Miscible transfer of solute in different types of rough fractures: from random to multiscale fracture walls heights.

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Abstract

Miscible tracer dispersion measurements in transparent model fractures with different types of wall roughness are reported. The nature (Fickian or not) of dispersion is determined by studying variations of the mixing front as a function of the traveled distance but also as a function of the lateral scale over which the tracer concentration is averaged. The dominant convective dispersion mechanisms (velocity profile in the gap, velocity variations in the fracture plane) are established by comparing measurements using Newtonian and shear thinning fluids. For small monodisperse rugosities, front spreading is diffusive with a dominant geometrical dispersion (dispersion coefficient $D \propto Pe$) at low Péclet numbers Pe; at higher Pe values one has either $D \propto Pe^2$ (*i.e.* Taylor dispersion) for obstacles of height smaller than the gap or $D \propto Pe^{1.35}$ for obstacles bridging the gap. For a self affine multiscale roughness like in actual rocks and a relative shear displacement $\vec{\delta}$ of complementary walls, the aperture field is channelized in the direction perpendicular to δ . For a mean velocity \vec{U} parallel to the channels, the global front geometry reflects the velocity contrast between them and is predicted from the aperture field. For \vec{U} perpendicular to the channels, global front spreading is much reduced. Local spreading of the front thickness remains mostly controlled by Taylor dispersion except in the case of a very strong channelization parallel to \vec{U} .

Key words: fractures, roughness, dispersion, multiscale, self-affine, shear-thinning

1. Introduction

The geothermal reservoir of Soultz-sous-Forêts, like most geological systems, contains structures of various size along which flow occurs: three main types of structures were identified: individual fractures, fracture clusters and major faults [1]. In order to understand these flow systems and help with managerial decisions, large scale numerical models incorporating such heterogeneities have been developed. Yet, when the transport of solutes is involved, the choice of a dispersion law (possibly scale dependent) valid at the scale of an individual fracture remains an open issue [2].

At this scale, tracer dispersion results from the combined action of the complex velocity field (varying both in the gap of the fracture and in its plane) and of mixing by molecular diffusion. The latter allows the tracers to move from one streamline to another and homogenizes the spatial distribution of the tracers. In the classical approach, tracer particles are assumed to perform a random walk superimposed over a drift velocity. The *Preprint submitted to Elsevier* latter is the average of the fluid velocity over an appropriate volume (the representative elementary volume or REV) while smaller scale variations induce tracer spreading. At the REV scale, the average $\overline{C}(x, t)$ of the tracer concentration over a section of the medium normal to the mean displacement satisfies the convection-diffusion equation [3]:

$$\frac{\partial \overline{C}(x,t)}{\partial t} = U \frac{\partial \overline{C}(x,t)}{\partial x} + D \frac{\partial^2 \overline{C}(x,t)}{\partial x^2}$$
(1)

where *D* is the longitudinal dispersion coefficient, \vec{U} the mean velocity of the fluid (parallel to *x*). The value of *D* (or equivalently of the dispersivity $l_d = D/U$) is independent of both time and the travelled distance: it is determined by the combined contributions of molecular diffusion and advection. The relative order of magnitude of these two effects is characterized by the Péclet number : $Pe = Ua/D_m$ (D_m is the molecular diffusion coefficient; *a* is a characteristic length of the medium (here the mean fracture aperture).

Recent experimental studies of breakthrough curves of October 26, 2018 solutes in natural fractures [4, 5, 6, 7] measured dispersion coefficients increasing linearly with the mean flow U (or with Pe). Moreover, the value of the dispersivity $l_d = D/U$ observed agreed with the predictions of a perturbation analysis [8]. These results suggested that dispersion is controlled (like in 3D porous media [3]) by spreading due to velocity variations associated to the geometry of the void structure. The latter determines the correlation length of the velocity field, leading to the so called geometrical dispersion regime. However, flow in fractures is known to be frequently concentrated in long channels of high hydraulic conductance [2, 9, 10]. The velocity remains then correlated over distances which may be too large for establishing a Fickian dispersion regime. These previous experiments were all performed for a fixed path length: however, in order to test the validity of the Fickian description one must measure the variation of the width of the mixing front with time tand check whether it increases, as expected, as $t^{1/2}$).

