Transport of Particles in Fractured Chalk: Effect of Flow Velocity, Fracture Orientation and Fracture Aperture

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RESUME

Le transport de particules dans les roches est fortement influencé par la présence de fractures, qui servent de conduits primaires pour les particules solides. L'objectif de ce travail est d'étudier l'effet de la vitesse d'écoulement, de l'ouverture et de l'orientation des fractures sur le transport des particules de kaolinite dans la craie fracturée. Des expériences de transport sont réalisées à des vitesses d'écoulement élevées, en utilisant deux échantillons de craie fracturée avec deux ouvertures différentes. Trois orientations différentes ont été utilisées. Les données expérimentales couplées au modèle numérique développé, basé sur l'équation d'advection-dispersion avec un terme source, sont utilisées pour déterminer les paramètres de transport. Les résultats suggèrent que l'effet hydrodynamique est significatif sur le transport des particules dans les fractures. Le taux de restitution, les coefficients d'attachement et de détachement des particules de kaolinite augmentent avec l'augmentation de la vitesse d'écoulement. L'échantillon avec la plus grande ouverture de fracture présente un taux de restitution plus élevé en raison d'un coefficient d'attachement plus faible. L'orientation verticale de la fracture améliore la récupération des particules et conduit à des courbes de percée plus précoces. Les résultats de cette étude contribuent à une compréhension globale des mécanismes de transport des particules dans les fractures.

Mots-clefs Transport de particules en suspension, ouverture de fracture, effet hydrodynamique, effets de la gravité.

I. INTRODUCTION

Emerging problems of pollution and water quality deterioration have progressively imposed the need to master the hydrogeological functioning of pollutant-receiving soils. In order to define the vulnerability of aquifers and to optimize modelling of pollutants transport and retention in soil. The karst aquifers in the Haute-Normandy serve as the main freshwater resources (El Janyani et al. 2014; Massei et al. 2003). The quality of water within these aquifers typically, relies on the existence of high permeability fractures acting as conduits for potential contaminants in the form of solid particles and solutes (Mondal et al. 2012). The introduction of these particles into the groundwater results in its contamination and thus, compromising its quality. This in turn, have a crucial effect on the public health, particularly in regions where potable water comes from karst aquifers.

In geological formation with low matrix permeability, transport of particles occurs predominantly through fractures (Zhang et al. 2012; Zvikelsky et al. 2008; James et al. 2004). Particle transport through fractures is a complex process that includes many mechanisms: advection, diffusion, deposition and re-entrainment (Tang et al. 1981). Fractures play a key role for the transport of pollutants, as they constitute preferential pathways through which solid particles (colloids, bacteria, viruses, etc.) and dissolved substances can move rapidly (Albarran et al. 2013; Mondal et al. 2012). Factors affecting the transport mechanisms of particles in fractures are intricately numerous. These factors include solution chemistry (McCarthy et al. 2002), fracture characteristics (James et al. 2005; Baiyu et al. 2021; Ding et al. 2021a; b), fluid properties (Hawi et al. 2023) and particle properties (Albarran et al. 2013; Zvikelsky 2006). Therefore, it is crucial to understand the impact of these factors on the transport of particles in fractures.

Many studies on different scales have explored the effect of flow velocity on the transport processes of particles in fractures (Mondal et al. 2012; Albarran et al. 2013). However, most of these studies are limited to low flow velocities (less than 0.01 cm/s) and colloidal particles with diameter less than 1 μm. Yet, the narrow apertures of fractures can still yield in high flow velocities. Moreover, high groundwater flow velocities (up to 0.28 cm/s) in fractured carbonate aquifers were reported by Medici et al. (2019). Thus, considering high flow velocities in studying the transport of particles in fractured rocks is crucial.

