



# Gaining insights into reactive fluid–fractured rock systems using the temporal moments of a tracer breakthrough curve



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## ABSTRACT

In this paper, we show that the tracer breakthrough curves (BTCs), when the tracer chemically interacts with the solid matrix of a fractured rock, are considerably different than when it does not. Of particular interest, is the presence of a long pseudo steady state zone in the BTCs, where the tracer concentration is more or less constant over a long period of time. However, such a zone of constant concentration is not visible when either the tracer does not interact with the solid, or does so at an extremely fast rate. We show that these characteristics of the BTCs could be correlated to the parameters of the system. We develop expressions for the mean residence time and its variance for a chemically active and inactive tracer. We show that chemical interaction between the tracer and the solid increases the mean residence time and the increase depends on the distribution coefficient. We also show that the variance of residence time for a chemically active tracer is much larger than that for an inactive tracer, and it depends on both the distribution coefficient and the rate of chemical reaction. We verify these calculations against synthetic tracer BTCs, where the temporal moments are calculated by numerically integrating the tracer evolution curves. Even though we developed the mathematical expressions assuming an idealized fracture–matrix system, we believe that the mathematical expressions developed in this paper can be useful in gaining insights into reactive transport in a real fractured rock system.

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## 1. Introduction

In fractured rocks, diffusive exchange of solutes between the fractures and the rock matrix causes retardation effects on the solutes (Sanchez-Villa and Carrera, 2004). This process, hereafter referred to as matrix diffusion for sake of brevity, has been the focus of considerable research – see, for example Maloszewski and Zuber (1985, 1990, 1993) and Moench (1995); a more thorough review on this topic can be found in Zhou et al. (2007). As discussed in Zhou et al. (2007), many tracer tests have been conducted since the 1970s, and data from these tests have provided significant insights into the retardation effects of matrix diffusion on solute transport in fractured rock systems. Moreover, researchers have often utilized data from these tracer tests along with a relevant flow model to estimate flow and transport parameters (see Zhou et al., 2007 and references therein).

Matrix diffusion is a transport mechanism by which solutes transfer from the water flowing portions of permeable media to the non-flowing portions (matrix) and vice versa. The effects of matrix diffusion are diverse (Sanchez-Villa and Carrera, 2004). On one hand, a large volume of stagnant water becomes accessible to the solute by diffusion. This causes an apparent retardation with respect to solutes that do not enter the matrix (Goltz and Roberts, 1987; Maloszewski and Zuber, 1985). An interesting example is that of Zuber et al. (2001), who presented a case in Poland where matrix diffusion causes a huge retardation (on the order of 50). On the other hand, diffused solutes may take a long time to come out of the matrix, causing a large tailing in the breakthrough curves (BTC).

While the retardation effects of matrix diffusion on tracer transport and subsequently on the tailings in the BTCs have been widely analyzed, the impact of chemical interaction of the tracer (say, through precipitation and/or dissolution) with the solid phase of the matrix on transport and on the

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BTCs have not been investigated to such an extent. When a reactive tracer is instantaneously injected into a fractured rock system, the tracer enters the matrix by matrix diffusion, where it may precipitate depending on tracer solubility (i.e., if the injected tracer concentration exceeds solubility). At this early stage, transport is governed by matrix diffusion processes. Once sufficient precipitation has taken place, the local thermodynamic condition dictates the dissolution of the tracer from the rock matrix into the matrix fluid, wherefrom it then diffuses back into the fracture fluid. This later part of the transport problem is governed by the rates of reaction. Because the time scales involved in such geochemical precipitation and dissolution reactions are considerably longer than those associated with matrix diffusion alone (DePaolo, 2006; Liu et al., 2011; Mukhopadhyay et al., 2013), it is intuitive that the tailings in the BTCs of transport system in fractured rock involving tracer precipitation/dissolution is much larger than those resulting from matrix diffusion alone. It is our hypothesis that these differences in the BTCs (between a reactive system and a non-reactive diffusion controlled system) can be exploited to gain insight about the nature of the transport system. We also suggest that these can be exploited to estimate transport and reaction parameters. In this paper, we establish the mathematical and theoretical foundation of such an approach using an idealized flow system in fractured rocks.

