



## Research papers

# Validity of using large-density asymptotics for studying reaction-infiltration instability in fluid-saturated rocks

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## ABSTRACT

Reaction-infiltration instability, in which chemical reactions can dissolve minerals and therefore create preferential pore-fluid flow channels in fluid-saturated rocks, may play an important role in controlling groundwater quality in groundwater hydrology. Although this topic has been studied for many years, there is a recent debate, which says that the use of large-density asymptotics in the previous studies is invalid. However, there is a crucial conceptual mistake in this debate, which leads to results and conclusions that are inconsistent with the fundamental laws of physics. It is well known that in terms of distance, time and velocity, there are only two independent variables. But they are treated as three independent variables, a procedure that is the main source of the physically unrealistic results and conclusions in the debate. In this paper, we will discuss the results and conclusions related to the debate, with emphasis on the issues leading to the corresponding errors. In particular, we demonstrate that there is an unappreciated constraint condition between the dimensional/dimensionless distance, time and velocity in the debate. By using this constraint condition, it can be confirmed that as the ratio of the reactant concentration in the incoming fluid stream to the mineral concentration approaches zero, the dimensionless transport parameter,  $H$ , automatically approaches infinity. Therefore, it is further confirmed that the previous work conducted by Chadam and others remains valid.

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## 1. Introduction

Reaction-infiltration instability, in which chemical reactions can dissolve minerals and therefore create preferential pore-fluid flow channels in fluid-saturated rocks, may play an important role in controlling groundwater quality in groundwater hydrology (Imhoff et al., 1996, 2003; Maji and Sudicky, 2008; Miller et al., 1990; Seyedabbasi et al., 2008). To solve this problem, Chadam et al. (1986) conducted a seminal study by using large-density asymptotics, in which the density ratio,  $\gamma$ , of the reactant concentration in the incoming fluid stream,  $c_{in}$ , to the mineral concentration,  $c_{sol}$ , is approaching zero. In their study, chemical dissolution fronts were considered to propagate in the infinite space, which is filled with fluid-saturated rocks. The following two main conclusions were drawn from their study: (1) as the density ratio ( $\gamma$ ) approaches zero, the thickness of a chemical dissolution front also approaches zero, leading to a sharp shape of the chemical dissolution front; (2) for a given reaction-infiltration system with a given inflow velocity in the fluid-saturated rock, a chemical dissolution

front is unstable if the wavelength of an applied small perturbation is greater than the critical wavelength of the system. Due to the theoretical importance of this seminal study, extensive theoretical analysis and computational simulations have been further followed in the past years (Ortoleva et al., 1987a,b; Ormond and Ortoleva, 2000; Chen and Liu, 2002, 2004; Chen et al., 2009; Lai et al., 2014, 2016). In particular, Imhoff and Miller extended the work of Chadam et al. (1986) to the theoretical study of non-aqueous phase liquid (NAPL) dissolution instability in fluid-saturated porous media (Imhoff and Miller, 1996), which plays an important role in removing NAPLs from contaminated groundwater resources (Imhoff et al., 1996, 2003; Maji and Sudicky, 2008; Miller et al., 1990; Seyedabbasi et al., 2008).

However, there is a recent debate (Ladd and Szymczak, 2017) to question the validity of using large-density asymptotics in the work of Chadam et al. (1986). Unfortunately, there are serious mistakes that lead to questionable conclusions in the debate paper. First, the treatment of distance, time and velocity as three independent variables violates the fundamental principle in physics. This violation leads to a constraint condition between the density ratio,  $\gamma$ , and the dimensionless transport parameter,  $H$ . Note that this density ratio,  $\chi$ , is also called the mineral dissolution ratio in

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previous papers (Zhao et al., 2010; Zhao, 2014). Second, the main conclusions in the debate paper are questionable. Actually, we can use the related equations in the debate paper itself to demonstrate that the constraint condition between the mineral dissolution ratio,  $\gamma$ , and the dimensionless transport parameter,  $H$ , clearly requires that as  $\gamma \rightarrow 0$ , the accompanied condition of  $H \rightarrow \infty$  must be automatically satisfied. Thus, this fact not only demonstrates that  $\gamma$  indeed plays a role in determining the degree of reactant penetration in the downstream region, but also demonstrates that in the limit of  $\gamma \rightarrow 0$ , the reactant concentration in the downstream of the front indeed vanishes. In addition, this fact can be further confirmed when a physically-consistent set of dimensional/dimensionless distance, time and velocity are used to derive the dimensionless governing partial-differential equations of the reaction-infiltration instability problem.

In addition, the criterion for a reaction-infiltration instability derived by Zhao and coworkers (Zhao et al., 2007, 2008, 2010, 2013b) is significantly different from that derived by Chadam et al. (1986). In reality, there are two significantly different approaches to the establishment of a criterion for assessing the instability of a dissolution front when it propagates in a chemical dissolution system either with a given inflow velocity (Chadam et al., 1986) or with a variable inflow velocity (Zhao et al., 2007, 2008, 2010, 2013b). Unfortunately, this significant difference was neglected in the debate (Ladd and Szymczak, 2017). In the former case, the main focus is on the *computational simulation aspects of morphological evolution* of chemical dissolution fronts in *supercritical* chemical dissolution systems, while in the later case, the main focus is on the *theoretical analysis aspects of instability* of chemical dissolution fronts in *critical* chemical dissolution systems. In the following sections, we will address the above-mentioned issues in detail. To facilitate the following discussions, the same definitions of the variables as those used in the debate (Ladd and Szymczak, 2017) are employed in this paper.