In reality, fractures are characterized by complex and irregular apertures, and tend to be randomly oriented according to the formation stress and rock properties. While numerous studies focused on the impact of fracture roughness (Baiyu et al. 2021), and aperture distribution (Chrysikopoulos et al. 2003), the effect of fracture aperture remains largely unexplored, despite its significant impact. Furthermore, the effect of gravitational settling, specifically for particles larger than 1 μm, is another crucial factor that cannot be neglected. Several studies employed numerical methods to study the effect of fracture orientation on particle transport in fractures (James et al. 2005; Ding et al. 2021a; b). However, as far as we are aware, no experimental studies have been conducted to explore this effect.

The present study investigated the effect of flow velocity at 0.017 cm/s and 0.085 cm/s. Also, the effect of fracture orientation is explored using three different orientation angles (0°, 45° and 90°) under the flow velocity 0.017 cm/s. Moreover, the effect of fracture aperture is studied using two fractured chalk samples of different apertures (0.81 mm and 1.41 mm). Transport tests in the chalk fractures are performed using kaolinite particles. The experimental data are fitted to the developed numerical model based on the Advection-Dispersion equation with a sources term. Finally, the transport parameters are determined and correlated to the flow velocity, orientation angle and fracture aperture.

II. MATERIALS AND METHODS

A. Experimental setup and procedure

Chalk samples were collected from Saint-Jouin Bruneval on the Normandy coast (France). Two fractured chalk samples were formed (Hawi et al. 2023) of apertures b_1=0.81 mm (±0.08 mm) and b_2=1.41 mm (±0.20 mm). The density and porosity of the chalk utilized is 1.7 g/cm^3 and 0.33 (±0.03) respectively. The geometrical characteristics of the chalk samples, presented in figure 1a, were determined using X-ray computed tomography (RX solutions, France). The suspended particles utilized in this study are kaolinite

P300 of density 2.65 g/cm³. Kaolinite particles were selected using sedimentation and centrifugation to obtain an average particle size of 2.3 µm (0.16 µm <diameter< 10 µm). The diffusion coefficient of fluorescein through chalk was measured using through-diffusion method. It was found to be 0.55×10^{-6} cm²/s, after one year of measurements.

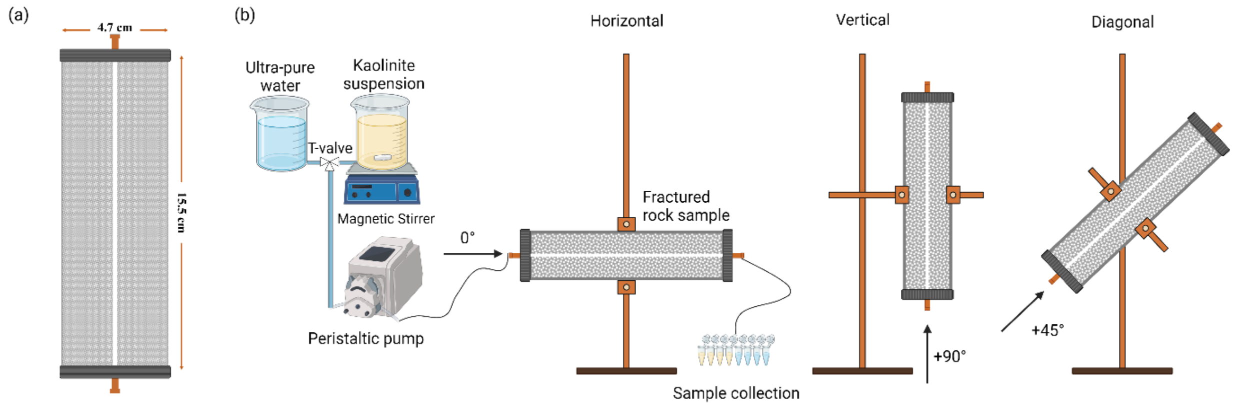

FIGURE 1. Schematic representation of the (a) fractured chalk sample with the geometrical characteristics shown and (b) the experimental setup with fracture oriented horizontally (0°), and vertically (90°), diagonally (45°).

