Reversible Reactive Flow Displacements in Homogeneous Porous Media

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Abstract—**The viscous fingering instability of miscible flow displacements in a homogeneous porous media is examined in the case involving reversible chemical reactions between the fluids. The problem is formulated using continuity equation, Darcy's law, and volume-averaged forms of convection-diffusion-reaction equation for mass balance. Full nonlinear simulations using a pseudo-spectral method combined with semi-implicit finite-difference time-stepping algorithm, allowed to analyze the mechanisms of fingering instability that result from the dependence of the fluids viscosities on the concentrations of the different species. In particular, the study examined the effects of varying important parameters namely the Damkohler number that represents the ratio of the hydrodynamic and chemical characteristic time scales, and the chemical reversibility coefficient, and analyzed the resulting changes in the finger structures.**

Index Terms—**Viscous fingering, miscible displacements, Reversible chemical reaction, Homogeneous porous media.**

I. INTRODUCTION

When a viscous fluid is used to displace another one of a larger viscosity, a frontal instability appears at the interface between the two fluids, which may dramatically affect the overall efficiency of the displacement process. This instability may grow to form fingers that propagate in both upstream and downstream directions and is referred to as fingering or Saffman-Taylor instability [1]. The instability can be triggered by either viscosity mismatch and is referred to as viscous fingering or density mismatch, where it is known as the Rayleigh-Taylor instability. Such instabilities are encountered in a wide variety of processes that include enhanced oil recovery, soil remediation, chromatography and CO_2 sequestration. Many experimental and theoretical studies have focused on the frontal instability of non-reactive displacement processes, where hydrodynamic interactions between the fluids result in the viscous fingering instability. In these studies the effects of different parameters were examined and most of these studies were reviewed in [2-4].

The viscous fingering instability may develop in conjunction with chemical reactions in a wide variety of processes such as underground water treatment, tertiary heavy oil recovery, spreading of chemical pollutants

chromatographic separation, polymer synthesis and processing as well as fixed bed regeneration [3]. There has been a growing interest in analyzing such reactive flow instability and a number of studies have examined the reactive flow. One of the earliest studies of reactive displacements in porous media was conducted by out by Naser-El-Din et al. [5], where the reaction leads to an interfacial tension decrease in a secondary oil recovery process. A number of subsequent experimental studies examined the effects of different parameters such as stoichiometry [6], geometry orientation [7], finger growth rate [8], chemical composition [9], external electrical field [10], variation in the physical properties of the phases [11], and precipitation [12].

Analytical and numerical Modelling of reactive flow displacements has been carried out by a limited, but growing number of studies [13-18]. These studies have considered either auto-catalytic or non-autocatalytic reactions.

All existing studies dealing with reactive viscous fingering have assumed the chemical reaction to be complete. However the reversibility of the reaction plays an important role in many phenomena studied in physics, chemistry, biology, and geology [19]. For example, in the in-situ soil remediation, promising results were reported by Zhang, 2003 [20], where a reactive fluid was injected to remove the pollutant from the underground water. The first study on reactive-diffusive systems with reversible reaction was carried out by Chopard et al., (1993) [21] who studied the properties of a reversible reactive front with initially separated reactants. It was reported that the dynamics of the reactive front can be described as a crossover between irreversible and reversible regimes at short and long times, respectively. A subsequent study [22] confirmed the existence of a crossover between short time "irreversible" and long-time "reversible" regimes. In a recent study, Sinder et al., (2011) [23] investigated the reaction rate of a reversible reactive-diffusive process when the reactants are initially mixed with different diffusion coefficients by using the boundary layer function method. The authors reported that the reactive-diffusive process for this case can be considered as a quasi-equilibrium process and analyzed the dependence of the reaction rate on the initial distribution of the reactants.

In all previous studies of reversible reactive displacement processes, only the diffusive effects were considered. However, when the injection of one of the reactants is required, the convective term needs to be considered in addition to the diffusive one. In such a case, the displacement process is expected to be dominated by convection rather than diffusion. Therefore, it is important to analyze reversible reactive-diffusive-convective flow displacements to understand the effects of reversibility on

the flow dynamics as well as on the overall efficiency of the displacement process.