Another key factor is dispersion resulting from the flow profile in the gap of the fracture: the variation of the velocity between the walls (where it cancels out) and the middle of the gap (where it has a maximal value) stretches the solute front. This creates a concentration gradient across the gap which is balanced by transverse molecular diffusion. The decorrelation of the velocity of the solute is then determined by the characteristic time for the diffusion of solute particles across the gap. This differs from the geometrical regime in which the decorrelation is determined by the geometrical structure of the fracture. Then, the longitudinal dispersion coefficient scales like $D \sim Pe^2$ in this so-called Taylor dispersion (instead of $D \sim Pe$ for geometrical dispersion).

In fractures, both dispersion regimes are expected to coexist (see refs. [11, 12, 13]): at low Péclet numbers (but large enough to neglect pure molecular diffusion), dispersion is controlled by the disordered geometry, while, at higher ones, Taylor dispersion becomes the leading dispersion mechanism. Yet, the critical Péclet number characterizing the transition still has to be determined. Also, the robustness of this model when contact points between the fracture walls are present must be tested.

We discuss in this paper dispersion experiments dealing with these issues and carried out in transparent fractures with various degrees of heterogeneities. The geometries of the void space and the roughness of the walls of these models are described in Sec. 2.1. They range from a random wall roughness with a correlation length of the order of the aperture to a multiscale rough wall geometry similar to that observed in the field [14]; this latter case often leads to a strong flow channelization [10]. In the present models a relative shear displacement $\vec{\delta}$ of complementary matching rough walls is introduced: high aperture channels oriented normal to the displacement and spanning over the fracture are then created leading to an anisotrope aperture field [15, 16]. This phenomenon increases with the magnitude of $\vec{\delta}$ and becomes noticeable as soon as δ is of the order of the mean aperture [17]. The influence of the contact area between the fracture walls was also investigated by performing flow experiments in a transparent model fracture with an array of contact points.

In order to address these various issues, dispersion has been studied as a function of :

• the distance traveled by the tracer.

• the lateral scale of observation in the fracture plane over which the concentration is averaged. This scale ranges from a (meso)microscopic scale (*i.e.* the typical fracture aperture) up to the fracture width.

• the fluid rheology in order to determine, without ambiguity, the main mechanisms controlling the dispersion: *i.e.* velocity profile in the fracture gap or velocity fluctuations in the fracture plane.

This contrasts with previous measurements realized at the outlet of the samples and in which the development of the mixing region and its spatial structure cannot be investigated.

2. Experimental setup and procedure

2.1. Experimental models and injection set-up

• Model 1: this model (see ref. [18] for details) has two transparent surfaces of size 350×120 mm without contact points. The upper one is a flat glass plate and the lower one is a rough photopolymer plate. The wall roughness corresponds to randomly distributed cylindrical obstacles of diameter $d_o = 1.4$ mm and height 0.35 mm protruding out of the plane surface. The minimum aperture a_m of the model is the distance between the top of the obstacles and the flat glass plate with $a_m =$ 0.37 ± 0.02 mm; the maximum and mean values are respectively $a_M = 0.72\pm0.02$ mm and $\bar{a} = 0.65\pm0.02$ mm.

• Model 2: this model uses a periodic square array of obstacles of similar size as in model 1 but of rectangular and variable cross section and with their top in contact with the top plate. Flow takes then place in a two dimensional network of channels of random aperture (see ref. [19] for a full description). The model contains 150×140 channels (real size 150×140 mm) with an individual length equal to l = 0.67 mm and a depth $a_M = 0.5$ mm; their average width is $\bar{w} = 0.33$ mm and its standard deviation $\sigma(w) = 0.11$ mm. Following the definition of Bruderer and Bernabe [20], the degree of

heterogeneity of the network can be characterized by the normalized standard deviation $\sigma(w)/\bar{w}$. In the present work : $\sigma(w)/\bar{w} \simeq 0.3$.

