $V_x$ is the average interstitial velocity, $L$ is a length scale defined as the average lateral size of a fracture and $D_L$ is the longitudinal dispersion coefficient. Huseby et al. (2001) defined $L$ as the radius of the component circular fractures making up their network and it is therefore explicit in their study. In reconstructing the fracture network in three dimensions, Gonzalez-Garcia et al. (2000) were able to establish the value of $L$ by measurement for their network. The problem in the present study is to estimate adequately the appropriate value of $L$ from the 2-D pattern of fractures observed in SEM micrographs. This problem is directly analogous to that encountered in field-scale exposures where fractures and joints networks are only seen as essentially 2-D fracture patterns on near-planar exposure surfaces. Gonzalez-Garcia et al. (2000) examined this problem by comparing the measured characteristics of their network with those derived by assuming the fractures were randomly oriented disks of radius $L$. They report that the simple isotropic model provides a good estimate of $L$ to within a factor of two despite their network being significantly anisotropic. As the fracture network in micro-fractured ACM has a very low degree of anisotropy (Clint et al., 2001) an estimate of $L$ based on the application of the isotropic model should be even more robust.

An equivalent net of 12 fractures with 23 junctions was derived from the fracture pattern observed from the SEM micrograph shown in Fig. 1b. By counting the number of junctions with other fractures, the average number of junctions per fracture is 3.1. Because the network is known to have a low degree of anisotropy it can be assumed that these fracture characteristics are also valid in the directions normal to the SEM micrograph. This allows us to estimate the fracture density as $3.79 \times 10^4$ fractures/mm$^3$. If the network is modelled as an array of intersecting ‘penny-shaped’ disks the total surface area will be:

$$S = N_f \pi r^2.$$  

(5)

where $N_f$ is the number of fractures in the sample and $r$ is the radius of the disks making up the fracture network.
As the average aperture of the network is 0.3 \( \mu m \) and the total active volume of the fracture network can be estimated from the impedance analysis to be 1.572 cm\(^3\), we can estimate the surface area of the network, \( S \), to be 5.24 m\(^2\). From this we derive a value of 20.5 \( \mu m \) for \( L \). Gonzalez-Garcia et al. (2000) also derive two other important parameters for a network. The excluded volume, \( V_{ex} \), is defined as the volume surrounding an object in which the centre of another object must be in order for them to intersect (Balberg et al., 1984). For isotropic networks:

\[
V_{ex} = \pi^2 L^3
\]  

Substituting our value of 20.5 \( \mu m \) for \( L \) in Eq. (6) gives a value for \( V_{ex} \) of \( 8.5 \times 10^4 \) \( \mu m^3 \) for micro-fractured ACM. The concept of \( V_{ex} \) is important in the present context as it allows the calculation of the dimensionless fracture density, \( \rho' \), which is equal to the number of intersections per fracture:

\[
\rho' = N_i V_{ex}/V
\]  

where \( V \) is the total volume of the fractured matrix considered.

The value of 3.21 derived for \( \rho' \) for the micro-fracture network is close to the average number of junctions/fracture of 3.1 derived above giving confidence in the estimation of \( L \).

Using the value of 20.5 \( \mu m \) for \( L \) in Eq. (4) it is also possible to calculate the macroscopic Peclet number \( (Pe) \) for each of the flow rates (Table 1).

The relationship of \( Pe \) to \( D^* \), the ratio of \( D_L \) to the diffusion coefficient of NaCl in the fluid, is shown in Fig. 10. When considered as a single group the data for ACM forms a trend of rising \( D^* \) towards higher \( Pe \), the highest four data increasing proportionally to \( Pe \) with a correlation coefficient of 0.93.

Also plotted in Fig. 10 are the data derived by random walk simulation of the naturally fractured sample by Gonzalez-Garcia et al. (2000). These data fall very close to those derived in this study for micro-fractured ACM despite the radical differences in the two fracture networks (isotropically micro-fractured ACM has \( 3.96 \times 10^{13} \) fractures/m\(^3\) whereas the fracture network of Gonzalez-Garcia et al., 2000) has a fracture density of \( 6.6 \times 10^5 \) fractures/m\(^3\) in two sets of differing orientation). The results of a theoretical study of synthetic fracture networks by Huseby et al. (2001) are also shown in Fig. 9. Their results for a range of fracture densities between 2.5 and 12 fall at much lower values of dispersion for a given \( Pe \) but have similar slopes at \( Pe>1 \). Gonzalez-Garcia et al. (2000) noted this discrepancy and attributed it to the differences between the networks (a natural fracture network vs. an infinite periodic network of monodisperse hexagonal fractures) and the time interval for which the dispersion was modelled. As the \( \rho' \) value calculated for the network (3.21) examined in this study is based on estimates of the three-dimensional nature of the ACM network, the similarity between the dispersion behaviour in the two granites may be simply coincidental. However, if born out by further study of the ACM network, the similarity between our data and the conditional simulations of Gonzalez-Garcia et al. (2000) presents the interesting possibility that the dispersive properties of a network depends principally on the fluid velocity and degree to which the fracture segments intersect, and is less sensitive to fracture geometry or spatial distribution. This type of \( Pe-D^* \) relationship has long been known to hold for particle beds and well sorted sandstones of a wide range of particle sizes (Perkins and Johnson, 1963) due to the strong mean-field properties of close-packed near-spherical particles. It is obviously premature, however, to propose that the \( Pe-D^* \) relationship will hold for all fracture networks. In particular it can be hard to define a representative elemental volume, as assumed in Eq. (2), for fracture populations with higher correlation lengths and thus strong channelling characteristics. What is
clearly needed is to extend both the theoretical modelling and physical measurement of dispersion in a wide range of fracture networks under varying conditions to explore further whether this relationship is relatively simple, as in the case of sandstones, or is more complex than these limited results indicate.

