Invasion flow enhanced solute mixing at rough-walled rock fracture intersections

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Key points:
- Invasion flow phenomenon at a 3D rough-walled rock fracture intersection;
- Impact of invasion flow on solute mixing at the fracture intersection;
- Dependence of mixing ratio on Péclet number (Pe) and flow directionality.

Abstract: The processes of fluid flow and solute transport through rock fractures are of primary importance in environmental engineering and geosciences. This study presented numerical modeling results of fluid flow and solute transport in a 3D rock fracture-matrix system with an orthogonal intersection of two rough-walled rock fractures. The rough-walled fracture geometry models were built from laser-scanned data of a real rock surface, for a realistic representation of natural rock fracture surface roughness. The fluid flow in the two intersected fractures and solute transport in the fracture-matrix system were simulated by solving the Navier-Stokes equations (NSE) and transport equation in the entire system. The dependence of mixing on Péclet number (Pe), flow directionality and interaction with matrix diffusion were analyzed. The results showed important invasion flow patterns that significantly enhanced the solute mixing process, which cannot be described by traditional complete mixing and streamline routing models. It also cannot be simulated by simplified 2D geometry models ignoring the surface roughness as widely used in previous published studies. The finding of invasion flow and associated impacts on mixing in this study is particularly important in prediction of solute transport in natural fractured rocks, especially when discrete fracture network (DFN) approach is applied.

Keywords: Rough-walled rock fracture intersection; Navier-Stokes equations; invasion flow; mixing ratio; Péclet number.

1 Introduction

Modeling of flow and transport in intersected rock fractures plays an important role in understanding of fluid flow and solute/contaminant transport processes in fractured rocks. In particular, the fracture intersections allow different fluid and solute to mix along the flow
paths, which significantly affect the solute transport processes in fractured rocks (Johnson et al. 2006).

To deal with solute mixing at fracture intersections, two traditional approaches, complete mixing and streamline routing, have been widely adopted for modelling the diffusion-controlled and advection-controlled transport processes, respectively (e.g., Wilson and Witherspoon, 1976; Robinson and Gale, 1990). These two idealized schematic models for the solute redistribution at a continuous intersection are illustrated in Figure 1 (Hull and Koslow, 1986; Park and Lee, 1999).

The complete mixing and streamline routing models provided simple treatments for concentration redistribution at the intersections, which have been extensively used for modeling solute transport in fractured rocks, especially in DFNs models (e.g., Bodin et al., 2007; Zhao et al., 2011; Frampton and Cvetkovic, 2011; Cvetkovic and Frampton, 2012; Zhao et al., 2013).

Meanwhile, many analytical, experimental and numerical studies have shown that the redistributions of solute at fracture intersections are based on the mixing ratio that depends on the Péclet number (Pe) of solute transport processes (Philip, 1988; Park and Lee, 1999; Mourzenko et al. 2002; Park et al. 2003; Hu et al. 2005; Hellou and Bach, 2011; Johnson and Brown, 2001; Gruber 2001; Mourzenko et al. 2002; Li 2002; Johnson et al. 2006; Zafarani and Detwiler, 2013). In consideration of irregular geometry conditions of natural fracture intersections, various numerical models using lattice gas method (e.g., Stockman et al. 1997), lattice Boltzmann method (e.g., Gruber 2001; Stockman et al. 2001), finite element methods

Fig. 1 Solute redistribute at continuous intersection: (a) complete mixing model and (b) streamline routing model. The symbol C denotes solute concentration, and Q denotes flowrate. The curved arrow indicates appearance of solute mixing.
(e.g., Kosakowski and Berkowitz, 1999; Li et al., 2016) and smoothed particle hydrodynamics (e.g., Zou and Jing, 2013) were developed to model fluid flow and solute transport at the fracture intersections.

