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Numerical investigation of oscillatory multiphase flow in porous medium with chemically active skeleton

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Abstract. Self-oscillating mode of reaction front propagation in multiphase flow in the porous medium with chemically active skeleton is investigated numerically. The considered flow represents an immiscible displacement process, such that the displacing fluid and the skeleton of the porous medium have chemically active components which react with production of gaseous phase. The calculations have demonstrated strong influence of the reaction kinetics on stability of the reactive flow. The presence of a time delay between the change of concentration of the reactants and the change of the reaction rate is shown to stimulate transition of the reaction front propagation to the oscillatory mode.

1. Introduction

The unstable stationary-propagating fronts (burning and phase transitions included) appear within different branches of mathematical physics and continuum mechanics: in the case of the Kolmogorov–Piskunov non-linear parabolic equations, in the theory of combustion (the diffusion-thermal instability, the Darrieus–Landau hydrodynamic flame instability [1], the instability of detonation waves). Instability can manifest itself with respect to one- or multi-dimensional disturbances. The result of the development of small perturbations is the often-pulsating self-oscillating mode of propagation.

There is hydrodynamic and structural instability of the waves. The instability of the first type can be caused, for example, by the non-standard thermodynamic properties of substances. The second type of instability is caused by the internal structure of the wave. The instability may be caused by both the release of energy in the front (for example, the detonation wave [2]) and the loss of energy (radiation wave [3]).

Within the framework of filtration combustion theory, a multi-component single-phase flow of gas through the porous medium is usually studied. The components of such flow include oxidizer, combustion products and inert components. Within the framework of the theory of filtration combustion a multi-component (oxidizer, combustion products, inert components) single-phase (usually a gas) flow through the porous matrix is considered. The reaction of the oxidizer with the fuel contained in the skeleton leads to an increase in temperature and combustion wave propagation under the influence of heat conduction. The theory of such processes relative to the synthesis of new materials has been developed over the past few decades. It has accumulated a large experimental and theoretical data.