In figure 1b, the schematic representation of the experimental setup is shown. The beaker containing the suspended kaolinite solution is placed on a magnetic stirrer and connected to a peristaltic pump (Dinko instruments, Model D-25plus). Before each experiment, the fractured chalk sample is flushed using 1 mMol NaOH to remove any deposited particles. The chalk samples are saturated using ultra-pure water (Milli-Q Gradient System, 18 MΩ cm) for 72 hours before experimentation. All experiments were performed at room temperature (20°C). Fracture volume (FV) represents the volume of the fracture using the average fracture aperture. The experiment begins with the injection of 3 FV of kaolinite solution of initial concentration C_0 = 0.2 g/l. Then, around 7 FV of ultra-pure water only are injected. At the outlet of the fractured chalk, samples were collected, and their concentrations were determined using a spectrophotometer (UV-1600PC Spectrophotometer) calibrated (R2 > 0.99) with respect to kaolinite.
The obtained concentrations of kaolinite particles (C) were normalized to the initial concentration (C/C_0). The breakthrough curves (BTCs) representing the normalized concentrations of the recovered particles as a function of number of fracture volume (NFV) were plotted. NFV is calculated according to the following equation:

$$NFV = t\frac{Q}{FV} \quad (1)$$

where t is the time [T], Q is the volumetric flow rate [L³/T]. The recovery rate R (%), presenting the percentage of recovered mass of particles relative to the total mass of particles injected, is obtained by integrating the BTC, according to Eq. (2). The relative residence time is calculated according to the moment method (Fried et al. 1971). Finally, the arrival time is the time at which the concentration of particles first becomes to increase, indicating the arrival of particles at the outlet of the fracture.

$$R = \int_0^t C(t)\,dt = \sum_{i=1}^{n-1} \frac{C_{i+1}+C_i}{2}.(t_{i+1} - t_i) \quad (2)$$

Using the peristaltic pump, it was possible to vary the flow velocity. The effect of flow velocity is studied across a range of six different flow velocities between 0.017 cm/s and 0.3 cm/s. In this paper we focus on the outcomes obtained at the two flow velocities 0.017 cm/s and 0.085 cm/s. Moreover, the fracture orientation is varied, using a bracket, between 0°, 45° and 90°, employing upward and downward injection under various flow velocities. Reynolds number has maximum value of 2.32 (Table 1) for the highest tested flow velocities, indicating laminar flow.

B. Modelling approach of transport in fractures

The main transport mechanisms of particles in fractures are advection and dispersion (Tang et al. 1981). In this study the diffusion into the matrix is neglected due to the large particle size compared to the pore size and the low diffusion coefficient of fluorescein (section II.A.). The transport parameters were determined by fitting the experimental data to the one-dimensional advection-dispersion equation with a source term (Equation 2) (Corapcioglu and Jiang, 1993).

$$\frac{\partial C}{\partial t} - D_L \frac{\partial^2 C}{\partial x^2} + U_T \frac{\partial C}{\partial x} = -\rho_p \frac{\partial s}{\partial t} \quad (3)$$

where D_L is the hydrodynamic longitudinal dispersion coefficient [L^2/T], x is the coordinate direction along the fracture [L], U_T is the total particle velocity [L/T], ρ_p is the particle density [M/L^3], s is the mass fraction of the deposited particles on the fracture [M/M]. The source term is expressed as following:

$$\rho_p \frac{\partial s}{\partial t} = k_{att} e^{-\frac{s}{s_c}} C - \rho_p k_{det} s \quad (4)$$

where, k_{att} is the attachment coefficient [1/T], s_c quantifies the decline in attachment coefficient [M/M], and k_{det} is the detachment coefficient [1/T]. To study the influence of fracture orientation, the gravitational effect should be included in the numerical model. Thus, the influence of gravity is integrated in the mathematical model by combining in the total velocity U_T [L/T] (Equation 4), the flow velocity U [L/T] and the sedimentation velocity U_s [L/T] (Equation 5) (Chrysikopoulos et al. 2003).