These previous studies generally showed Pe-dependent feature of the mixing ratio and the application range of the two models (i.e., complete mixing and streamline routing models) for solute mixing at fracture intersections. However, most previous studies were based on the simplified 2D intersection models formed by two smooth parallel plates, so that the effects of rock fracture surface roughness, as well as associated 3D effects, on flow and transport were ignored. Such geometrical simplification is an inadequate representation of natural rock fractures and intersections, leading to unknown uncertainties in the modelings and predictions. As reported in Johnson and Brown (2001) and Johnson et al. (2006), flow through variable aperture fractures leads to flow channelization, which enhances the solute mixing compared to the 2D intersecting fractures formed by smooth parallel plates. Moreover, most analytical and numerical studies simplified the flow patterns as governed by the Reynolds or Stokes equations, based on assumptions of local or general validity of the cubic law developed for smooth parallel plate models of fluid flow in fractures. Therefore, the simulated flow fields may contain uncertainties, especially for natural 3D rough-walled fractures (e.g., Kosakowski and Berkowitz 1999; Zou et al. 2016a). Last but not the least, almost all of above studies did not considered the matrix diffusion process in models of intersecting rock fractures. In reality, however, the matrix diffusion plays a significant role in solute retardation in fractured rocks (e.g., Neretnieks, 1980; Zou et al., 2016b). Therefore the interaction between matrix diffusion and mixing at fracture intersections is still an open issue.

The objective of this study is to investigate the complex fluid flow and solute mixing behaviors at 3D model of fracture-matrix system with two intersected rough-walled rock fractures, with the process of matrix diffusion included. The two fractures were orthogonally intersected in a porous rock matrix, such that interactions between matrix diffusion and advection and diffusion in fractures can be properly simulated. The Navier-Stokes equation (NSE) was solved for the fluid flow in the fractures, and the advection-diffusion equation was solved for the solute transport in the entire fracture-matrix system. The fluid invasion flow patterns at the 3D intersection were demonstrated and its impacts on solute mixing process were analyzed by comparing with previously published studies. The dependences of mixing ratio on Pe and flow directionality at the intersection are also discussed. Such detailed numerical modelling has not been systematically performed in an integrated fracture-matrix
system model with realistic 3D fracture surface roughness by solving the NSE, and the results will highlight important issues for modeling of flow and transport in fractured porous rocks.

2 Fracture intersection model and physical considerations

In this study, we consider two orthogonally intersected rough-walled fractures located in a fully saturated porous crystalline rock matrix of very low permeability, such as granite, as shown in Fig. 2. The size of the fracture-matrix model is 20 mm in length (Y-axis), 10 mm in width (X-axis) and 20 mm in height (Z-axis). The two intersecting rough-walled fractures in the middle of the model (in blue color) were created by using a laser-scanned natural granite rock fracture surface (Koyama et al., 2008; Zou et al., 2015).

To generate the natural fracture model with variable apertures and asperity contacts, the two fracture models were built by a numerical ‘uplift’ step that simply extrudes up the laser-scanned surface, and then a ‘shear’ step that shifts the upper surface along the length direction with a shear displacement of 1.0 mm (Zou et al. 2016a). The fracture models created in this way were not from direct measurements in laboratory, but they provided a realistic representation of surface roughness in natural rock fractures with contacting asperities and complex spaces of variable apertures, for a generic study. The two fractures are intersecting at

![Fig. 2 Geometry model of the 3D rough-walled fracture intersection with rock matrix. The red numbers are indexing the four fracture branches of the intersection.](image)
a right angle with four branches, which are indexed by red numbers in Fig. 2. The mean apertures of the four branches are the same (0.63 mm). Note that the contacts caused by the numerical shearing play important roles for flow velocity fields in fractures, and the invasion and mixing processes, which is of importance for mechanical loading conditions and deformation processes.

We consider fluid flow in the fractures and solute transport through the whole fracture-matrix system. For simplicity, we limit our consideration to single phase fluid flow through the intersecting fractures as a ‘continuous’ flow type of intersections (Kupper et al. 1995). Specifically, the boundaries of fracture branches 1 and 2 were specified as inflow branches and the boundaries of fracture branches 3 and 4 were set as outflow branches. The source of solute with a constant concentration ($C_0$) was specified at the inlet boundary of fracture branch 1. Mechanical processes were not considered but its effects were represented by the asperity contacts (Zou et al., 2016).

The main solute transport phenomena in natural rock fracture-matrix systems included solute advection and dispersion/diffusion in the fracture, matrix diffusion from the fracture into the matrix, fracture surface and matrix sorption, decay and chemical reactions (Bodin et al., 2003). For simplicity and highlighting the effects of fracture surface roughness and matrix diffusion, we limited our focus on the following key transport mechanisms: 1) solute advection and diffusion in the fractures; and 2) solute diffusion from fractures into the rock matrix. The matrix was assumed to be fully saturated with zero flow velocity.