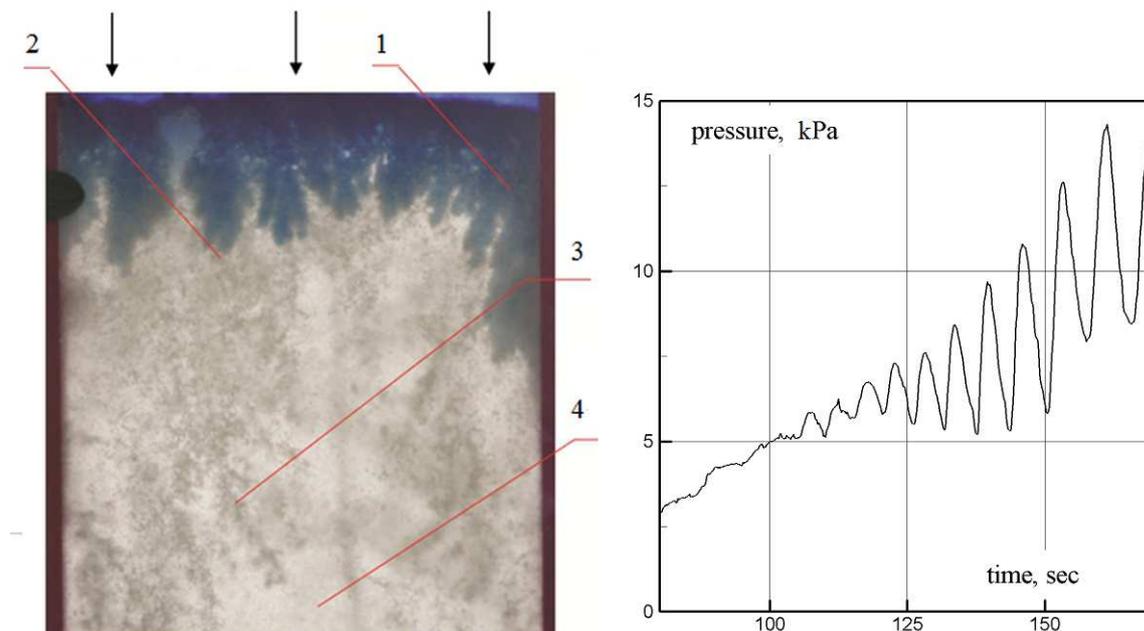


Figure 1. Left panel—photograph of the flow field in the experiment on the acid solution-oil (petroleum jelly) displacement in the porous medium containing NaHCO_3 [13]: 1—acid solution (blue), 2—the displacement front, 3—the flow of the gas (CO_2), 4—oil. Right panel—example of the pressure sensor data demonstrating spontaneous oscillations (the sensor is located at the center of the cell).

The speed of propagation of the flame front is established, which depends on the thermal properties of the fluid phase and the skeleton, of the kinetics of chemical reactions and energy in the co-current, and the counter-current flows. Stability of stationary filtration combustion waves was considered in [4]. Criteria for the instability of waves of filtration combustion have been derived [5–7]. Diffusion (the rate of combustion is limited by the diffusion fluxes) and kinetic (determined by the rate of reaction kinetics) propagation modes of the combustion front have been considered. Self-oscillating modes of multiphase flow with phase transitions in the porous medium were obtained in [8,9]. The mechanism of self-oscillation, however, remains unclear [10].

The oscillating modes of propagation of the combustion front in relation to the oil and gas topics were discovered in the experiment [11]. Recent experimental work in which the oscillations have been detected is [12]. In [13] it was shown experimentally that the oscillation modes of propagation of waves of reactive displacement are possible under isothermal conditions.

In [13] homogeneous mixture of glass balls and NaHCO_3 powder is placed into the cell with transparent walls. The porous medium is saturated with oil. Then the displacement of the resident fluid (oil) by the acetic acid solution is conducted. The multi-phase flow is driven by the pressure difference between inflow and outflow boundaries (figure 1). Propagation of the reaction front along the cell is accompanied by intensive production of the gas phase at the reaction front. Since the heat of the reaction is small, the conditions may be considered to be isothermal. Spontaneous transition of multiphase flow to oscillatory mode has been observed under nearly stationary boundary conditions. The mechanism of the self-oscillating mode of the reactive displacement includes rise of porous pressure in the reaction zone caused by gas phase production and non-stationary multi-phase transport which provides the flux of the acid solution to the reaction zone.

The appearance of self-oscillating reaction waves is suggested to depend on the reaction kinetics. In the next sections a mathematical model is introduced, which takes into account the time delay between change of the concentrations and the rate of the reaction, then influence of the kinetics on the oscillating mode is investigated numerically. In connection with the multiphase reactive flow in porous medium the time delay may be explained by the capillary nonequilibrium. This possibility is discussed in the last section.

2. Problem formulation

The problem of injection of oxidizing solution into the porous medium with chemically active skeleton saturated initially by an inert fluid is considered. The skeleton contains chemically active component, which reacts with the component contained in the injected fluid. One-dimensional problem formulation is considered. At the inlet and outlet boundaries constant pore pressure is assumed. During filtration, the active components react with production of the gas phase $A + B = C + W + G$, A is the active component of the fluid, B is the active component of the skeleton, W is the water, G is the gas (in the experiment [13], $A = \text{CH}_3\text{COOH}$, $B = \text{NaHCO}_3$, $C = \text{CH}_3\text{COONa}$, $G = \text{CO}_2$).