• model 3: Models 3, 4 and 5 have complementary self-affine walls of size 350×90 mm, reproducing the roughness of natural fractures (see ref. [21]). In model 3, a relative shear displacement $\delta = 0.75$ mm parallel to the direction of the flow is applied between the walls. the mean of the fracture aperture is $\bar{a} = 0.75$ mm and its standard deviation is $\sigma_a = 0.11$ mm. This shear configuration is referred to as $\vec{\delta} \parallel \vec{U}$.

• model 4: In order to analyze the influence of the direction of the shear displacement, the direction of the shear for model 4 is now perpendicular to the direction of the flow (the corresponding standard deviation of the aperture is $\sigma_a = 0.15$ mm). This configuration (and that of model 5) is referred to as $\vec{\delta} \perp \vec{U}$. All other characteristics (wall size, mean aperture, map of the roughness of each wall, amplitude $\delta = 0.75$ mm of the shear displacement) are identical to those of model 3.

All models are transparent and placed vertically with their open sides horizontal. The upper side is fitted with a leak tight adapter allowing one to suck the fluids at a constant flow rate. The lower open side can be dipped into a bath containing the liquid. When the pump is switched off, the bath can be lowered before changing the fluid inside it. This allows one to obtain a flat initial front between the fluids (See Figure 1 in ref. [21]).

The models are illuminated from the back by a light panel and images are acquired using a high resolution camera. The pixel size is around 0.2 mm, i.e. lower than the typical fracture aperture. About 100 images of the distribution of the light intensity I(x, y, t) transmitted through the fracture are recorded at constant intervals during the fluid displacement using a digital camera with a high dynamic range. Reference images with the fracture saturated with the clear and dyed fluids (dye concentration c_0) are also recorded before the experiments and after the full saturation by the displacing fluid. A calibration curve obtained independently through separate measurements is then used to map the local relative dye concentration $0 \le c(x, y, t)/c_0 \le 1$ (in the following, c_0 is omitted and c(x, y, t) refers directly to the normalized dye concentration). The two fluids are of equal density: this is verified by performing twice the experiments at each flow rate value with the dyed fluid either displacing or displaced by the clear fluid. Comparing the results allows one to to detect possible instabilities induced by residual density differences (the corresponding experiments are discarded). The two fluids are, of course, miscible and have the same viscosity.

| Fluids | n | $\dot{\gamma_0}$ | μ_0 |
|------------|---------------|-------------------|----------------|
| | | s^{-1} | mPa.s |
| W-Glycerol | 1 | _ | 10 |
| 500 ppm | 0.38 ± 0.04 | 0.077 ± 0.018 | 410 ± 33 |
| 1000 ppm | 0.26 ± 0.02 | 0.026 ± 0.004 | 4500 ± 340 |

Table 1: Rheological parameters and Péclet numbers for the 500 and 1000 *ppm* scleroglucan solutions solutions used in the present work. W - Glycerol refers to the water glycerol mixture.

2.2. Fluids preparation and characterization

The solutions used in the present work are either a Newtonian water-glycerol mixture or shear-thinning water-polymer (scleroglucan) solutions with a 500 or 1000 ppm polymer concentration. In all cases, the injected and displaced fluids have identical rheological properties. The Newtonian solution contains 10% in weight of glycerol and has a viscosity equal to 1.3×10^{-3} Pa.s at $20^{\circ}C$. The preparation and characteristics of the shear-thinning solutions are the same as reported in ref. [21]. The variation of the viscosity μ with the shear rate $\dot{\gamma}$ is well fitted by the Carreau function:

$$\mu = \frac{1}{(1 + (\frac{\dot{\gamma}}{\dot{\gamma}_0})^2)^{\frac{1-n}{2}}} (\mu_0 - \mu_\infty) + \mu_\infty.$$
(2)