3 Numerical simulation of flow and transport

3.1 Governing equations and numerical method

The solute transport processes considered in this study through the fracture intersection model with the rock matrix are governed by the general transport equation without source terms, written as

$$\frac{\partial \theta C}{\partial t} + \mathbf{u} \cdot \nabla C - \nabla \cdot \mathbf{D} \nabla (\nabla C) = 0$$

(1)

where $C \ (kg/m^3), t (s), \mathbf{u} \ (m/s), \mathbf{D} \ (m^2/s)$ and $\theta$ denote the volumetric solute concentration in fluid phase, the time, the fluid velocity, the dispersion coefficient and the rock matrix porosity, respectively. For the fracture, with accurate velocity field by solving NSE, its dispersion coefficient $D$ is the molecular diffusion coefficient $D^*$ of solute in the fracture fluid i.e., $D = D^*$. For the porous rock matrix, the diffusion coefficient is equal to the effective diffusion coefficient, and it was defined as related to the matrix tortuosity ($\tau$), written as $D = \tau D^*$ (Tang et al. 1981).
For the isothermal, steady-state and incompressible single Newtonian fluid flow in the fracture, the governing equations are the NSE, which represent mass and momentum conservation respectively, written as

\[
\nabla \cdot \mathbf{u} = 0 \quad (2)
\]

\[
\rho \mathbf{u} \cdot \nabla \mathbf{u} - \mu \nabla^2 \mathbf{u} = -\nabla P \quad (3)
\]

where \( \rho \) (kg/m\(^3\)), \( \mathbf{u} \) (m/s), \( P \) (Pa), \( \mu \) (Pa \cdot s) and \( t \) (s) denote the density of a fluid, the velocity vector, the pressure, the viscosity coefficient and time, respectively.

In this study, the commercial finite element software of COMSOL Multiphysics 5.1 was employed to sequentially solve the NSE and transport equations (COMSOL, 2016).

3.2 Initial and boundary conditions

The inlet flow boundaries of the two inflow branches were set the same as constant flowrate \( Q_1 = Q_2 = Q \). The outlet flow boundaries were set as pressure free (\( P = 0 \)). The rest of fracture surfaces were set as sealed non-slip walls (\( \mathbf{u} = 0 \)). Initially, no solute was assumed in the entire fracture-matrix system (\( C = 0 \) at \( t = 0 \)). At the inlet boundary of the inflow fracture branch 1, the transport boundary condition was set as constant concentration (\( C = C_0 = 1.0 \) kg/m\(^3\)). All the other boundaries are set as gradient free of concentration (\( \mathbf{n} \cdot \nabla C = 0 \), where \( \mathbf{n} \) is normal vector of boundary).

For convenient comparison of results, the concentration was normalized as \( C' = C/C_0 \). Unless specified otherwise, the results reported in the following sections used the normalized time as \( t' = t/t_0 \) for comparison, where \( t_0 \) is the mean residence time for fluid flow through the fracture (\( t_0 = L/\bar{u} \), \( L \) is the length of fracture, \( \bar{u} \) is the averaged flow velocity in the fracture in the principal direction of each branch). The molecular diffusion coefficient was set as \( D^* = 2.03 \times 10^{-9} \) m\(^2\)/s. The matrix porosity \( \theta \) and tortuosity \( \tau \) were set as 0.01 and 0.1, respectively (Tang et al., 1981).

In order to illustrate the sensitivity of mixing on \( \text{Pe} \), a series of simulations with different values of \( \text{Pe} \) in the range of 0.1 to 500 were performed, by changing the flowrate \( Q \) in fractures from \( 2.03 \times 10^{-12} \) m\(^3\)/s to \( 1.015 \times 10^{-8} \) m\(^3\)/s. This range of \( \text{Pe} \) numbers was chosen because the mixing ratios often transit typically in this range in rock fractures as reported in previous studies (e.g., Johnson et al., 2006; Zafarani and Detwiler, 2013). The \( \text{Pe} \) number was defined as in Mourzenko et al., (2002) and Zafarani and Detwiler (2013, written as