Let m be the number of moles of the component B per unit volume of the porous medium, S —volume fraction of pore space occupied by the solution, c —concentration of the oxidizer in the solution. Consider volume averaged rate of the chemical reaction (mol/(m³s)) given by the relation:

$$\hat{r} = \frac{1}{\tau_{\text{chem}}} mSc. \quad (1)$$

Here, it is assumed that the product mS is proportional to the area of contact of oxidizing and reducing agents in unit volume of the porous medium and the flow of the oxidizer to the surface is proportional to its concentration. The multiplier $1/\tau_{\text{chem}}$, being the inverse characteristic timescale of chemical reaction, includes the factors of proportionality. The actual dependence for the volume averaged reaction rate can be significantly more complicated. For the purposes of this study it is important to take into account the time delay between change of the concentrations of the active components and the resulting change of the rate of chemical reaction. This time delay is caused by the presence of the resident phase. We introduce the retarded rate of the reaction on the basis of the following equation

$$\tau \frac{\partial r}{\partial t} + r = \hat{r}, \quad (2)$$

where τ is relaxation time.

Mathematical model of the flow is formulated using a number of simplifying assumptions (isothermal flow, incompressibility of liquids, independence of viscosity and density of the solution on its composition), which correspond approximately to the conditions of the laboratory experiment [13]. In view of these assumptions, the equations, which describe the flow under consideration, are as follows

$$\begin{aligned} \frac{\partial}{\partial t}(\phi S \rho_s) + \frac{\partial}{\partial x}(W_s \rho_s) &= -\zeta_A r + \zeta_W r + \zeta_C r, & \frac{\partial}{\partial t}(\phi S \rho_s c) + \frac{\partial}{\partial x}(W_s \rho_s c) &= -\zeta_A r, \\ \frac{\partial}{\partial t}(\phi S_g \rho_g) + \frac{\partial}{\partial x}(W_g \rho_g) &= \zeta_G r, & \frac{\partial}{\partial t}(\phi S_o \rho_o) + \frac{\partial}{\partial x}(W_o \rho_o) &= 0, & \frac{\partial m}{\partial t} &= -\zeta_B r, \\ \phi &= \phi_0 + (m_0 - m)/\rho, & k &= k_0 (\phi/\phi_0)^\lambda, \end{aligned} \quad (3)$$

where the following notations are used: ϕ is the porosity, S , S_o , S_g are water, oil and gas saturations, ρ_s , ρ_g , ρ_o are the molar densities of the acid solution, gas and oil, respectively, c is a concentration of the acid in the solution, ζ_A , ζ_B , ζ_C , ζ_W , ζ_G , are stoichiometric coefficients, r

is a rate of chemical reaction, m is averaged molar density of the reducing agent, W_s , W_g , W_o is the rate of filtration of water, gas and oil, respectively. It is assumed that the filtration rates are determined by the Darcy law for multiphase flow with quadratic relative permeabilities.

$$W_\alpha = -k \frac{f_\alpha}{\mu_\alpha} \frac{\partial p}{\partial x}, \quad \alpha = \{s, g, o\} \quad (4)$$

$$f_s = S^2, \quad f_g = (1 - S - S_o)^2, \quad f_o = S_o^2.$$

Now we will introduce the scales for the length L , molar density ρ_{ref} , viscosity μ_{ref} , porosity ϕ_0 , absolute permeability k_0 , pressure p_{ref} . Let the time scale be $t_{\text{ref}} = \phi_0 \mu_{\text{ref}} L^2 / (k_0 p_{\text{ref}})$ and the scale of averaged molar density m of the component B : $m_{\text{ref}} = \phi_0 \rho_{\text{ref}}$. It is assumed that L is the length of the cell, ρ_{ref} , and μ_{ref} are the density and viscosity of water phase, p_{ref} is the pressure difference between the inflow and outflow boundaries. After change to the dimensionless variables

$$\bar{t} = \frac{t}{t_{\text{ref}}}, \quad \bar{x} = \frac{x}{L}, \quad \bar{\rho}_\alpha = \frac{\rho_\alpha}{\rho_{\text{ref}}}, \quad \bar{\mu}_\alpha = \frac{\mu_\alpha}{\mu_{\text{ref}}}, \quad \bar{m} = \frac{m}{m_{\text{ref}}}, \quad \bar{\phi} = \frac{\phi}{\phi_0}, \quad \bar{k} = \frac{k}{k_0}, \quad \bar{p} = \frac{p}{p_{\text{ref}}}$$