$$U_T = U - U_s \quad (5)$$

$$U_s = -\frac{(\rho_p - \rho_w) d_p^2}{18\mu_w} g.\sin\beta \quad (6)$$

where ρ_w is the density of water [M/L^3], d_p is the particle diameter [L], μ_w is the hydrodynamic viscosity coefficient [M/L.T], g is the gravitational acceleration [L/T^2], and β is the angle of the flow direction [°], it varies between 0, 45°, and 90°. The initial and boundary conditions are as following:

$$C(x,0) = 0; \qquad s(x,0) = 0; \quad (7)$$

$$C|_{(x=0,t)} = \begin{cases} C_0, & 0 \le t \le t_{inj} \\ 0, & t > t_{inj} \end{cases}, \quad \text{where } t_{inj} \text{ is the injection time} \quad (8)$$

$$\left.\frac{\partial C}{\partial t}\right|_{(x=L,t)} = 0 \qquad (9)$$

III. RESULTS AND DISCUSSION

Experimental analysis coupled with numerical modelling is used to obtain the dispersion, attachment and detachment coefficients controlling the transport of kaolinite particles. The obtained transport parameters, with the experimental recovery rate are presented in Table 1, for the different experiments conditions.

Table 1. **Summary of the kaolinite transport experiments conditions, and the fitted parameters values, and estimated mass recoveries.**

b (mm)	U (cm/s)	β (°)	D_L cm²/s	k_{att} (1/h)	k_{det} (1/h)	Recovery Rate (%)	Reynolds Number
1.41 mm	0.085	0°	0.0550	8.70	0.100	63.9	2.32
	0.017		0.0075	2.70	0.010	43.6	0.46
0.81 mm	0.085		0.0900	11.88	0.200	57.9	1.33
	0.017		0.0080	3.72	0.015	40.9	0.27
		45°	0.0140	3.18	0.002	43.7	
		90°	0.0250	1.50	0.001	71.5	

A. *Effect of flow velocity and fracture aperture*

Figure 2a compares the fitted breakthrough curves of the transport experiments at the two tested flow velocities 0.085 cm/s, and 0.17 cm/s. The results indicate that hydrodynamic effect is significant. When the flow velocity is increased from 0.017 cm/s to 0.085 cm/s, the plateau of the breakthrough curve increased from about 47% to 70%. Similar results were observed in previous studies using flow velocities less than 0.01 cm/s. Albarran et al. (2013) studies the effect of flow velocity for four different types of colloids under flow velocities ranging between 8.5×10^{-4} cm/s and 9.95×10^{-2} cm/s. They found that the peak height of the breakthrough curves decreases with the decrease in flow velocity, showing that particles have been retained in the fracture aperture. Decreasing flow velocity slows down the particle's lotion which in turn leads to longer transport duration. This contributs to particle deposition and inhibits particle transport.

According to Table 1. the dispersion coefficient increases with the increase in flow velocity in both fractured chalk samples. The increase in flow velocity can create some irregularities in the flow, even for laminar flow, leading to the spreading of particles. Therefore, increasing the flow velocity leads to higher dispersion coefficient. This correlation was also shown by several studies before (Keller et al. 1999; Dronfield et al. 1993). Similarly, the attachment coefficient increased when the flow velocity was increased in both fractured samples. Because of the greater spreading of particles at higher flow velocities, the likelihood of particle approaching the fracture surface becomes higher. Eventually, enhancing the probability of attachment to the fracture surface. Moreover, the detachment coefficient also increased with flow velocity due to the increase in hydrodynamic shear force.

Figure 2b. represents the breakthrough curves of kaolinite particles in the two different fractured chalk samples of apertures 1.41 mm and 0.81 mm at the highest flow velocity 0.085 cm/s. The BTCs and the recovery rates (Table 1.) show that the retention and the attachment coefficient (Table 1.) in the fractured chalk sample of the smaller aperture (0.81 mm) is higher. Due to the shorter distance the particles have to travel to reach the fracture surface, the retention of particles in the smaller aperture might occur more easily. Therefore, the recovery of kaolinite particles is higher in the fractured sample with the larger aperture. However, the detachment coefficient in the smaller fractured chalk sample, generally, is higher. Shear stress is inversely proportional to the fracture aperture and increasing shear stress would increase the detachment of particles. Therefore, the detachment coefficient in higher in the smaller fractured sample.