\[
\text{Pe} = \frac{\bar{u} \varepsilon}{W D^*} = \frac{Q}{W D^*} \quad (4)
\]
where $U_\text{m/s}$ is the characteristic velocity at the intersection and is taken as the mean value of the normal velocity in the two fractures; $\bar{e}$ (m) is the average aperture of two fractures; $W$ (m) is the width of fracture, $Q$ (m$^3$/s) is the volumetric flowrate and $D^*$ (m$^2$/s) is the molecular diffusion coefficient. The density and viscosity of water were taken as $\rho = 0.9997 \times 10^3$ kg/m$^3$ and $\mu = 1.307 \times 10^{-3}$ Pa·s (water at 10°C), respectively.

Note that Pe may also be defined as $\text{Pe} = \frac{U r}{w D^*}$, where $U$ is the average velocity at the intersection, $r$ is the radius of the fracture intersection (Berkowitz et al., 1994; Stockman et al., 1997; Park and Lee, 1999; Johnson et al., 2006). In cases of laminar flow in smooth fractures of constant apertures and orthogonal intersection models, above two definitions are the same. However, in the rough-walled fracture models with complex surface roughness and varying apertures, it is difficult to determine the radius of the fracture intersection, since the local geometry of the intersection is often irregular.

The established fracture-matrix system model was discretized into around 3.8 million tetrahedral elements. Additional finer elements (around 0.05 mm) were set for the fracture intersection part to properly represent the complex geometry of surface roughness. The resolution of meshes was determined through a mesh-size sensitivity analysis procedure, to reach mesh-independency and numerical stability of the simulation results.

4 Results

4.1 Invasion flows at the 3D fracture intersection

Figure 3 presents flow streamlines in the fractures for $Q = 2.03 \times 10^{-12}$ m$^3$/s, Pe = 0.1, as the representative result of flow patterns at the intersection. Due to the same constant flowrate through the two inflow branches ($Q_1 = Q_2 = Q$) and the same mean apertures of the two fractures, the flowrate at the outflow branches are almost equal ($Q_3 \approx Q_4 \approx Q$).

In order to clearly show the flow redistribution at the intersection, the flow in different inflow branches 1 and 2 were differentiated by different colors, i.e. red and black for branches 1 and 2, respectively. Apparently, the continuous flow streamlines from inflow to outflow branches show channeling flow behavior around the contacts areas in the fractures, and significant invasion flow patterns at the intersection. Specifically, a small amount of the red streamlines from the inflow branch 1 were invaded into the outflow branch 3, and a similar amount of the black streamlines from the inflow branch 2 were also invaded into the outflow branch 4. This means that the flow itself is mixing at the 3D rough-walled intersection because of the irregular fracture surface roughness and irregular geometry shapes at the intersection. This invasion flow cannot be obtained when smoothed parallel fracture
models were used. It also cannot be simulated by using 2D intersection models, since the mixing invasion flow occurs on the perpendicular direction of the intersection plane which is ignored in 2D models.

4.2 Concentration distributions

In order to illustrate concentration distribution features and the interaction between matrix diffusion and mixing processes at the fracture intersection for different Pe values in the 3D fracture-matrix system model, the distributions of solute concentration fields when $t = 2t_0$, for Pe = 0.1, 1, 10, 100, 200 and 500, are presented in Figure 4.

For Pe = 0.1 (Figure 4a), the solute spread almost homogeneously along the branch 1 to branches 3 and 4 with generally uniform evolution of matrix diffusion process from the fracture into the matrix, since the diffusion plays a dominant role in transport for small values of Pe (i.e., Pe < 1). When the Pe values increase from 1 to 500 (Figure 4b-f), the corresponding concentration fields gradually show much higher concentration in branch 4 than in branch 3, indicating that the mixing at the fracture intersection reduces with increasing Pe. In particular, when Pe is relatively high (i.e., Pe > 10), the concentration fields in fracture
branches 3 and 4 show strong channelization following the invasion flow paths as shown in Figure 3. Therefore, the mixing process at the 3D rough-walled fracture intersection is highly Pe-dependent. The matrix diffusion process may affect the mixing and concentration distributions in the entire fracture-matrix system when Pe is relatively small (i.e., Pe ≤ 1). More importantly, the mixing invasion flows significantly enhance the solute mixing process when Pe is relatively high (i.e., Pe > 1).