II. MATHEMATICAL MODEL

A two-dimensional displacement is considered in which both fluids are incompressible and fully miscible. The flow takes place in horizontal direction in a homogeneous medium of constant porosity ϕ and permeability K. A schematic of the two-dimensional porous medium is shown in Fig. 1. The length, width and thickness (z-direction) of the medium are Lx, Ly and b respectively.

The medium is assumed to be initially filled with a solution of a reactant (B) of viscosity μ_B. A miscible fluid (A) of viscosity μ_A is injected from the left-hand side with a uniform velocity U to displace fluid (B). The direction of the flow is along the x-axis and the y-axis is parallel to the initial plane of the interface. A reversible chemical reaction occurs between the two fluids leading to the formation of a product (C) of viscosity μ_C:

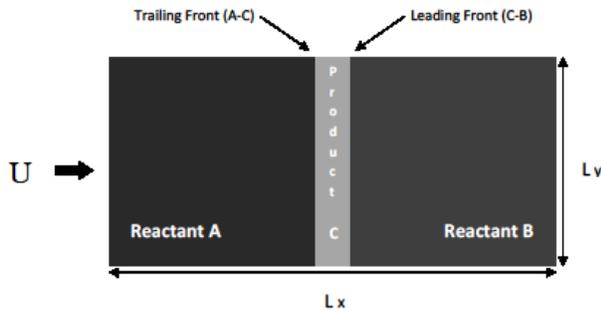$$A + B \leftrightarrow C \tag{1}$$

Figure 1: Schematic of a two-dimensional porous medium.

As time proceeds, more product accumulates at the interface between the two reactants. Figure 1 shows an idealized distribution of the two reactants (A) and (B) and the product (C), with two fronts. One between the reactant (A) and the product (C); (A - C) while the other is between the reactant (B) and the product(C); (C - B), and they are referred to as the trailing and the leading front, respectively. It should be stressed that this is an idealization of the system and the three chemical species are actually present to a more or less degree everywhere in the region where the reaction takes place. However, this concept of a leading and trailing front will be helpful in the interpretation and explanation of the results.

The flow is governed by the equations for conservation of mass, momentum (Darcy's Equation) and the transport of the three chemical species.

$$\nabla . \mathbf{v} = 0 \tag{2}$$

$$\nabla p = -\frac{\mu}{K} \mathbf{v} \tag{3}$$

$$\frac{\partial A}{\partial t} + \frac{u}{\varphi}\frac{\partial A}{\partial x} + \frac{v}{\varphi}\frac{\partial A}{\partial y} = D_A\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2}\right) - kA.B + k_r C \tag{4}$$

$$\frac{\partial B}{\partial t} + \frac{u}{\varphi}\frac{\partial B}{\partial x} + \frac{v}{\varphi}\frac{\partial B}{\partial y} = D_B\left(\frac{\partial^2 B}{\partial x^2} + \frac{\partial^2 B}{\partial y^2}\right) - kA.B + k_r C \tag{5}$$

$$\frac{\partial C}{\partial t} + \frac{u}{\varphi}\frac{\partial C}{\partial x} + \frac{v}{\varphi}\frac{\partial C}{\partial y} = D_C\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2}\right) + kA.B - k_r C \tag{6}$$

In the above equation, $\mathbf{v} = u\mathbf{i} + v\mathbf{j}$ is the velocity vector with u and v the x- and y- components respectively, p the pressure, μ the viscosity, K the medium permeability and ϕ its porosity. The concentrations of the two reactants and the product are denoted by A, B and C, respectively while D_A, D_B and D_C are their corresponding diffusion coefficients. Furthermore, k is the reaction constant while k_r represents the reverse reaction constant. For simplicity, it will be assumed that all species have the same diffusion coefficient, i.e. $D_A = D_B = D_C = D$.

Since the characteristic velocity for the fluid flow through the porous medium is U/ϕ, we adopted a Lagrangian reference frame moving at a velocity U/ϕ. Furthermore, diffusive time $D\phi^2/U^2$ and diffusive length $D\phi/U$ are chosen to make the length and time dimensionless. The constant permeability K is incorporated in the expression of the viscosity by treating μ/K as μ, and we shall refer to ratios of μ as either viscosity or mobility ratios. The rest of the scaling is as follows: the velocity is scaled with U/ϕ, the viscosity and pressure with μ_A and $\mu_A D\phi$, respectively, and the concentration with that of the pure displacing fluid, A_0. The dimensionless equations are:

$$\nabla^* . \mathbf{v}^* = 0 \tag{7}$$

$$\nabla^* p^* = -\mu^*(\mathbf{v}^* + \mathbf{i}) \tag{8}$$

$$\frac{\partial A^*}{\partial t^*} + \mathbf{u}^* . \nabla^* A^* = \nabla^{*2} A^* - D_a A^* B^* + D_r C^* \tag{9}$$

$$\frac{\partial B^*}{\partial t^*} + \mathbf{u}^* . \nabla^* B^* = \nabla^{*2} B^* - D_a A^* B^* + D_r C^* \tag{10}$$

$$\frac{\partial C^*}{\partial t^*} + \mathbf{u}^* . \nabla^* C^* = \nabla^{*2} B^* + D_a A^* B^* - D_r C^* \tag{11}$$

In the above equations, dimensionless variables are represented with asterisk while $D_a = kA_0 D/U^2$ is the Damkohler number representing the ratio of hydrodynamic to chemical characteristic times and $D_r = k_r D/U^2$ represents a reversible Damkohler number.

Two additional dimensionless groups are also involved, namely the Péclet number $Pe = UL_x/D$ and the cell aspect-ratio $A_r = L_x/L_y$ that appear in the boundary conditions [26].

Following previous studies, an exponential concentration dependent viscosity model is adopted to complete the model [24-26],

$$\mu^* = exp\left(R_b B + R_c C\right) \qquad (12)$$

where Rb and Rc are the log-mobility ratios between the species as follows:

$$R_b = ln\left(\frac{\mu_B}{\mu_A}\right), \qquad R_C = ln\left(\frac{\mu_C}{\mu_A}\right) \qquad (13)$$

An associated mobility ratio at the chemical front between the chemical product (C) and the reactant (B), and between the reactant (A) and the product (C) can be also defined as:

$$R_{CB} = ln\left(\frac{\mu_B}{\mu_C(C=0.5)}\right) = R_b - \frac{R_C}{2} \qquad (14)$$

$$R_{AC} = ln\left(\frac{\mu_C(C=0.5)}{\mu_A}\right) = \frac{R_C}{2} \qquad (15)$$

It should be stressed here that the different fronts, whether it is the initial reactive front between (A) and (B) or the idealized leading and trailing ones will be unstable whenever their mobility ratios are strictly positive, while they will be stable if the mobility ratios are negative or zero.

For convenience, in all that follows, the asterisks will be dropped from all dimensionless variables.

III. NUMERICAL TECHNIQUE

The above problem is formulated using a stream-function vorticity formulation, where the velocity field, the stream-function ψ and the vorticity ω are related as:

$$u = \frac{\partial \psi}{\partial y}, \quad u = -\frac{\partial \psi}{\partial x}, \quad \nabla^2 \psi = -\omega \qquad (16)$$

In the above equation ∇^2 is the Laplacian operator. With this formulation, the continuity equation is satisfied automatically and the convection dispersion equations take the forms:

$$\frac{\partial A}{\partial t} + \frac{\partial \psi}{\partial y}\frac{\partial A}{\partial x} - \frac{\partial \psi}{\partial x}\frac{\partial A}{\partial y} =$$
$$\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2}\right) - D_a\, A.B\ + D_r C \qquad (17)$$

$$\frac{\partial B}{\partial t} + \frac{\partial \psi}{\partial y}\frac{\partial B}{\partial x} - \frac{\partial \psi}{\partial x}\frac{\partial B}{\partial y} =$$
$$\left(\frac{\partial^2 B}{\partial x^2} + \frac{\partial^2 B}{\partial y^2}\right) - D_a\, A.B\ + D_r C \qquad (18)$$

$$\frac{\partial C}{\partial t} + \frac{\partial \psi}{\partial y}\frac{\partial C}{\partial x} - \frac{\partial \psi}{\partial x}\frac{\partial C}{\partial y} =$$
$$\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2}\right) + D_a\, A.B\ - D_r C \qquad (19)$$

The pressure term is eliminated by taking the curl of equation (8) resulting in the following relationship between the vorticity and the concentrations of the three chemical species:

$$\omega = R_b\left(\frac{\partial \psi}{\partial x}\frac{\partial B}{\partial x} + \frac{\partial \psi}{\partial y}\frac{\partial B}{\partial y} + \frac{\partial B}{\partial y}\right)$$
$$+ R_c\left(\frac{\partial \psi}{\partial x}\frac{\partial C}{\partial x} + \frac{\partial \psi}{\partial y}\frac{\partial C}{\partial y} + \frac{\partial C}{\partial y}\right) \qquad (20)$$

