



## Research papers

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

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## ABSTRACT

In this comment we draw attention to an elementary but consequential mathematical error in this recently published article.

The recent paper “Validity of using large-density asymptotics for studying reaction-infiltration instability in fluid-saturated rocks” (Zhao et al., 2018), hereafter referred to as ZHO, attempts to undermine our critique (Ladd and Szymczak, 2017) of previous work by the same authors (Zhao et al., 2008, 2010, 2013b; Zhao, 2014; Zhao et al., 2014, 2015). However, the main conclusions of ZHO are based on an elementary mathematical error and must therefore be discarded.

In Eq. (8) of ZHO, the authors have confused Lagrangian and Eulerian descriptions of the fluid motion. Although partial differential equations describing reactive transport can be written in either frame (Bear, 1972), in Ladd and Szymczak (2017), hereafter referred to as LS, an Eulerian (or laboratory) frame was chosen; ZHO also use an Eulerian coordinate system, as indicated by Eqs. (22)–(24). In an Eulerian (or laboratory) frame  $r$  and  $t$  are independent variables; therefore, contrary to Eq. (8) of ZHO,  $dr/dt$  is zero and unrelated to the fluid velocity. The core of ZHO (Sections 2 and 3) follows directly from Eq. (8) and therefore their conclusions are not valid. Moreover, the result  $\gamma H = 1$  from Eq. (12) of ZHO makes no physical sense, because  $\gamma H$  is a product of six independent quantities.

ZHO also err in their criticism of the scaling in LS, apparently not understanding the difference between independent and dependent variables. Scaling  $r$  and  $t$  (the independent variables) introduces a change of length and time scales into the equations via the differential operators. But scaling the velocity (a dependent variable) merely introduces a multiplicative factor, which just needs to be kept track of in the equations. That is why the parameter  $H$  has  $v_0$  in it. It is simple to check that Eqs. (22)–(24) from LS are equivalent to the dimensional

form given in Eqs. (1)–(3) of the same paper.

It might escape the reader's notice that the origin of our disagreement with Zhao, Hobbs and Ord does not lie in mathematical details, important as they are. The source of the disagreement comes from their repeated insistence (Zhao et al., 2008, 2010, 2013b,a; Zhao, 2014; Zhao et al., 2014, 2015) that the thickness of a dissolution front is determined solely by the ratio of aqueous to mineral concentrations, the small parameter denoted by  $\gamma$ . According to Zhao et al., in the limit  $\gamma \rightarrow 0$  the dissolution front is inevitably sharp. In this, they are simply repeating a mistake from 30 years ago. Contrary to Chadam et al. (1986), Ortoleva et al. (1987b), it is well-established in the reactive-transport literature that the thickness of the dissolution front is not determined by  $\gamma$ , but by a combination of fluid velocity, reaction rate, and diffusion constant (Bear, 1972; Lichtner, 1988; Phillips, 1990; Steefel and Lasaga, 1990). A number of authors (Hinch and Bhatt, 1990; Steefel and Lasaga, 1990; Wangen, 2013; Szymczak and Ladd, 2014) have noted that the reaction-infiltration model in Chadam et al. (1986), Ortoleva et al. (1987b) is only valid in the limit of fast reactions. In LS we located the error in the asymptotic analysis of Chadam et al. (1986), Ortoleva et al. (1987b) and by implication in the work of Zhao et al.. The validity of the critique made in LS is unaffected by the erroneous analysis presented in ZHO.

Finally, we note that the stability analysis from (Zhao, 2014), summarized in Section 4, is equivalent to previously published work Chadam et al. (1986), Ortoleva et al. (1987b). After correcting a typographical error, Eq. (32) of ZHO reads (Zhao, 2014)

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$$\bar{\omega} = \frac{\gamma Zh}{(1 + \Gamma)(\phi_1 - \phi_0)} [Zh - \sqrt{Zh^2 + 4\bar{m}^2} + (1 - \Gamma)\bar{m}]. \quad (1)$$

As was shown in Section 5 of LS (Eq. (47)), the apparent dependence of the growth rate  $\omega$  on the dimensionless parameter  $Zh$  is illusory. Using the definitions of length and time scales from Eqs. (25) and (26) of ZHO and the definition  $Zh = v_0/\sqrt{Dks\gamma}$  (Zhao et al., 2013a; Zhao, 2014), the dimensionless growth rate  $\bar{\omega}$  and wavenumber  $\bar{m}$  can be written in terms of dimensional quantities as

$$\bar{\omega} = \frac{\omega}{ks\gamma} = Zh^2 \frac{\omega D}{v_0^2}, \quad \bar{m} = \frac{mD^{1/2}}{(ks\gamma)^{1/2}} = Zh \frac{mD}{v_0}. \quad (2)$$

After canceling the common factor of  $Zh^2$  from both sides of Eq. (1), we recover the original result from Ortoleva et al. (1987a) (Eq. (VIII.3)),

$$\frac{\omega D}{\gamma v_0^2} = \frac{1}{(1 + \Gamma)(\phi_1 - \phi_0)} [1 - \sqrt{1^2 + 4(mD/v_0)^2} + (1 - \Gamma)(mD/v_0)], \quad (3)$$

which is independent of reaction rate. Thus the plots of Figs. 1 and 2 of ZHO only repeat the same result with different scalings of the axes. In order to account for finite reaction rates, a more complex theory, including both upstream and downstream concentration fields, is needed (Szymczak and Ladd, 2013, 2014).

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