and henceforth omitting the bar over the dimensionless variables the differential equations in (3) retain their appearance, at the same time, dimensionless averaged reaction rate, scaled porosity and permeability are given by

$$r(t) = \frac{1}{\tau} \int_0^t \hat{r}(\xi) \exp\left(\frac{\xi - t}{\tau}\right) d\xi, \quad \hat{r} = \text{Da } m S c, \quad \text{Da} = t_{\text{ref}} / \tau_{\text{chem}}, \quad (5)$$

$$\phi = 1 + (m_0 - m) / \rho_c, \quad k = \phi^\lambda.$$

The solution to the problem depends on the parameter $\text{Da} = \phi_0 \mu_{\text{ref}} L^2 / (k_0 p_{\text{ref}} \tau_{\text{chem}})$, boundary conditions and initial data. The initial data and boundary conditions are

$$S(0, x) = 0, \quad S_m(0, x) = 1, \quad c(0, x) = 0, \quad m(0, x) = m_0, \quad p(0, x) = 0 \quad \text{at } 0 < x < L$$

$$S(t, 0) = 1, \quad S_m(t, 0) = 0, \quad c(t, 0) = c_0, \quad m(t, 0) = 0, \quad p(t, 0) = 1, \quad p(t, L) = 0 \quad \text{at } t > 0.$$

The following values of dimensionless parameters were used in the calculations

$$\text{Da} = 20, \quad c_0 = 0.5, \quad m_0 = 1, \quad \mu_g = 0.1, \quad \rho_g = 2 \times 10^{-3} (1 + 0.1p), \quad \mu_m = 1, \quad \rho_c = 2$$

$$\phi_0 = 1, \quad k_0 = 1, \quad \zeta_A = \zeta_B = \zeta_G = \zeta_W = 1.$$

It should be noted here, that taking into account conditions of the experiment [13] (namely, the length of the cell, $L = 0.35$ m, the driving pressure, $p_{\text{ref}} = 10^4$ Pa, the diameter of glass spheres, $d \approx 3 \times 10^{-4}$ m, the absolute permeability given by the Kozeny–Carman equation [14], $k_0 = d^2 \phi_0^3 / (180(1 - \phi_0)^2) \approx 0.33 \times 10^{-10}$ m², where the porosity is $\phi_0 \approx 0.3$) one can estimate the reference time $t_{\text{ref}} = \phi_0 \mu_{\text{ref}} L^2 / (k_0 p_{\text{ref}}) \approx 120$ s. The inverse dimensionless period of the oscillations is estimated as $t_{\text{ref}} / 6 \approx 20$. The characteristic time of the volume averaged reaction τ_{chem} has not been determined experimentally; however, numerical calculations have shown that it has the same order of magnitude as the period of the oscillations. The order of magnitude of the Peclet number is $\text{Pe} = (WL) / (\phi_0 D) = (k_0 p_{\text{ref}}) / (\mu \phi_0 D) \approx 10^7$, where the diffusion coefficient of chemical components in the water is estimated as $D \approx 10^{-5}$ cm²/s. This justifies the absence of the diffusion terms in transport equations; nevertheless, both the diffusion and the capillary forces can affect averaged reaction kinetics. The characteristic time of the diffusion in the pore scale is $d^2 / D \approx 10^2$ s. This value is much greater than the observed period of the oscillations