## 2. The treatment of distance, time and velocity as three independent variables leading to an unappreciated constraint condition between $\gamma$ and $H$

It is well known that for continuum mechanics problems in physics, distance and time are two fundamentally independent variables, while velocity and acceleration are derivative variables. We will demonstrate what happens when this most common knowledge is violated in the section of “Asymptotic Limits” in the debate paper (Ladd and Szymczak, 2017). Toward this end, we need to introduce the following equations involved to the reaction-infiltration instability problem:

$$\bar{\mathbf{r}} = \frac{\mathbf{r}v_0}{D} \quad (1)$$

$$\bar{\mathbf{v}} = \frac{\mathbf{v}}{v_0}, \quad \bar{c} = \frac{c}{c_{in}} \quad (2)$$

$$\bar{t} = \frac{t}{t_d} = ks\gamma t \quad (3)$$

where  $\mathbf{r}$  and  $\bar{\mathbf{r}}$  are the dimensional and dimensionless distance vectors;  $D$  is the constant diffusion coefficient of the fully dissolved matrix;  $v_0$  is the flow velocity in a uniform (undissolved) matrix;  $\mathbf{v}$  and  $\bar{\mathbf{v}}$  are the dimensional and dimensionless flow velocities;  $k$  is the rate constant with a unit of  $m/s$ ;  $c_{in}$  is the reactant concentration in the incoming fluid stream;  $c$  and  $\bar{c}$  are the dimensional and dimensionless concentrations;  $s$  is the constant surface area per unit volume of the undissolved mineral phase with a unit of  $1/m$ ;  $t$  is the dimensional time with a unit of second;  $\bar{t}$  is the dimensionless time;  $\gamma$  is the (dimensionless) ratio of the reactant

concentration in the incoming fluid stream to the mineral concentration;  $t_d$  is the dissolution time scale with a unit of second. The last two quantities are defined as follows:

$$\gamma = \frac{c_{in}}{c_{sol}} \quad (4)$$

$$t_d = \frac{t_r}{\gamma} = \frac{1}{ks\gamma} \quad (5)$$

where  $c_{sol}$  is the mineral concentration;  $t_r$  is the reaction time scale with a unit of second and is defined in the following form:

$$t_r = \frac{1}{ks} \quad (6)$$

Note that Eqs. (1)–(3) are in correspondence to Eqs. (16), (17) and (21) in the debate paper (Ladd and Szymczak, 2017) respectively. The detailed corresponding relationships between equations used in this paper and in the debate paper (Ladd and Szymczak, 2017) can be found in Table 1.

By considering Eqs. (1)–(3), the following equation exists mathematically:

$$\frac{d\bar{\mathbf{r}}}{d\bar{t}} = \left( \frac{v_0}{ks\gamma D} \right) \frac{d\mathbf{r}}{dt} \quad (7)$$

From fundamental definitions in physics, we have the following relationships:

$$\mathbf{v} = \frac{d\mathbf{r}}{dt}, \quad \bar{\mathbf{v}} = \frac{d\bar{\mathbf{r}}}{d\bar{t}} \quad (8)$$

Substitution of Eq. (8) into Eq. (7) yields the following equation:

$$\bar{\mathbf{v}} = \left( \frac{v_0^2}{ks\gamma D} \right) \frac{\mathbf{v}}{v_0} \quad (9)$$

In the debate paper (Ladd and Szymczak, 2017), the dimensionless transport parameter,  $H$ , is defined as follows:

$$H = \frac{Dks}{v_0^2} \quad (10)$$

Substitution of Eq. (10) into Eq. (9) yields the following equation:

$$\bar{\mathbf{v}} = \left( \frac{1}{\gamma H} \right) \frac{\mathbf{v}}{v_0} \quad (11)$$

It is straightforwardly concluded that the validity condition of the first expression (i.e.  $\bar{\mathbf{v}} = \frac{\mathbf{v}}{v_0}$ ) in Eq. (2) can be described as follows:

$$\frac{1}{\gamma H} = 1 \quad \text{or} \quad \gamma H = 1 \quad (12)$$

Clearly enough, the constraint condition in Eq. (12) is directly contradictory to the quasi-steady state condition, which is derived and presented in Eq. (36) of the debate paper (Ladd and Szymczak, 2017) as follows:

$$\gamma H \ll 1 \quad (13)$$

## 3. The main conclusions in the debate are questionable

In the section of conclusions in the debate paper (Ladd and Szymczak, 2017), there is a paragraph, which says that: “we have offered a critical review of the reaction-infiltration instability, pointing out two significant misconceptions that have arisen from hidden assumptions in the asymptotic analysis. First, that the mineral dissolution ratio ( $\gamma$ ) plays a role in determining the degree of reactant penetration; second, that in the limit  $\gamma \rightarrow 0$  the reactant concentration

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