4.3 Breakthrough curves of the outflow branches

Figure 5 presents breakthrough curves of the outflow branches for different Pe numbers, which illustrates the mean concentration evolution on the outlets of the outflow branches 3 and 4 as a function of transport time.

The breakthrough curves for all Pe generally reach a stable state around t = 5t₀. Similarly, only when Pe = 0.1 (Figure 5a), the breakthrough curves at the outlet of outflow branches 3 and 4 are almost identical, indicating that the solute was well mixed at the fracture intersection in the low Pe cases (i.e., Pe ≤ 0.1). When Pe increases from 1 to 100 (Figure 5c-d), the concentration gradually decreases on the outflow fracture branch 3 and increases for the branch 4. The increasing difference between the breakthrough curves of outflow branches 3 and 4 is caused by the gradually less extent of mixing at the intersection. However, when Pe
increases from 100 to 500 (Figure 5d-f), the breakthrough curves of outflow branches 3 and 4 are approximately the same, which is similar to the results of concentration distributions shown in Figure 4d-f. This indicates that the transport is fully dominated by advection when Pe > 100.

4.4 Mixing ratio

The mixing ratio ($M_r$), is widely used to describe the mixing behavior at the fracture intersections in the literature (e.g., Stockman et al., 1997; Johnson et al. 2006; Zafarani and Detwiler, 2013), defined as

$$M_r = \frac{C_3}{C_3 + C_4}$$  \hspace{1cm} (5)

where $C_3$ and $C_4$ are the mean concentration at the outlets on branch 3 and 4, respectively. The mixing ratio for different Pe between 0.1 and 500 when $t = 5t_0$ were calculated and presented in Figure 6. For comparison, earlier studies applied the same equal flowrate boundary conditions, as used in this study (Berkowitz et al., 1994; Stockman et al., 1997; Park and Lee, 1999; Li, 2002; Zafarani and Detwiler, 2013); results from these studies are also plotted in Figure 6. These earlier studies were based on the 2D simplified geometry models without considering the effects of fracture surface roughness.
As shown in Figure 6, all the mixing ratio results were varied in the range between 0 and 0.5, as decreasing functions with the increasing Pe from 0.1 to 500. Note that the mixing ratio values of complete mixing model and streamline routing model equal to 0.5 and 0, respectively, which are the respective upper and lower limit values of the mixing ratio at the fracture intersection. Obviously, both the complete mixing model and the streamline routing model leads in over- or underestimation of the mixing ratio, due to lacks of proper description of the Pe-dependent transport process.

More importantly, the results obtained from this study have much higher mixing ratio values than all of abovementioned earlier studies, especially for Pe > 10. In particular, unlike earlier studies that the mixing ratio curves gradually decrease toward an asymptotic value of 0 with the increasing value of Pe. Instead the mixing ratio values from this study decrease toward an asymptotic value of 0.24. Such important differences quantitatively illustrate that the invasion flows due to the 3D fracture surface roughness and shear caused asperity

Fig. 6 Comparison of mixing ratio as functions of Pe.
contacts, significantly enhance the solute mixing process at natural fracture intersections. The enhanced mixing ratio results indicate that fracture surface roughness and the associated complex invasion flow patterns may be important issues to include in modeling of solute transport in fracture networks.

5 Discussion

In this study, our numerical modeling results of flow patterns, concentration distributions and the mixing ratios have demonstrated the important invasion flow behavior and its significant impacts on the mixing process at natural 3D rock fracture intersections. Despite the fact that such mixing invasion flow did not affected the total flowrates at outflow branches, it significantly enhanced the solute mixing process, especially for transport cases with high Pe values. Such enhancement of mixing ratio at 3D rough-walled fracture intersections was also found in earlier experimental study results by Johnspn et al. (2006) that was based on a synthetic fracture intersection model using pieces of textured and flat glass. Comparing with their study, our model showed clearer invasion flow patterns at the natural rock intersection and solute transport behavior in the entire fracture-matrix system model, created by using the laser-scanned surface roughness data from a real rock sample, with shear induced contacts. Such direct and detailed invasion flow patterns and mixing behavior is still unavailable in most of the current experimental observations.

