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## Cyclic time-dependent reactive flow displacements in porous media

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#### HIGHLIGHTS

• This is the first study to analyze time-dependent velocity (t-d-v) reactive flows in porous media.

• T-d-v can destabilize flows that are stable under constant velocity and lead to more chemical production.

- Effects of the t-d-v depend on the viscosity distribution of the chemicals.
- The characteristics of the t-d-v can be used to control the flow and the rate of chemical production.
- Differences between t-d-v and constant velocity flows are discussed and explained.

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#### ABSTRACT

Reactive miscible flow displacements in homogeneous porous media with time-dependent injection velocities are analyzed. The displacements consist of periodic cycles that involve alternating stages of injection and extraction. Nonlinear simulations are used and the dynamics of the flows are characterized qualitatively as well as quantitatively in terms of the amount of generated chemical product. The simulations revealed that, when compared with constant velocity flows with the same overall amount of injected fluid, time dependent displacements show drastically different flow structures and chemical productivity. These differences are strongly dependent on the period of the velocity cycles and the scenario of whether the displacement is initiated through an injection or an extraction process. In particular, for *non-monotonic* viscosity profiles, displacements with larger period result in stronger chemical production while the effects of the velocity scenarios depend on the nature of the instability at the initial front. Moreover, for *monotonically decreasing* viscosity profiles known to be stable under constant injection velocity, only displacements starting with extraction can trigger instabilities and increase the amount of chemical product. Finally for *monotonically increasing* viscosity profiles, displacements that start with injection tend to generate more chemical product than the constant velocity flow and its counterpart starting with an extraction instead.

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#### 1. Introduction

In a flow displacement in a porous medium, there is a tendency of a less viscous displacing fluid to move faster than the more viscous displaced one. This results in an instability that manifests itself in the form of finger shaped intrusions, which is referred to as viscous fingering. This instability can affect very seriously the efficiency of many processes encountered in enhanced oil recovery, fixed bed regeneration, groundwater flows, CO<sub>2</sub> sequestration, soil remediation and filtration. A considerable amount of literature has been published on viscous fingering since it was first examined by Hill (1952). Extensive literature reviews on this instability are found in Homsy (1987) and McCloud and Maher (1995).

While most authors have focused on the miscible displacements for non-reactive flows, there are reactive flows where chemical reaction takes place between two fluids. This kind of displacements can be found in fixed bed regeneration, chromatographic separation, polymer synthesis and processing (Hejazi and Azaiez, 2010b). Discussions of the most recent studies dealing with the reactive flows are found in Ghesmat and Azaiez (2009), Hejazi and Azaiez (2010a), Nagatsu and De Wit (2011), Riolfo et al. (2012) and Alhumade and Azaiez (2013).

To date, most of the existing studies dealing with miscible displacements in homogeneous porous media have been limited to flows where the injection velocity is constant. However, in many processes the injection velocity may actually be time dependent resulting in different flow dynamics. For example, in

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trickle-bed reactors (Zhuang et al., 2012), different cycling liquid feed strategies were developed to increase the mass transfer rate of the limiting reactant and to prevent flow maldistribution and hot spot formation (Boelhouwer et al., 2002). The flow rates are forced to be time-dependent by using appropriate algorithms where sinusoidal, pulsing, and heaviside velocity profiles were adopted (Nguyen et al., 2009). Other examples of time-dependent injection velocities are encountered in the energy sector, namely in some processes of enhanced oil recovery. These include the cyclic steam stimulation (CSS) process which involves three stages of injection, soaking and extraction/production (Mago et al., 2005). A similar process known as CO<sub>2</sub> huff-and-puff involves cyclic injection of liquid CO<sub>2</sub> for heavy as well as light oil enhanced recovery (Monger et al., 1991). The efficiency of these processes, which can be run as either miscible or immiscible, depends on the importance and duration of each stage of the cycle.