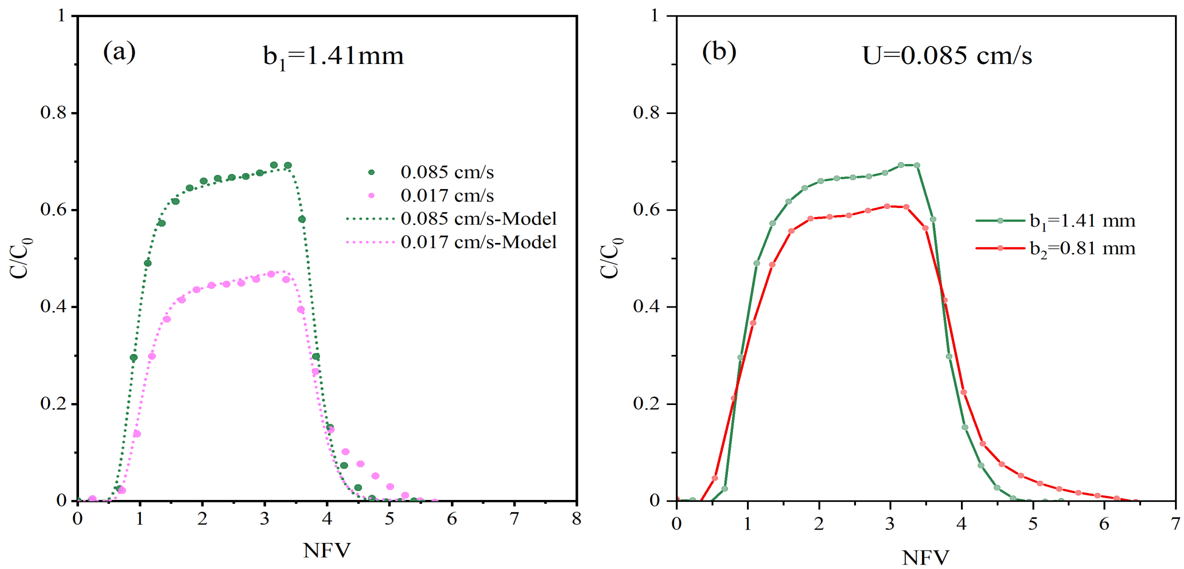

FIGURE 2. Experimental data and fitted model of the breakthrough curves of kaolinite particles in the fractured chalk sample, placed horizontally (0°). (a) Hydrodynamic effect. (b) Fracture aperture effect.

B. Effect of fracture orientation

Figure 3. shows the variation of the breakthrough curves of kaolinite particles with different fracture orientations at the flow velocity 0.017 cm/s. The breakthrough curves become more pronounced with increasing the fracture orientation vertically. When the fracture was placed in the horizontal orientation (0°), the plateau of the breakthrough curve reached around 41%. Then when the fracture orientation was increased to 90°, or in other words placed vertically, the plateau increases to reach around 76%. Similarly, the recovery rate of kaolinite particles increased from 41% at 0° to 71% at 90°. No significant difference was noticed between the horizontal and the inclined orientation. This phenomenon was observed previously in many theoretical studies (Ding et al. 2021a; b; James et al. 2005). Ding et al. (2021a, 2021b) reported that in vertical orientation, the settlement of particles diminishes. So, particles are not dragged away from the main streamline towards the fracture surface. However, in the horizontal orientation, particles can deposit in the

fracture by settlement due to gravity, specifically with low flow velocities. This explains the higher recovery rates at the vertical orientation angles.