The values of the rheological parameters characterizing the fluids are listed in Table 1. For the non Newtonian fluids and at low shear rates $\dot{\gamma} \lesssim \dot{\gamma_0}$, the viscosity is constant like for a Newtonian fluid with $\mu \simeq \mu_0$ (Newtonian plateau regime). At higher shear rates $\dot{\gamma} \gtrsim \dot{\gamma}_0$, the viscosity follows a power law: $\mu \propto \dot{\gamma}^{(n-1)}$. Practically, μ_{∞} is taken equal to 1 mPa.s, *i.e.* the viscosity of water (the solvent): this limiting value would indeed only be reached at shear rates above the experimental range. The two main dispersion mechanisms *i.e.* Taylor dispersion $(D \sim Pe^2)$ and geometrical dispersion $(D \sim Pe)$ are affected in opposite directions when a Newtonian fluid is replaced by a shear thinning solution. More precisely, the velocity contrasts between different flow paths are enhanced for a shear thinning fluid, resulting in an increase of the geometrical dispersion (without modifying the scaling law $D \sim Pe$). By contrast, the velocity profiles in the gap become flatter: this reduces therefore Taylor dispersion, but still with $D \propto Pe^2$. Varying the fluid rheology modifies the relative influence of the two main dispersion mechanisms in opposite ways: the dominant one can therefore be identified unambiguously for each fracture geometry and flow rate.

3. Experimental results

3.1. Fracture model 1

In this model, flow takes place in the free space between a flat plate and a second one with protuberant obstacles. The latter perturbs the flow velocity field: the local mean fluid velocity (averaged over the gap) is greater between the obstacles, where the aperture is largest than at their top, where it is minimal. These mean velocity variations in the fracture plane result in geometrical tracer spreading. As for the velocity profile in the fracture gap, it induces Taylor like dispersion. The variation of the dispersivity $l_d = D/U$ as a function of *Pe* confirms that it is the sum of the two contributions discussed above with:

$$\frac{l_d}{a} = \alpha_G + \alpha_T P e, \tag{3}$$

where $\alpha_T Pe$ corresponds to Taylor dispersion and α_G to geometrical dispersion. For a fracture with two flat parallel plates and a Newtonian fluid, one has: $\alpha_G = 0$ and $\alpha_T = 1/210$; also, one has $\alpha_G \neq 0$ only for fractures with rough walls. Moreover, if the correlation length of the velocity field is small compared to the fracture size and if the ratio ϵ of the amplitude of the velocity fluctuations to the mean velocity U is small, then the perturbation theory predicts that $\alpha_G \propto \epsilon^2$ (a complete expression of α_G is given by Eq. (3) of ref. [18]).

Experimental dispersivity variations as a function of Pe are plotted in Fig. 1 for the three fluids. These data sets are well adjusted (see lines in Fig. 1) by functions of the type shown in Eq. (3): the dispersivity increases at first slowly with Pe above $Pe \simeq 20$ from a nearly constant plateau value before displaying a linear variation at higher velocities. The plateau value corresponds to α_G in Eq. (3) and increases with the polymer concentration. It can be shown that the amplitude of the velocity fluctuations is larger for shear thinning fluids: for a power law dependence of the viscosity on the shear rate $(\mu \propto \dot{\gamma}^{(n-1)})$, the parameter ϵ would increase theoretically by a factor (1 + 1/n)/2 compared to a Newtonian fluid. The velocity fluctuations (and, as a result, the dispersivity) increase therefore when n decreases *i.e* when the shear thinning character of the fluids is stronger. Unlike α_G , the parameter α_T for shear-thinning fluids is lower than the Newtonian value 1/210 (see [18]).

The values displayed in Fig. 1 were obtained by fitting the local concentration variation on each individual pixel by solutions of Eq. (1). A similar analysis was performed on the average of the local concentrations over the fracture width. The results are displayed by empty symbols in Fig. 1: they almost fall on the filled symbols



Figure 1: Variation of the experimental dispersivity l_d as a function of the Péclet number in model 1. (\Box): water-glycerol solution; (\triangle): 1000 ppm, (\circ): 500 ppm polymer solutions. Solid, dotted and dashed lines: fit of the respective data with Eq. (3).

demonstrating the lack of large scale heterogeneities in this model fracture.