To the best knowledge of the authors of this article, there are no studies dealing with time-dependent velocities that have focused on *reactive* flows. There are however few studies that have examined such flows in the case of non-reactive displacements. In particular, an experimental study of time-dependent flow rates in Enhanced Oil Recovery (EOR) was conducted by Davidson et al. (1999) and Spanos et al. (2003). The authors used a Consistent Pulsing Source (CPS) to generate pulsing water injection to immiscibly displace the oil in a sand pack. It was found that time-dependent flow rates can indeed suppress the fingering instability and lead to a sweep efficiency more than 10% larger than that of constant flow rates. The same authors have also examined a process involving first the injection of oil in the producer and then the displacement with water in the injection well, and reported that this allows to produce more oil in an old reservoir with full water breakthrough. However, no definitive explanation or criteria to predict the flow rate enhancement were proposed. In the field of solute transport in groundwater systems, time-dependent velocities were also considered to study the effects of seasonal groundwater velocity and water level on contaminant concentration in water (Singh et al., 2009). These authors proposed analytical solutions of the one-dimensional concentration transport equation for sinusoidal and exponential velocities but did not actually solve the coupled momentum and mass transport equations and hence the flow instability was not addressed. More recently, Dias et al. (2010), Dias and Miranda (2010), and Dias et al. (2012) studied the suppression of viscous fingering of two-phase immiscible flows in a radial geometry with time-dependent flow rates. They reported the optimal flow rates for linear flow regime and nonlinear flow regime. Motivated by the study of Dias and Miranda (2010), Chen et al. (2010) investigated miscible flow displacements in a radial Hele-Shaw cell with injection rates that decayed in time like  $t^{-1/3}$ . The width of fingers was found to be more uniform and symmetric than that in corresponding constant injection flows. In addition, it was reported that this time dependent injection rate leads to less unstable displacements than constant velocity with the same amount of injection at equal times.

In the present study we propose to examine the dynamics of the reactive interfaces instability in a rectilinear geometry under step-size time dependent velocity profiles. The objective is to characterize the development of the instability and analyze the effects of the flow cycle period and the injection scenarios in the case of reactive flows. The ultimate objective being to determine the conditions that result in the largest or the smallest amount of product and the intent is to provide a better insight of the flow as well as criteria to optimize practical process where reaction is to be enhanced or suppressed. The study is different from the few existing ones that have modeled time-dependent flows in a number of aspects. First, the rectilinear geometry adopted here is different from the radial one examined by Dias et al. (2010) for immiscible flows and by Chen et al. (2010) for non-reactive ones, since the former involves a fixed initial interface while the latter has a point source injection and results in a contact interface that expands as the flow evolves. Furthermore, unlike the previous study dealing with miscible flows (Chen et al., 2010), the present work examines the reactive displacements and analyzes the effects of the velocity period as well as scenarios. As shall be seen later, depending on the flow conditions, the amount of chemical product of the time-dependent displacement velocity can actually be larger or smaller than that of its constant velocity counterpart, with the same average flow rate.

The formulation of the problem and the numerical techniques are presented in Section 2. The discussions of the results of nonlinear reactive flows dynamics for time dependent velocities are given in Section 3. This is followed by Section 4 where quantitative analysis is presented and the effects of velocity period and different velocity scenarios are analyzed. Finally, a conclusion is given in Section 5.

#### 2. Model equations

#### 2.1. Physical problem

A horizontal two-dimensional displacement in a homogeneous porous medium is considered. The porous medium is assumed to have constant porosity  $\phi$  and permeability K and the fluids are incompressible, Newtonian and completely miscible. The flow is taking place along the *x*-axis and the initial interface between the fluids is parallel to the *y*-axis. A schematic of the two-dimensional porous medium is shown in Fig. 1.

A fluid (A) of viscosity  $\mu_A^*$  is injected from the left-hand boundary to displace another one B of viscosity  $\mu_B^*$ . The displacement velocity  $u^*(t^*)$  is time dependent, with a positive velocity corresponding to an injection process, a negative one representing an inverse displacement or a production (extraction) process while a zero velocity corresponds to soaking process. A simple bimolecular chemical reaction (BCR) (Gálfi and Rácz, 1988) occurs between the two fluids resulting in the formation of a product (C) with viscosity  $\mu_C^*$ :

$$(A) + (B) \to (C) \tag{1}$$

It is assumed that initially and before the start of the displacement, the interface between the two reactants (A) and (B) is stable and their respective initial concentrations are  $a(x, 0) = a_0H(x)$  and  $b(x, 0) = b_0H(-x)$ , where H(x) is the heaviside step function. A chemical product (C) is generated by a second order chemical reaction with a reaction rate RR(x, t) = ka(x, t)b(x, t), where k is the reaction constant. The mixing between the two reactants is initially slow and dominated by diffusion, but will be enhanced



Fig. 1. Schematic of the reactive flow system.

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