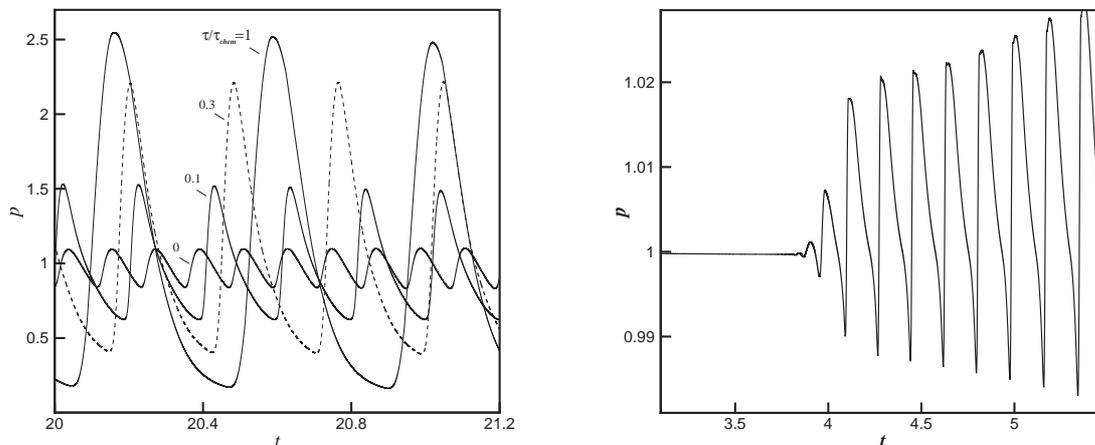


Figure 2. The pressure at the reaction front versus time: left panel—dependence on the time lag τ/τ_{ref} ; right panel—spontaneous transition to the oscillatory mode.

(≈ 6 s). The average size of NaHCO_3 particles ($70 \mu\text{m}$) is considerably less than the particle of glass ($300 \mu\text{m}$), in the same time, such particles cannot migrate freely in the porous space. In the considered model the migration of NaHCO_3 was neglected (it was assumed to be a part of the skeleton). In the experiment the oscillatory mode has been observed particularly at the mole fraction of acid in the solution equal 0.5 and the mass fraction of the sodium bicarbonate in the skeleton 0.1. Thus, the total volume occupied by NaHCO_3 is significantly smaller than the volume of the porous space. The initial value of the saturation S in the experiment was negligible; however, it rapidly became nonzero due to propagation of the displacement fronts, which propagate much faster than the reaction zone. The governing equations have been integrated numerically using unconditionally stable fully implicit scheme. The first-order accurate scheme includes upwind approximation of phase mobilities and concentration at the cell faces. Numerical tests have shown that computational grids with about 10^3 cells provide grid convergent solutions to the considered problem with good accuracy.

3. Numerical results

The calculations have demonstrated the instability of the considered multiphase reactive flow accompanied by spontaneous transition to self-oscillating mode, which is characterized by oscillations of pressure, phase saturations and a rate of the reaction with well-defined frequency (figure 2, left panel). Depending on the parameter values, the oscillations can start at the beginning of the displacement process or the transition can occur at some point in time, see figure 2 (right panel), where pressure-time dependence indicates spontaneous transition to the oscillatory mode. The reaction wave propagates in a stable manner when concentrations of active components or Da number are sufficiently small. The similar behavior was observed in the experiment [13].

In figure 3 phase saturations, concentration of the active component (A), and averaged density of the component B are shown at two points in time, before (left panel) and after transition to the oscillatory mode (right panel). In the latter case, the evolution of the saturation S during one period of the oscillations is presented. The saturation of the resident phase (oil) decreases rapidly due to displacement by the reaction products in gas and liquid phases (figure 3). However, the

