

# Reactive solute transport in a filled single fracture-matrix system under unilateral and radial flows



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## ARTICLE INFO

### Article history:

Received 13 October 2016

Revised 23 February 2017

Accepted 29 March 2017

Available online 2 April 2017

### Keywords:

Fracture-matrix system

Reactive solute transport

Injection well

Mobile-immobile model

## ABSTRACT

The study of transport processes in a single fracture is the basis of understanding transport in complex fractured networks. Many single fractures in the field are filled with sediments, and the transport in such filled single fractures has received much less attention up to present. When the fracture is partially filled with sediments, a mobile-immobile approach is considered necessary. This study deals with a coupled three-domain transport problem using mobile and immobile domains to characterize a filled single fracture and a matrix domain to characterize the rock body. Mathematical models are developed for such a coupled three-domain transport problem with new semi-analytical solutions to analyze the spatial-temporal concentration and mass distributions in the fracture and rock matrix with the help of Laplace transforms. This study addresses transport in a filled fracture-matrix system under two different flow conditions: unilateral flow, and radial flow. The new solutions have been tested extensively against previous solutions under various special settings and are proven to be robust and accurate. This study has the following findings: 1) Longitudinal dispersion in the fracture often plays an important role in such a coupled system in unilateral flow, 2) Mass partitions in three domains follow similar patterns in respect to the influence of fracture apertures, mobile/immobile ratios, and first-order mass transfer rates, 3) The system is most sensitive to the dispersivity and least sensitive to the first-order mass transfer rate and the mobile/immobile ratio in the unilateral flow model over a wide range of time scales (if the longitudinal dispersivity and Darcian flow velocity remain constant), 4) The system is most sensitive to the dispersivity, less sensitive to the mobile/immobile ratio, and least sensitive to the first-order mass transfer rate in the radial flow model (if the radial dispersivity and injection rate remain constant).

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## 1. Introduction

Transport in fractured media has been considered as an important research subject by many scholars for more than three decades because of its broad range of applications in different disciplines (Roubinet et al., 2012; Grisak and Pickens, 1981), for instance, for dealing with problems such as the disposal of radioactive materials, CO<sub>2</sub> geological sequestration and storage (Pouya, 2012), and groundwater pollution in fractured reservoirs (Bodin et al., 2003). Since fractures are much more permeable than the surrounding rock matrix (Wilson and Witherspoon, 1970), fractures have the potential for being the most effective pathways for solute migration. For this reason, transport in the rock matrix is often highly simplified in mathematical models, for instance, limited to a diffusion-dominating process (Tang et al., 1981). Specifically,

some experiments have been designed and conducted to confirm that advective transport in the rock matrix can often be ignored but diffusive transport in the rock matrix must be considered in most cases (Roubinet et al., 2012; Maloszewski and Zuber, 1993).

Understanding transport in a single fracture is the foundation of understanding transport in fracture networks (Tang et al., 1981; Long and Billaux, 1987). Analytical solutions developed at the scale of fracture-matrix systems can be applied directly to models of solute transport in fracture networks. This is the case for the particle-tracking methods developed by Dershowitz and Miller (1995) and Cvetkovic et al. (2004) and reviewed by Noetinger et al. (2016). The solutions developed for a single fracture-matrix system can also be extended rather straightforwardly to deal with transport in fractured rocks with multiple parallel fractures which may or may not have the same apertures, as demonstrated in details by Sudicky and Frind (1982), Zhu et al. (2016), and others.

There are multiple reasons to explain why investigators are interested in single fracture transport. First, this is the simplest possible fracture transport case that may be solved using an analytical

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approach, which can offer insights on various transport processes. Such analytical or semi-analytical solutions may serve the purpose of benchmarking numerical solutions developed for transport in a fracture-matrix system, which may suffer from non-negligible (and often hidden) numerical errors, partially because of the sharp differences of fracture and rock matrix parameters (Grisak and Pickens, 1981). One may consult Seo and Mittal (2011) for numerical challenges related to sharp interfaces (such as along a fracture-matrix boundary). Also, a thin fracture, often with an aperture on the order of millimeter or less, requires a very fine grid to discretize the fracture in numerical simulations which may not be practical for dealing with large-scale field transport problems (Weatherill et al., 2008). Secondly, a single fracture offers simple enough setting to test different transport theories related to a fracture-matrix system. Thirdly, isolated single fractures exist in real geological settings (Moreno et al., 1988; Raven et al., 1988). Because of its importance, significant effort has been put into conducting tracer transport experiments in single fractures (Brown et al., 1998; Esposito and Thomson, 1999), in addition to the theoretical works that will be briefly reviewed in the following.

A widely-used analytical solution of contaminant transport in a single fracture system was proposed by Tang et al. (1981). Chen (1986) also derived an approximate solution for radial transport from an injection well into a single fracture. Theoretical studies of transport in a single open fracture have been reported by numerous investigators, including Moreno et al. (1988), and Esposito and Thomson (1999). In the above-mentioned studies, fractures were assumed to be open without infillings.