Equations (16)-(20) form a closed set that can be solved for the concentration and velocity fields. The system of partial differential equation is solved by decomposing the variables as a base-state and a perturbation. The perturbation terms consist of a random noise centered at the initial interface between the reactants A and B, with the magnitude of the noise decaying rapidly away from the interface. The resulting system of three partial differential equations is solved using a highly accurate pseudo-spectral method based on the Hartley transform [26, 27]. This method allows to recast the partial differential equation in time and space into an ordinary differential equation in time. The solution for the time stepping of the reactive-diffusive-convective equations was generated by using a semi-implicit predictor-corrector method along with an operator-splitting algorithm.

IV. RESULTS

A. Numerical Code Validation

The numerical code has been validated by comparing the time evolution and the related viscous fingers interactions to those presented by Hezaji and Azaiez (2010) [26] for the non-reversible case (Dr=0). It has been noted that the dynamics of fingering were identical when the same parameters were used along with the same spatial resolution and time step size. In addition, the numerical convergence of the numerical results has also been tested by varying the spatial resolution and the time step. In this study, unless mentioned otherwise, a spatial resolution of 256 x 256 is used along with a time step dt=0.005.

The flow evolution will depend on the mobility ratios of the different species; R_b and R_c as well as Pe, A_r, Da and D_r. In order to limit the analysis to the effects of the reversibility of the chemical reaction, the following parameters are fixed as Ar=2, Pe=1000 and Da=1, unless mentioned otherwise.

For brevity and illustration purposes, the time sequences will not be always presented necessarily at the same time intervals, and only the frames that reveal new and interesting finger structures that help in the discussion, are shown. Moreover the analysis and discussion will examine the case where the initial front between the two reactants is unstable, i.e. $\mu_A < \mu_B$ or equivalently for $R_b>0$.

B. Effects of the Chemical Reaction

Prior to discussing the effects of chemical reversibility, a brief explanation of the effects of chemical reaction on the viscous fingering instability is presented. More details can be found in [26]. Figure 2 shows a time sequence of contours of the displacing fluid in the case of non-reactive flow displacement for a mobility ratio of Rb=3. This implies that the displacing fluid (A) is less viscous than the displaced one and as a result, an instability in the form of finger shaped structures develops at the interface. As the flow evolves, the fingers extend in the flow and grow through different mechanisms of interactions.
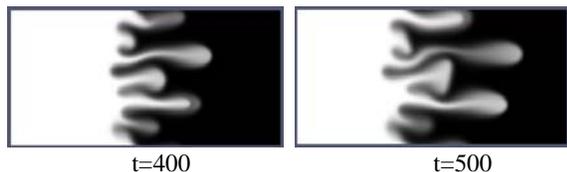

t=200	t=300

Figure 2: Contours of (A) for a non-reactive flow: Rb=3.

Corresponding contours when a reaction takes place in the flow leading to a chemical product (C) with Rc=5, are shown in Figure 3. The Damkohler number in this case is equal to 0.5. Clearly the frontal instability is modified by the reaction resulting in a larger number of fingers that tend to be thinner and to have more complex structures. In what follows, we will focus on the effects of the chemical reversibility (Dr) on the flow in the case of a reactive flow displacement.

For the purpose comparisons with later results, contours of the chemical product (C) for case depicted in Figure 3 are shown in Figure 4. First, it should be noted that contours of the chemical product actually allow to illustrate the finger structures of the displacing fluid (A) (compare the trailing front in Figure 4 with those in Figure 3) as well as those of the displaced one (B) through the leading front. This indicates that plots of the chemical product contours actually allow to show the development of the instability at all fronts, and hence it will be used in all subsequent figures. Note that both the leading and trailing fronts are unstable. Furthermore, as time proceeds and more product is generated, the fingers develop more and extend both upstream and downstream.
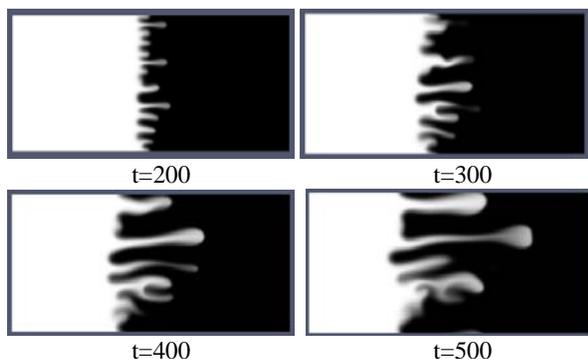