Fig. 7 Slice of concentration distribution at cross-section X = 5 mm mm over a relatively long time ($t = 10 t_0$) for Pe = 1.
Matrix diffusion has been recognized as an important process affecting solute transport in fractured rocks, especially for the long-term diffusion-dominated transport processes in applications, such as modeling of radionuclide transport in the subsurface (e.g., Neretnieks, 1980; Zou et al., 2016b). Many analytical and numerical models have been developed to consider the matrix diffusion for single fracture-matrix systems (e.g., Neretnieks, 1980; Tang et al. 1981). However, the complex fracture intersections in natural fractured rocks significantly increase the difficulties and uncertainties in application of such models, especially for the prediction of concentration distribution in the matrix. The reason is that the matrix diffusion is determined by the transport processes in the fractures. The mixing behaviors at the intersections will consequently affect the diffusion process into the adjacent matrix. In order to illustrate the concentration distribution feature in the matrix, a slice of concentration field at the cross-section \( X = 5 \text{ mm} \) of the intersecting fracture-matrix model over longer time \((t = 10t_0)\) is presented in Figure 7 for \( \text{Pe} = 1 \). It shows the irregular distribution of solute concentration in the matrix around corners of the fracture intersection, due to the interaction between fracture branches. Such irregular distribution of solute concentration may cause important uncertainties in modeling of solute transport in large scale fracture network systems, which calls for proper consideration or upscaling treatment in applications.

Due to the 3D nature of rock fracture surface roughness, the transmissivity and flow fields in rough-walled fractures often show anisotropy, so that the flow behavior is dependent on the flow directionality (e.g., Cardenas et al. 2009). In this study, we also found that the invasion flow patterns and mixing ratios were significantly dependent on the flow directionality. Figure 8 presents an exemplified results of flow and transport in the opposite direction where inflows from the branch 3 and 4, and outflows from the branches 1 and 2 were specified, for \( \text{Pe} = 1.0 \). The flow streamlines and concentration distributions show significant differences in the invasion flow patterns, channeling and mixing behaviors, comparing with the results shown in Figure 3 and 4. The corresponding mixing ratio value in this case with a changed flow direction is 0.21, which is slightly smaller than original case (around 0.24).

In the context of such upscaling analysis as DFN modeling, the invasion flow and enhanced solute mixing may cause additional uncertainties, due to more complicated intersection structures and flow conditions, as well as the large number of fractures in the networks. In order to quantify such uncertainties in DFN modeling, it requires proper characterization of the intersection structures and further stochastic modeling. However, a
A better understanding of flow and mixing at single fracture intersection systems is primarily important.

Fig. 8 Invasion flow and mixing behavior in the opposite direction where inflows from the branch 3 and 4, and outflows from the branches 1 and 2, for Pe = 1. (a) Streamlines show different invasion flow patterns; and (b) concentration distributions show a different mixing ratio value.
In this study, we only considered a simple approximately orthogonal intersection of two fractures, with the ‘continuous’ type of flow. The mean apertures of the two intersecting fractures are approximately the same, and the inflow boundary conditions on the inlet branches are equal. However, in natural fractured rocks, the apertures of each fracture, intersection angle, inflow ratio of the two inflow branches and the flow types vary spatially, depending on the local geological and hydraulic conditions. How these factors affecting the invasion flow patterns and mixing processes are important topics requiring further studies.

6 Conclusions

The mixing invasion flow patterns at 3D rough-walled fracture intersections significantly enhance the solute mixing process at the intersection. The invasion flows are caused by the irregular geometry conditions of the fracture surface roughness. Such enhancement of mixing behavior cannot be properly described by simplified fracture models built on smooth parallel plates. The mixing process at fracture intersections also strongly depends on Pe in the range between 0.1 and 500, which cannot be well described by the conventional full mixing or streamline routing models. The mixing ratio simulated in this study is much larger than that of previous studies based on simplified geometry models as well as the streamline routing models, especially for cases with relatively high Pe (i.e., Pe > 10). The concentration distribution in the fracture-matrix system becomes more uncertain due to interactions between matrix diffusion and solute mixing processes. The mixing behavior is also shown to be dependent on the flow direction at the intersection, because of the dependence of invasion flow patterns on flow directionality. Therefore, it is important to consider the fracture surface roughness, invasion flow patterns, Pe-dependent transport behaviors and flow directionality in modeling of solute transport in fractured rock.

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