However, fracture infillings are commonly observed in real applications (Wealthall et al., 2001; Bradner and Murdoch, 2005). Wealthall et al. (2001) conducted an investigation of fractures filled with sediments and compared the preferential flow pathways in such filled fractures to those in an equivalent open (unfilled) fractures. Kemp et al. (2003) evaluated filled fractures in Permo-Triassic sandstones in southwest Scotland and presented a sampling method for such fractures. Bradner and Murdoch (2005) investigated the gas-phase permeability in sand-filled fractures in a soil vapor extraction system.

A filled fracture usually has a different transport behavior from an open one. For instance, Lunati et al. (2003) found that a propagation front was smoother in single fractures filled with glass beads at extremely low flow velocity, compared to those in open fractures. The existence of infilling materials often leads to disconnections or dead-end water pockets between pore spaces within a fracture, which can be regarded as immobile domains (Jodar et al., 2009; Qian et al., 2011). Such a problem cannot be dealt with using the advection-dispersion equation (ADE) which is commonly used for dealing with transport in homogeneous porous media or open fractures, as done by Tang et al. (1981) for unilateral flow, and by Chen (1986) for radial flow. Regarding to transport in an open fracture, there is much evidence showing that ADE performs poorly, and cannot explain the so-called non-Fickian transport phenomena such as the early breakthrough and long tailing of the breakthrough curves (BTCs), that can, however, be satisfactorily explained with mobile-immobile models (MIM) as first proposed by Van Genuchten and Wierenga (1976).

The objective of this study is to develop new mobile-immobile models for two types of transport problems in a filled single fracture-matrix system, which has not been attempted before. The following transport processes are considered: advection, longitudinal dispersion, first-order reaction, and linear sorption in the fracture; transverse molecular diffusion, first-order reaction, and linear sorption in the rock matrix; first-order mass transfer between the mobile and immobile domains in the filled fracture. The first model concerns a unilateral flow field; while the second model concerns a radial flow field caused by an injection/pumping well.

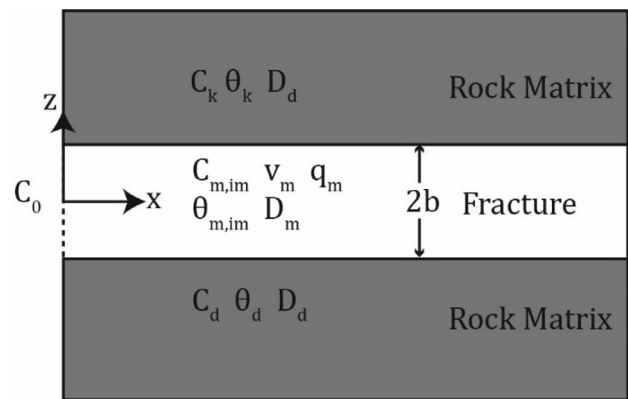


Fig. 1. The conceptual model of unilateral flow.

## 2. The conceptual model and physical setup

In this study, a filled single fracture is oriented horizontally with a constant aperture  $2b$  and extends sufficiently far from the domain of interest. The fracture is bounded by a rock matrix whose permeability is at least several orders of magnitude less than that of the fracture. The rock matrix is wide enough so that the effect of the limit boundary of the rock matrix can be ignored. This is justified, as the primary transport process in the rock matrix is often limited to regions close to the fracture since it is a much slower process.

In this model, a constant-rate unilateral flow field from left to right is established (Fig. 1). A Cartesian coordinate system is used with the origin at the intercept of the left boundary and the middle of the fracture. The  $x$ -axis is along the same direction with the unilateral flow, and the  $z$ -axis is vertically upward. The  $y$ -axis is perpendicular to the  $x$ -axis and is horizontal as well. A solute source is located at  $x=0$  with a constant concentration  $C_0$  and extends to sufficiently far distance from the domain of interest along the  $y$ -axis. Thus, the problem can be conceptualized as a two-dimensional (2D) model in the  $xz$  plane. Such a simple boundary condition at  $x=0$  (constant concentration) can be relaxed later to accommodate more realistic boundary types, such as prescribed time-dependent concentration (first-type), prescribed flux (second-type), or Robin (third-type). Two points are notable. First, the fracture aperture is so small that the vertical mixing throughout the fracture aperture is completed almost instantaneously, thus transverse dispersion in the fracture along the  $z$ -axis has no discernible effect on transport processes in the fracture-matrix system and is not considered. Roubinet et al. (2012) further reported that longitudinal diffusion in the rock matrix affected solute transport only when the Peclet number was very low (less than 0.01). Rezaei et al. (2016) also investigated the horizontal dispersion in the aquitard (HDA) on reactive solute transport in an aquifer-aquitard system, which is similar to the fracture-matrix system investigated here except that slow water flow (advection) in aquitard was considered by Rezaei et al. (2016) and water flow in matrix is excluded in this study. Rezaei et al. (2016) concluded that HDA was negligible for most practical cases of transport. Therefore, longitudinal diffusion in the rock matrix is neglected in both models of this study. Secondly, despite the fact that the fracture is filled, it could still be very porous and very permeable.

The conceptual model of radial flow is the same as that of unilateral flow but with the following exceptions (Fig. 2). A radial flow field rather than a unilateral flow field is established due to an injection/pumping well with a constant rate  $Q$  (positive for injection and negative for pumping). Accordingly, a cylindrical coordinate system rather than a Cartesian coordinate system is

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