3.2. Fracture model 2

In this model, the obstacles extend over the full gap height and mimic gouge particles created by the failure of the rock and evenly distributed in the fracture (see Sec. 2.1). The model appears then as a plane array of channels of random width: it can be considered as a 2D porous medium in which the pores correspond to the junctions between the channels. We show now that mixing at these pore junctions has a crucial influence on dispersion.

Fig. 2 displays variations of the dispersivity l_d with the Péclet number deduced from time variations of the local concentration at the pore scale (filled symbols) and of its average over the fracture width (open symbols); it is seen that the values of l_d obtained in both cases are similar so that, in the following, only global measurements will be discussed.

For Pe < 10, $l_d = D/U$ is nearly constant (*i.e.* $D \propto Pe$), suggesting dominantly geometrical dispersion. As discussed in Sec. 3.1, the value of l_d in this regime should depend strongly on the rheology of the solution: more precisely, it should increase with the polymer concentration as indeed observed here (like for model 1).

For Pe > 10, a second dispersion regime is observed, in which l_d increases with Pe. Furthermore, the linear trend observed in a log-log coordinate shows that l_d follows a power law of Pe (more precisely, $l_d \propto Pe^{0.35}$ for Pe > 10). This result is in agreement with numerical simulations by Bruderer and Bernabe [20] and differs from the Taylor dispersion regime $l_d \propto Pe$ observed in model 1 at high Pe values.



Figure 2: Variation of the dispersivity l_d (mm) with the Péclet number for experiments with water-polymer solutions : (\Box) , (\blacksquare) : 500 ppm concentration - (\circ), (\bullet) 1000 ppm. Open (resp. filled) symbols : averaging interval : 35 (resp. 0.4) mesh sizes. Dashed lines : Mean dispersivity values for the geometrical dispersion regime. Dotted line: power law fit of the variation for Pe > 10 (exponent 0.35 ± 0.03).

This difference is explained by the influence of the pore junctions. At low flow velocities (typ. Pe < 10), tracer particles can explore effectively the local flow field by molecular diffusion during their transit time through a given junction: this distributes evenly the tracer concentration inside it which represents a perfect mixing condition. Then, the tracer concentration is equal in all outgoing paths and the probability to follow one of them is proportional to the corresponding flow rate [22, 25]. Therefore, in this regime, dispersion is controlled by the disordered geometry of the array of channels.

At higher *Pe* values (typ. *Pe* > 10), mixing at the junctions is no more perfect and the tracer concentration in slower channels (like those transverse to the mean flow) is lower compared to the perfect mixing situation. The dispersion characteristic becomes more similar to the case of capillary tubes (representing the fast flow channels) oriented along the flow direction. In this case, one would observe Taylor dispersion with $l_d \propto Pe$ (or $D \propto Pe^2$) but the influence of flow redistribution at the junctions is quite large: this leads to a variation of l_d as $Pe^{0.35}$ intermediate between those observed in the geometrical and Taylor regimes.

3.3. Fracture model 3

Like in model 1, the walls of this fracture do not have any contact point but, in contrast with it, the rugosities of the wall have been selected to reproduce the multiscale roughness of most natural fractures (see Sec. 2.1). Such fractures are known to display high aperture channels perpendicular to the relative shear displacement $\vec{\delta}$



Figure 3: Variation of the normalized dispersivity l_d/d as a function of *Pe* for model 3 and two different polymer concentrations. (•, \blacksquare): global dispersivities determined from concentrations averaged over the fracture width. (\circ , \Box): local dispersivities determined from concentration variations on individual pixels. Lines: Taylor dispersion for plane parallel walls with the same mean gap as for model 3. (\circ , •), dotted line: 1000ppm polymer solution; (\Box , \blacksquare), dashed line: 500ppm polymer solution; continuous line: Newtonian solution. Insert: variation of the ratio of the local and global dispersivities as a function of the Péclet number. (\Box): 500 ppm solution. (\circ): 1000 ppm solution.

of the walls; they are characterized by an anisotropic permeability field with a larger permeability in the direction parallel to the channels. While most studies of these systems have dealt with their permeability, little is known about the influence of such a structure on tracer dispersion.