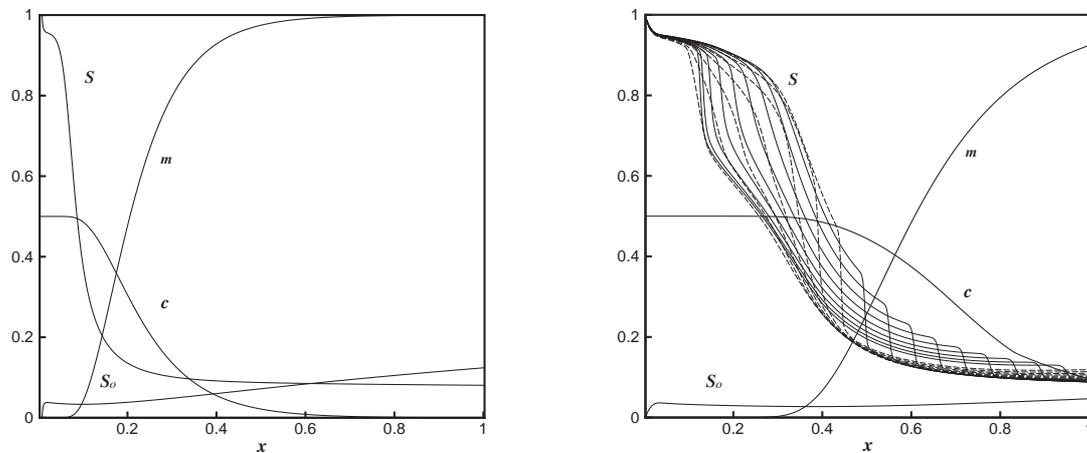


Figure 3. Multiphase reactive displacement in a porous block with chemically active skeleton: the distribution of saturations S , S_o , concentration c , averaged density of the active component of the skeleton m are shown at some time before (left panel) and after transition to the self-oscillating mode (right panel).

residual oil can limit the rate of the reaction. The case represented in figure 3 corresponds to $\tau = 0$.

A series of calculations has been performed with different kinetics ($\tau/\tau_{\text{chem}} = 1; 0.3; 0.1; 0$) to study influence of the time delay between the change of the concentrations and the rate of the reaction. Figure 3 (right panel) demonstrates time dependence of the reaction rate according to the kinetics (1-3) for spatially uniform case with initial reaction rate $r(0) = 0$. The results of numerical solution to the reactive displacement problem are presented in figure 4 (left panel), where the upper and lower envelope functions outlining the extremes of oscillating front pressure are shown for the cases under consideration. The front pressure is defined as pressure at the point, where the concentration of active component of the skeleton, m , equals to half of its initial value m_0 . The area between the lower and upper envelope curves is shaded for the case $\tau/\tau_{\text{chem}} = 0$. The comparison of the curves for different cases shows that the amplitude of the oscillations increases with growth of τ .

The effect of retarded kinetics appears to be significant even in the case when the time delay τ is small in comparison with the characteristic time of the reaction τ_{chem} . This is demonstrated by the difference between calculations for $\tau/\tau_{\text{chem}} = 0.1$ and 0 in the left panels in figure 4. It should be noted that the average speed of propagation of the reaction wave is higher in the self-oscillating mode and increases with τ .

The qualitative behavior of the problem solutions is as follows.

- The period of the oscillations is on the order of characteristic time of the reaction τ_{chem} . The dependence of the period on τ is illustrated in figure 2, left panel.
- The width of the reaction zone increases, on average, with time. It is subject to oscillations in the oscillatory regime. In figure 2, the width of the reaction zone can be defined as the region, where S , c and m are not equal to 0 simultaneously.
- Both the period of oscillations and the width of the reaction zone are found to be decreasing functions of Da , all other parameters being equal.