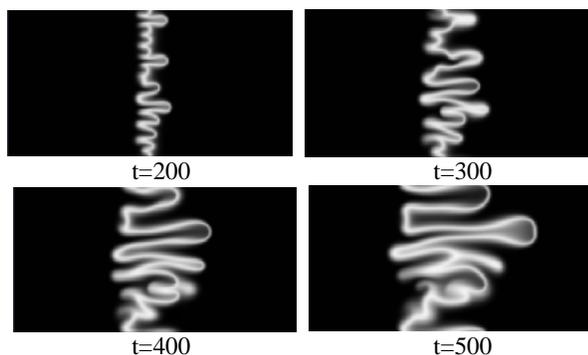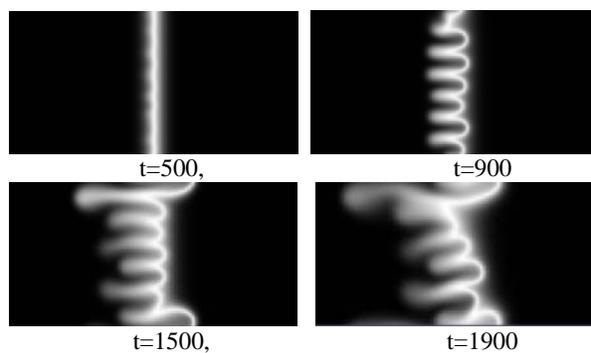**Figure 3**: Contours of (A) for a reactive flow: Rb=3, Rc=5, Da=0.5.

Figure 4: Contours of the chemical product (C) for a reactive flow: Rb=3, Rc=5, Da=0.5.
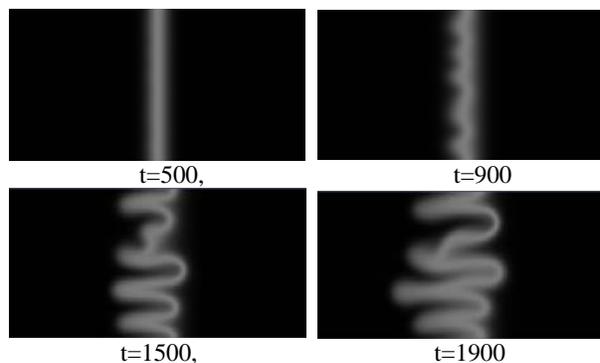
C. Effects of the Chemical Reversibility

Figure 5 depicts contours of the chemical product (C) in the case of a chemical reaction with $R_b=1$ and $R_c=3$. These numerical values imply that the chemical product (C) is more viscous than the reactant (B) which itself is more viscous than the reactant (A). It should be noted that in this case the trailing front is expected to be unstable (Rc/2>0) while the leading one should be stable (Rb-Rc/2<0). Figure 5-a shows the contours for the non-reversible case (Dr=0) while figure 5-b presents the corresponding contours for the reversible reaction case with Dr=0.3.

In the non-reversible reaction flow, the instability develops early in time and result in quite complex fingers that extend mainly in the upstream direction. Note that by t=1900, the fingers have almost reached the left side of the domain.

t=500,   t=900

t=1500,   t=1900

(a)

t=500,   t=900

t=1500,   t=1900

(b)

Figure 5: Contours of the chemical product (C) for a reactive flow: Rb=1, Rc=3, Da=1, (a) Dr=0, (b) Dr=0.3.

It is clear in this case that reversibility results in a less number of fingers, which actually are less developed and extend less in the upstream and downstream directions. Note also that the fingers are more diffuse and less complex than in the non-reversible reaction flow (Figure 5-a). Furthermore, as expected, the amount of product in the medium has also decreased. It is however interesting to note that the leading front between (C) and (B) is virtually stable in the case of the flow involving a non-reversible reaction (Figure 5-a) while actually it has developed instabilities when there is chemical reversibility (see Figure 5-b namely at t=1900). This result may be explained by the fact that in the non-reversible case the leading (downstream) front is

actually stable because the chemical product (C) is more viscous than the displaced reactant (B). However, as the reaction reverses, less chemical product is expected to be present at the leading front, giving rise to more reactants to be in contact and therefore a more unstable front (recall Rb>0).