In model 3, flow is parallel to $\vec{\delta}$ (*i.e.* normal to the channels): in this case ($\vec{\delta} \parallel \vec{U}$), both the local and global concentration variation curves are well adjusted by the solution of the convection-dispersion equation (1). Moreover, dispersivity values determined from these curves become constant after a long enough path inside the fracture. Like for models 1 and 2, the dispersion process is therefore Fickian. Fig. 3 displays variations of both the local and global dispersivities with *Pe* for the two polymer solutions. Theoretical Taylor dispersivities for a fracture of same mean aperture with plane smooth walls and for the different fluid rheologies are also plotted in Fig. 3 as dashed and dotted lines (differences between these curves reflect the effect of the velocity profile in the gap).

For Pe > 12, the local dispersivity increases with Pein qualitative agreement with theoretical expectations $(l_d = D/U \sim Pe)$ and is also lower for the strongly shear-thinning 1000 ppm solution (open symbols). For both solutions l_d is larger than predicted, particularly for the 500 ppm solution for which it is close to the Newtonian value. This may be due to the vicinity of the "plateau" domain of the rheological curve in which the solution behaves like a Newtonian fluid at low shear rates. For $Pe \sim 12$, in which both solutions should be in this "plateau" regime, the dispersivities are, as expected, the same for the two solutions but still slightly higher than the theoretical value. At Pe < 10, l_d rises again due to the influence of longitudinal molecular diffusion and its value is also the same for the two solutions (the (\circ) and (\Box) symbols coincide).

These value of the local dispersivity are compared in Fig. 3 to the global dispersivities determined from time variations of the concentration averaged over the fracture width (filled symbols): as seen in Fig. 3 and its inset, the local dispersivities are significantly smaller (at a same Péclet number and for a same solution). The front contours (c = 0.5) displayed in Figs. 4a and b for model 3 reveal fine structures of the mixing front: they reflect fluctuations of the velocity induced by the fracture wall roughness. Their magnitude is large enough to account for the additional increase of the global dispersivity with respect to pure Taylor dispersion (compared to local dispersion) but not enough to allow for the observation of a geometrical dispersion regime.

To conclude, in model 3 with $\vec{\delta} \parallel \vec{U}$, dispersion is mostly controlled by the Taylor dispersivity component due to the velocity profile between the walls as soon as $Pe \gtrsim 12$; there is however an amplification of the dispersion due to the fracture roughness.

3.4. Fracture model 4

In model 3, the mean flow was perpendicular to the channels or to the ridges induced by the shear displacement: the correlation length of the velocity is then determined by the typical width of these structures. Model 4 has the same size as model 3, a same mean aperture and complementary rough walls with a self-affine geometry exactly identical to that used for model 3. However, the shear $\vec{\delta}$ is, this time, perpendicular to the mean flow \vec{U} . In this configuration $(\vec{\delta} \perp \vec{U})$, \vec{U} is parallel to the channels and ridges created by the shear: the correlation length of the flow velocity is then determined by the length of the channels which is much larger than their width.

The dispersion characteristics are then very different as can be seen by comparing isoconcentration fronts obtained for model 4 (Figs. 4c-d) and model 3 (Figs. 4ab) at different times and in identical experimental conditions. More precisely, large fingers and troughs are observed for model 4 while none appears for model 3. Also, the amplitude of these features parallel to \vec{U} is larger at the higher velocity for which the solution has a shear-thinning behaviour (Fig. 4d) than at the lower velocity at which it behaves like a Newtonian