NaCl, the solute chosen for the tests, is widely regarded as a conservative tracer and should not show significant retardation due to adsorption/desorption processes within the fracture network. However, de-convolution of the time–impedance curves requires significant retardation values (Table 1) that are dependent on fluid velocity. The highest retardation value (4.9) occurs at the lowest average linear fluid velocity and falls steeply with increasing fluid velocity to a minimum of 2.9 at an average linear velocity of 2.0 \times 10^{-4} \text{ cm s}^{-1}. From this point the retardation increases again at a more modest rate to a value of 3.2 at the highest fluid velocity of 1.1 \times 10^{-3} \text{ cm s}^{-1}. This behaviour can be interpreted in terms of the network containing a significant number of fractures that are not ‘through connections’ and so do not contribute to the permeability of the sample. At low fluid velocities, diffusion will cause significant loss of solute to these ‘blind’ or stagnant sections of the network and thus slow the rate of advance of the advection front. As the fluid velocity rises, however, the rate of diffusive loss from the front relative to its advection rate decreases, reducing the apparent retardation, as observed (Fig. 9). The subsequent increase in retardation with further increase in fluid velocity, however, cannot be assigned to diffusive loss. One possible explanation for this increase may be that the increasing fluid velocity initiates eddies in the flow paths, both within individual fractures and in fracture sets within the network. This process would cause localised negative fluid velocities on many scales within the network thus decreasing the average front velocity and increasing the apparent retention.

Huseby et al. (1997) have shown that the percolation threshold in random isotropic networks takes place at a dimensionless fracture density (\(\rho'\)) of 2.26 ± 0.04. As the \(\rho'\) of micro-fractured ACM has value of 3.2 it is clear that it is well above the percolation threshold and thus well connected. Many numerical studies of both natural and synthetic networks predict that non-Fickian dispersion is to be expected from well connected networks and attribute this behaviour to a combination of channelling and dead space within the network (e.g. Moreno et al., 1988; Koudina et al., 1998; Huseby et al., 2001). Why, then, does the micro-fractured ACM show the apparently disparate characteristics of high solute retention combined with near Fickian dispersion? As the retention rates are high it is clear that although the fracture network is well connected it must also have a large fraction of dead-space within it and thus must exhibit flow channelling. If, however, the fracture dead space is distributed such that no part of it is distant from a ‘live’ fracture then diffusion will ensure a rapid exchange of solute between it and a fluid flowing along the live fracture. In this case, rapid arrival and tailing in the breakthrough behaviour typical of non-Fickian dispersion will be minimal whilst retention will be substantial. As the correlation length of the fracture network is small relative to the core volume, however, the non-Fickian dispersion effects caused by channelling occur only on a small scale. Thus the macro-dispersion will be the average of local dispersions and thus will approach the ‘ideal’ Fickian behaviour.

5. Conclusions

We have described novel measurements, based on impedance spectroscopy, of dispersion in low porosity media where conventional techniques are prone to instrumental artefacts. The novel techniques we have adopted alleviate the necessity for chemical micro sampling and allow large numbers of measurements to be made during the development of dispersion profiles. We have applied these techniques to thermally fractured Ailsa Craig micro-granite and report the first determination of dispersion in a microfracture network. Applying a simple assumption about the three-dimensional nature of the network allows the results to be compared to those of Gonzalez-Garcia et al. (2000) who modelled dispersion in a naturally fractured block of granite. Despite large differences in geometry and density of the two fracture networks there is a striking similarity in the Peclet number–dispersion relationships and, in turn, to those observed in particle bed experiments. As our data are limited and derived, in part, from assumptions about the 3-D nature of the network it is too early to claim a simple relationship between dispersion and Peclet number similar to that shown by particle beds. However, the data present the possibility that dispersion is dependent more on the fracture network connectivity than the spatial distribution, and that, at least for the relatively low fracture density examined here, there is a simple relationship between dispersion and Peclet number, similar to that known for particle beds and sandstones. The fracture network has been shown to exhibit significant levels of retardation which cannot be ascribed to simple adsorption/desorption behaviour. The retardation is dependent on fluid velocity and it is suggested that, at low fluid velocities, this is due to diffusive loss of solute to ‘blind’ sections of the fracture network. Increasing
retardation towards higher fluid velocity may be due to back eddies in the flow within the network and thus may mark the onset of turbulent flow. Thus fracture networks similar to that studied here can have an interesting combination of characteristics of dispersion similar to sandstones but with the ability to retard even those contaminants that are regarded to be unaffected by adsorption/desorption processes.

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