Spreading of particles increases with the increase in the fracture orientation. At the horizontal orientation, particles can sink with gravity to the fracture surface. However, at inclined and vertical orientations, particles are able to sample the parabolic flow velocity profile (James et al. 2005). Thus, the spreading or dispersion is higher when the fracture orientation is larger with respect to the horizontal. On the other hand, the attachment coefficient exhibits a contrary trend to the dispersion coefficient with the variation in fracture orientation. The attachment coefficient is maximum (3.96 h^{-1}) when the fracture is at horizontal direction (0°) and it decreases to a minimum value (1.56 h^{-1}) at the vertical orientation (90°). This is consistent with the numerical findings of Ding et al. (2021b). As mentioned before (Section II.A), with steeper angles the collision frequency of particles with the fracture surfaces is lower. Thus, with increasing fracture orientation vertically, the attachment coefficient decreases. This can also explain the higher recovery of particles at vertical and inclined orientations. The detachment coefficient was nil at all fracture orientations. The nearly negligible hydrodynamic shear at low flow velocities, explains the absence of detachment of particles at 0.0017 cm/s.

The breakthrough curves of Figure 3. show different arrival times for particles. In the vertical orientation, particles are transported rapidly, and their arrival tiles is about 0.1 NFV. While for the horizontal orientation, particles are mostly retarded with respect to the vertical and diagonal orientations, they arrive to the fracture outlet after about 0.8 NFV. This can be attributed to the mitigation in the interactions between kaolinite particles and fracture surface at the vertical and diagonal orientations. So, the effect of gravity on particle retardation cannot be neglected.

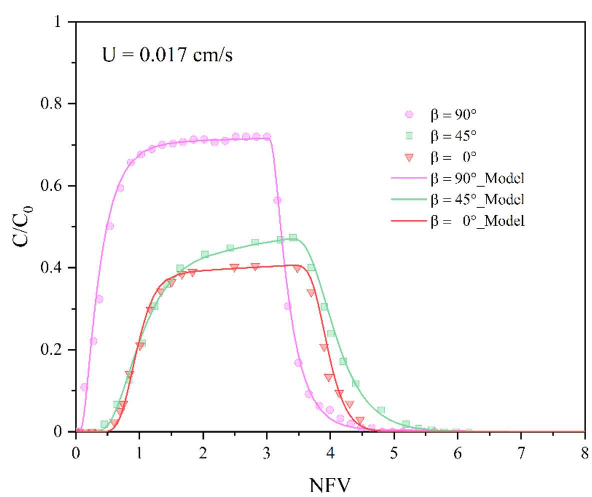

FIGURE 3. Fitted breakthrough curves of kaolinite particles with different fracture orientation (0°, 45°, and 90°) under the flow velocity 0.017cm/s.

IV. CONCLUSION

The effect of flow velocity, fracture aperture and fracture orientation on the transport of kaolinite particles was studied. Transport experiments were performed at two different flow velocities (0.017 and 0.085 cm/s), using two fracture chalk samples with different apertures (0.81 mm and 1.41 mm). The fracture orientation was also varied between 0° (horizontal), 45° (diagonal), and 90° (vertical). Experimental data were fitted to a numerical model based on the Advection-Dispersion equation with a source term. Then, transport parameters were determined and they were correlated with the flow velocity, fracture aperture and fracture orientation. Results indicated that the recovery of particles and the attachment coefficient increases with the increase in flow velocity. Moreover, fracture samples with larger apertures exhibit higher recovery rates with smaller attachment coefficients. The increase in fracture inclination from horizontal to vertical orientation, enhances the transport of particles. This research introduces a novel perspective, that deeply enhances our understanding in transport mechanisms of particles in fractures. Despite, the various studies performed on fractured rocks, to the best of our knowledge, the effect of fracture orientation and flow velocity under high flow rates is rarely explored. Also, no experimental investigation has been reported on particles transport with variable fracture orientation. Such an understanding is important for the assessment of groundwater quality, management of resources and contribute to the advancement in hydrogeology and environmental field.

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