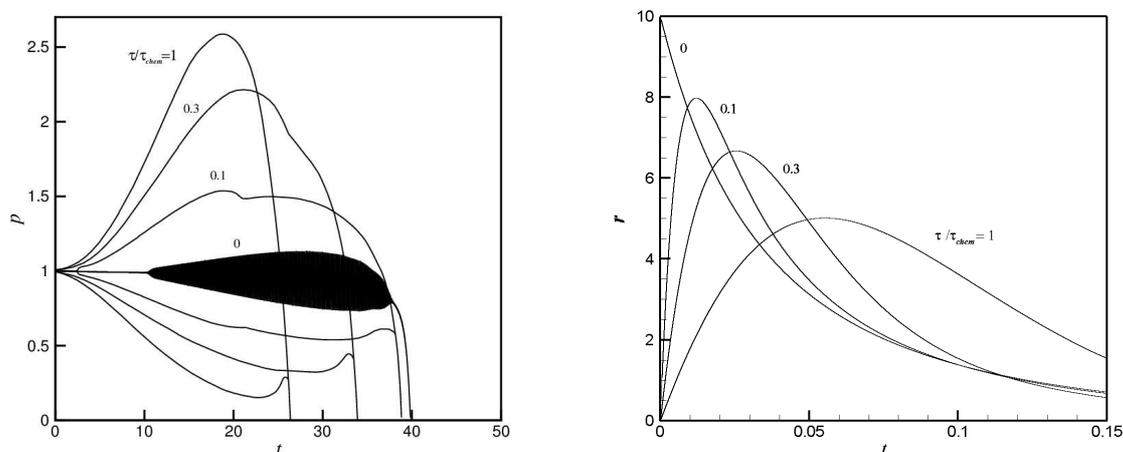


Figure 4. Effect of time lag τ/τ_{ref} on the oscillations (for the cases presented in figure 2): envelope curves of pressure oscillations at the front of the reaction (left panel) and the time dependence of the reaction rate (right panel, arbitrary units).

- If $\tau = 0$ and another dimensionless parameters of the problem are fixed, then there is a critical value of Da , such that if Da number is less than this critical value, the reaction wave propagates in a stable manner (without oscillations).
- The oscillations also take place in the case when the gas phase is assumed to be incompressible ($\rho_g = \text{const}$ in (3)). In this case the pressure equation for the considered system changes its type.
- The oscillating mode can occur both in converging and in diverging geometry, when problem is formulated in a cylindrical layer.

Thus the results of the calculations confirm appearance of self-oscillating mode of the reaction wave observed in [13] and demonstrate strong destabilizing effect of the retarded kinetics.

4. Discussion

In the previous section we have demonstrated that transition to self-oscillatory mode, which occurs in the process of propagation of the reaction front in the multiphase flow through the porous layer with chemically active skeleton, is stimulated by the presence of the time lag between the change of concentration of the reactants and the change of the reaction rate. The reaction rate averaged over the volume of porous medium depends on area of contact between active liquid and the skeleton. In multiphase flow at given saturations, this area is governed by capillary forces. There is a flux of the acid (from dark to light zones in figure 5) driven by capillary pressure gradient, which is caused by the gradient of the wetting phase saturation.

The average rate of the chemical reaction is suggested to be determined by capillary non-equilibrium, which is related to finite time of redistribution of phases in the pore channels under influence of capillary forces at constant saturations. For the sake of simplicity we consider slow reactions, which means that the condition $\tau \ll \tau_{\text{chem}}$ takes place, where τ is the characteristic time of capillary relaxation. Several approaches have been proposed to model capillary non-equilibrium effects in multiphase flow in porous media [15, 16], [17]). Consider evolution of the contact area using the thermodynamically consistent model [17]. In the framework of this



Figure 5. Zoomed-in photograph of the reaction front in experiment [13]: dark zones correspond to acid solution; the products of the reaction (gas phase) are distributed in light-colored regions.

approach the thermodynamic potential being a function of inner parameter of the system, which parameterizes capillary non-equilibrium states, is introduced. The kinetic equation for the non-equilibrium parameter is formulated in a thermodynamically consistent way. In the particular case of non-deformable, isothermal medium with non-deformable skeleton the potential is simply the surface energy of the system, which depends on saturation and the non-equilibrium parameter

$$\Psi = \Psi(s, \xi), \quad \frac{\partial \xi}{\partial t} = \Lambda(s, \xi). \quad (6)$$

Here, s is the saturation of the skeleton by the non-wetting fluid saturation; ξ is the nonequilibrium parameter. All properties, which depend on distribution of phases in pore channels, such as phase permeability, capillary pressure, etc., are functions of the phase saturation and of ξ . Consider the special case, such that the rate of change of the non-equilibrium parameter is proportional to the derivative of the potential of the skeleton with respect to this parameter. Hence, the dissipation is non-negative in an arbitrary process

$$\Lambda(s, \xi) = -\frac{1}{\tau(s, \xi)} \frac{\partial \Psi}{\partial \xi}, \quad \delta = -\Lambda(s, \xi) \frac{\partial \Psi}{\partial \xi} = \tau^{-1} \left(\frac{\partial \Psi}{\partial \xi} \right)^2 \geq 0. \quad (7)$$

The time of relaxation τ can be obtained from experiment on relaxation of capillary pressure. For the sake of simplicity it assumed to be constant.