Contours for flow displacements with Rb=3 and Rc=2 are shown in Figure 6. This mobility distribution implies that the chemical product is more viscous than the displacing reactant (A) but less viscous that reactant (B). Furthermore, in this case the leading and trailing fronts defined earlier are unstable (Rb-Rc/2>0, Rc/2>0), and hence one expects that both of these fronts will develop fingering structures. This is well illustrated in Figures, where one observes instability developing on both the trailing and leading fronts, though the one on the leading front is stronger. This is not surprising as R_{CB}=Rb-Rc/2 is larger than R_{AC}=Rc/2.
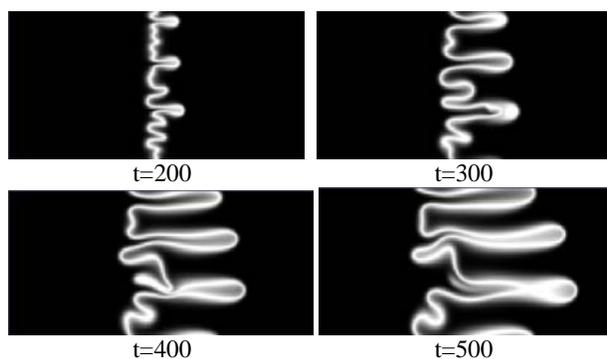

t=200	t=300
t=400	t=500

Figure 6: Contours of the chemical product (C) for a reactive flow: Rb=3, Rc=2, Da=1, Dr=0.
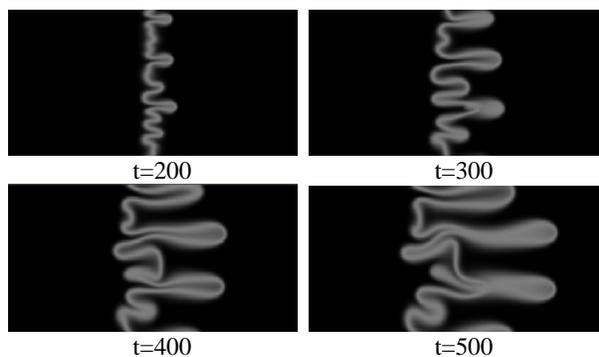

t=200	t=300
t=400	t=500

Figure 7: Contours of the chemical product (C) for a reactive flow: Rb=3, Rc=2, Da=1, Dr=0.3.

The corresponding contours for the reversible case with Dr=0.3 are shown in Figure 7. It is very interesting to note that in this particular case, even though reversibility leads to more diffusive finger structures and obviously less chemical product, the number of fingers, their general shape as well as their size are virtually unchanged. This result is to be contrasted with the previous one where the reversibility of the chemical reaction has actually led to a reduction of the number of fingers, their size and complexity. This interesting result can be explained by examining the values of the different mobility ratios, namely Rb=3, R_{AC}=1 and R_{CB}=2. As less product is generated in favour of the chemical reactants as a result of the reversibility, the mobility ratio at the trailing front (AC) will effectively

increase since now there is more of the two reactants in contact and the mobility of these two reactants, Rb, is larger than that at the trailing front R_{AC}. The same is also true at the leading front where one has again the mobility ratio Rb larger than R_{CB}. This increase in the expected instability at both the leading and trailing front, is however nuanced by the fact that it results in more mixing between the different chemicals that help instability to keep growing regardless of how fast product C is converted back to A and B.

V. CONCLUSION

The effects of chemical reversibility on the development of the viscous fingering instability in the case of reactive flow displacements in porous media were examined. It was found that even though reversibility leads to less chemical product being generated in the flow, it may or may not affect the hydrodynamic instability. Indeed, depending on the mobility ratios of the reactants and the chemical product, reversibility can attenuate the instability resulting is less fingers that are more diffuse and less complex than in the case of a non-reversible reaction. However, it was also found that reversibility may actually not affect the hydrodynamic instability of the displacement leading to similar finger structures as those observed in the non-reversible reaction case. This was observed in flows where the viscosity of the chemical product lies between those of the two chemical reactants.

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