## 2. Conceptual model and semi-analytical solution for transient tracer transport

The details of the conceptual model for transient transport of a reactive tracer in an idealized fracture–matrix system where the tracer can precipitate/dissolve from the matrix pore fluid to the matrix solid phase according to a specified rate law and controlled by an equilibrium distribution coefficient can

be found in Mukhopadhyay et al. (2013). For the sake of convenience, we provide here a summary of the same conceptual model, which is also schematically shown in Fig. 1. In this conceptual model, we consider fractures of length  $L$  in a system of parallel horizontal fractures with spacing of  $2B$  between two adjacent fractures. The fractures have a width of  $2b$ , and they are embedded in a rock matrix that has a porosity of  $\phi_m$ . In the fracture, groundwater flow rate is  $u_f$  and transport can be by advection, molecular diffusion, and longitudinal dispersion. A tracer is injected instantaneously into the fracture fluid, which while being transported along the length of the fracture by advection, may diffuse into the matrix. The water in the matrix is assumed to be stagnant, i.e., tracer may diffuse into or out of the matrix only by diffusion with a matrix diffusion coefficient of  $D_m$ . Once in the matrix, tracer may precipitate onto the rock matrix. Tracer may also dissolve out of the rock matrix into the matrix pore water and diffuse back into the fracture.

When solids dissolve, they contribute the tracer to the matrix pore fluid, and when solids precipitate from the matrix pore water, they incorporate the tracer according to a distribution coefficient,  $K$ . In other words, if the rate of the dissolution reaction is  $R'_d$  [kg-solid  $m^{-3} s^{-1}$ ] and the tracer concentration at the solid phase is  $c_s$  [mol/kg-solid], solutes are added to the fluid phase at the rate of  $R'_d c_s$  [mol  $m^{-3} s^{-1}$ ] through dissolution. On the other hand, assuming the rate of precipitation to be  $R'_p$  [kg-solid  $m^{-3} s^{-1}$ ] and the fluid-phase tracer concentration in the matrix water to be  $c_p$  [mol/kg-water], solutes are removed from the fluid phase at the rate of  $R'_p K c_p$  [mol  $m^{-3} s^{-1}$ ] because of precipitation. In the treatment that follows we assume that  $R'_d = R'_p = R'$ , i.e., the rate of precipitation is assumed to be same as the rate of dissolution (DePaolo, 2006; Mukhopadhyay et al., 2013). This assumption simplifies the mathematical derivation that follows and may be acceptable for systems where the rate

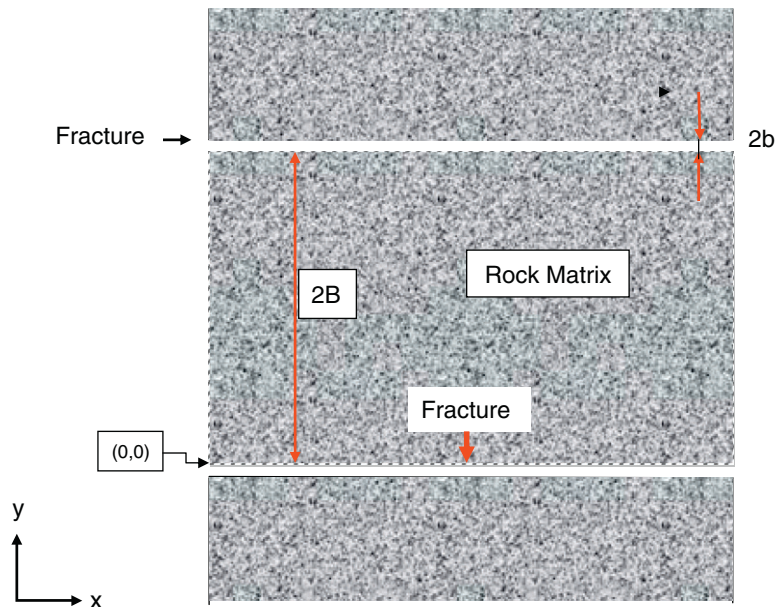


Fig. 1. Schematic representation of the conceptual model.

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