Following [17], consider the thermodynamic potential

$$\Psi(s, \xi) = \frac{1}{2} \xi^2 + \frac{1}{2} M s^2 - \alpha \xi s + \gamma s, \quad s = 1 - S, \quad (8)$$

where $\alpha, \gamma, M > 0$ are constants. It was shown that this definition enables to simulate basic capillary effects in a porous medium saturated by two fluids. Substitution to (6) gives the following equation for ξ :

$$\frac{\partial \xi}{\partial t} = \frac{1}{\tau} (\alpha s - \xi). \quad (9)$$

Thus, the equilibrium values of this parameter and of the potential are

$$\xi^e(s) = \alpha s, \quad \Psi^e(s) = \Psi(s, \xi^e) = \frac{1}{2} (M - \alpha^2) s^2 + \gamma s, \quad s = 1 - S. \quad (10)$$

It is naturally to suppose, that the area of contact between the non-wetting fluid and the skeleton is proportional to the surface energy of contact between them, $\sigma^{\text{nw}} = \lambda \Psi$. The energy of the surfaces, separating the fluids, is neglected, since usually the area of these surfaces is small compared with the area of contact of the fluids with the skeleton. Taking into account this definition, it follows from (8-10) that evolution of the contact area at $s = \text{const}$ from some non-equilibrium state is given by the relaxation equation

$$\frac{\partial \sigma^{\text{nw}}}{\partial t} = \frac{2}{\tau} (\lambda \Psi^e(s) - \sigma^{\text{nw}}), \quad (11)$$

and, hence, for the wetting phase

$$\frac{\partial \sigma^w}{\partial t} = \frac{2}{\tau} (\sigma^{\text{we}} - \sigma^w). \quad (12)$$

The rate of slow ($\tau \ll \tau_{\text{chem}}$) reaction is proportional to the product of the area of contact between the active fluid and skeleton, $m^{(s)} \sigma^w$, where $m^{(s)}$ is surface fraction of the active component of skeleton, and concentration $c^{(s)}$ of the reactant in fluid near the surface

$$r(t) \sim \frac{1}{\tau} \int_0^t m^{(s)}(\xi) c^{(s)}(\xi) \sigma^{\text{we}}(S) e^{(\xi-t)/\tau} d\xi. \quad (13)$$

Initial condition $\sigma^w(0) = 0$ is used. The kinetics (5) is particular case of this expression, provided that the surface values are proportional to the bulk concentrations $m^{(s)} \sim m$, $c^{(s)} \sim c$ and $\sigma^{\text{we}}(S) \sim S$. It has been shown (figure 3) that such kinetics has strong destabilizing influence and stimulates transition to the oscillatory mode. There are other reasons for the time lag to occur. It may be caused, for example, by multistage character of reaction kinetics or by diffusion. The considered mechanism can manifest itself in highly non-uniform media under conditions of strong capillary non-equilibrium.

5. Conclusion

The immiscible displacement process such that the displacing fluid and the skeleton of the porous medium have chemically active components which react with production of gaseous phase is studied. The redox chemical reaction accompanied by production of gaseous phase is considered. The calculations have demonstrated strong influence of the reaction kinetics on stability of the reactive flow. The presence of a time lag between change of concentration of the reactants and the change of the reaction rate is shown to stimulate transition of the reaction front propagation to the oscillatory mode. Capillary non-equilibrium of the reactive displacement provides possible explanation of this kind of kinetics.

Acknowledgments

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