Figure 4: Experimental isoconcentration fronts (c = 0.5) as a function of the normalized distance $x/\overline{x(t)}$ (\overline{x} = mean front distance) for different ratios α of the injected volume to the pore volume (a)-(b): fracture model 3 ($\vec{\delta} \parallel \vec{U}$); (c)-(d): fracture model 4 ($\vec{\delta} \perp \vec{U}$). Mean velocities: (a)-(c): U = 0.0125 mm/s, Pe = 14; (b)-(d): U = 0.25 mm/s, Pe = 285. (dots: $\alpha = 0.85$, dash-dots: $\alpha = 0.65$, dash-dot-dots : $\alpha = 0.5$, dashes: $\alpha = 0.15$). Continuous line: theoretical variation from Eq. 4. All experiments have been realized with identical 1000 ppm water-polymer solutions.

fluid (Fig. 4d). Another important feature is the good collapse of the large features of the front observed at different times when normalized by the mean distance: this shows that the size of these features parallel to the flow increases linearly with time. Such a collapse is not apparent in model 3, except near the sides of the model where they likely reflect wall effects.

These results show that front spreading is purely convective and that the total width Δx of the front parallel to \vec{U} (*i.e.* the distance between the tips of the fingers and the bottom of the troughs) increases linearly with distance as $x \Delta U/U$ ($\Delta U/U$ = typical large scale velocity contrast between the different channels created by the shear).

In order to predict these contrasts, we modelled the fracture aperture field as a set of independent parallel channels of aperture $a(y) = \langle a(x, y) \rangle_x$ ([23, 24]). A particle starting at a transverse distance *y* at the inlet is assumed to move at a velocity proportional to $a(y)^{(n+1)/n}$; The theoretical profile $x_f(y, t)$ of the front at a time *t* is then:

$$x_f(y,t) = \frac{\overline{x(t)} a(y)^{(n+1)/n}}{< a(y)^{(n+1)/n} >_y},$$
(4)

where $\overline{x(t)} = \langle x_f(y,t) \rangle_y$ and $\langle a(y)^{(n+1)/n} \rangle_y$ are averages over *y* of the local aperture a(x,y). Normalized profiles $x_f(y,t)/\overline{x(t)}$ computed using Eq. (4) and the actual aperture fields are plotted in Figs. 4a to d as con-

tinuous lines. The exponent *n* has been taken equal to 1 at the lowest velocity for which $\dot{\gamma} \sim \dot{\gamma}_0$ (Figs. 4a-c) and to 0.26 at the highest one for which $\dot{\gamma} > \dot{\gamma}_0$ (Figs. 4b-d) (as mentioned in Sec. 2.2, $\dot{\gamma}_0$ is the shear-rate value corresponding to the crossover from the Newtonian to the shear-thinning behaviour of the fluid).

Eq. (4) clearly predicts well the location and shape of the large "fingers" and "troughs" at both velocities for $\vec{\delta} \perp \vec{U}$. In contrast, the theoretical curves does not reproduce the front geometries in model 3 ($\vec{\delta} \parallel \vec{U}$) except for the small global slope.

This confirms that, for $\vec{\delta} \perp \vec{U}$ (model 4), the large scale features of solute transport are determined by the velocity contrasts between the channels created by the shear. The curves of Figs. 4c-d also reproduce well the difference between the sizes of the fingers at the two velocities investigated. This confirms that the difference between these sizes may be accounted for by the different rheological behavior of the fluid : the velocity contrasts (and, therefore, the size) are amplified for Pe = 285 (shear-thinning power law domain) compared to the vicinity of the Newtonian constant viscosity regime (Pe = 14).

For model 3, the hypothesis of the model are not satisfied and it does not reproduce the front geometry: however, the features of the front are generally visible at similar transverse distances *y* in Figs. 4a and 4b (at a given time). They reflect likely also in this case a convective spreading of the front due to velocity contrasts between the flow paths: however, there is no simple relation of the front geometry to the aperture field, in contrast with model 4.

The local dispersivity $l_d(x, y)$ has also been determined for model 4 from the variations of the concentration on single pixels: its values are overall larger and their distribution is much broader than for model 3.

The same measurements have been performed [21] on a model fracture with a similar wall geometry but with a smaller amplitude $\delta = 0.33$ mm of the shear (still with $\vec{\delta} \perp \vec{U}$). In this case, the values of the local dispersivity are very close to those predicted from Taylor dispersion. Thogether with a smaller amplitude of the large scale fingers, this reflects a weaker disorder of the flow field.

4. Discussion

The experiments reported in this paper for several model fractures demonstrate the key influence of wall roughness geometries on the dispersion processes and their dependence on Pe and on the fluid rheology. One can group the results in two sets:

• models 1 and 2: both models correspond to obstacles with a single characteristic size. The height of the obstacles is smaller than the aperture for model 1 and equal to it in model 2: this models the case of gouge (or proppant) particles bridging the gap. In both cases the variation with distance and time of the tracer concentration satisfies the convection dispersion equation (1); the values of D are independent of the fraction of the width of the model over which the concentration is averaged and also of the distance from the inlet.

At low Péclet numbers, one has, in both cases, $D \propto U$ corresponding to geometrical dispersion due to the disorder of the velocity; in this regime, $l_d = D/U$ increases with the polymer concentration (*i.e.* with the shear-thinning character of the fluids) due to an enhancement of the velocity contrasts. Moreover, for model 1, the value of l_d is close to that predicted from a small perturbation theory. At high Péclet numbers, there is, for model 1 a transition towards Taylor dispersion with $D \propto Pe^2$. In model 2, D increases at high Pe values as $Pe^{1.35}$: this exponent agrees with previous numerical simulations [20] and should depend on the distribution of the size of the obstacles. In model 2, the transition between the different regimes is controlled by mixing at the scale of individual junctions.

• models 3, 4 and 5: The roughness of the walls of these models has a multiscale self-affine geometry similar to that of many fractured rocks; the walls of these fractures are complementary with a relative shear displacement either parallel ($\vec{\delta} \parallel \vec{U}$) for model 3 or perpendicular ($\vec{\delta} \perp \vec{U}$) for models 4 and 5. The relative shear produces a channelization perpendicular to $\vec{\delta}$ of the aperture field: as a result, dispersion depends strongly on the relative orientation of $\vec{\delta}$ and \vec{U} .

For $\vec{\delta} \perp \vec{U}$, the global spreading of the mixing front is not dispersive. The global width of the front parallel to \vec{U} increases instead linearly with time and reflects directly the velocity contrasts between the channels created by the shear. The large scale structures of the front can be predicted from the aperture field and their size increases with the shear-thinning character of the fluid. The variation of the local thickness of the front remains instead dispersive, but with a magnitude larger than for Taylor dispersion. For model 3 ($\vec{\delta} \parallel \vec{U}$), the global spreading of the front is much weaker that in model 4 which has the same characteristics but for which $\vec{\delta} \perp \vec{U}$: local spreading is controlled by Taylor dispersion at large *Pe*'s and by molecular diffusion at lower ones.

5. Conclusion

The experiments reported here demonstrate that varying the fluid rheology is a powerful diagnostic tool for understanding hydrodynamic tracer dispersion mechanisms in rough fractures.

For models 1 and 2, both the size of the wall rugosities and the correlation length of the velocity field are small compared to the global size of the fracture: this allows one to reach a geometrical dispersion regime at low Pevalues. At higher Pe's, other characteristics of the structure of the void space such as the flow profile in the aperture (model 1) and the distribution of the tracer in the pore junctions (model 2) strongly influence dispersion. In these models, dispersion may be characterized by a single macroscopic dispersion coefficient: however the knowledge of the microscopic structure of the fracture aperture field (correlation length, pore size...) is necessary to predict its value and dependence on Pe.

Experiments performed on multiscale fractures (models 3 and 4) reproducing the roughness of natural fractures have demonstrated the strong influence of channelization and of its orientation with respect to the mean flow on the transport of tracer. An important issue is whether, in these cases, transverse exchange of tracer is large enough so that a diffusive spreading regime might be reached at very large distances. These results have potentially a strong relevance to the efficiency of the recovery of heat through water circulation in geothermal reservoirs. There are also other possible applications to the prediction of seismic events from water circulation in the rock layers under stress.

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