

Lattice Boltzmann Simulation on Reactive Transport of Calcite Dissolution by Injecting CO₂-saturated Brine during Sequestration

Qiheng Xie ^{a,b}, Wendong Wang ^{a,b}, Yuliang Su ^{a,b}, Sina Rezaei-Gomari ^c, Han Wang ^{a,b},

Zhouyuan Zhang ^{a,b}, Wubin Yan ^{a,b}

^a Key Laboratory of Unconventional Oil & Gas Development (China University of Petroleum (East China)), Ministry of Education, Qingdao, 266580, PR China

^b School of Petroleum Engineering, China University of Petroleum (East China), Qingdao, 266580, PR China

^c School of Computing, Engineering and Digital Technologies, Teesside University, Middlesbrough TS1 3BX, UK



Introduction

CO₂ as a kind of greenhouse gas has a greatly adverse impact on the environment, and fixing carbon into saline aquifers effectively alleviates the effect it brings. In the present study, the calcite dissolution during sequestration near wellbores at different temperatures is explored to elucidate the mesoscale mechanism of reaction transport during this process and the evaluation of hydraulic properties with time that affects the injectivity of CO₂-saturated brine.

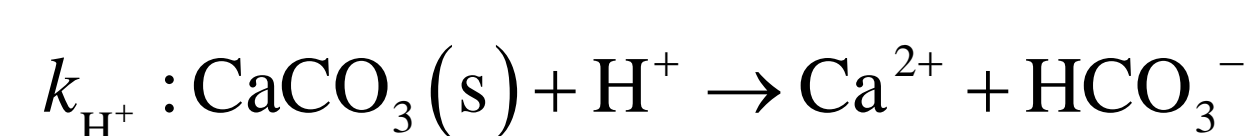
A multi-component lattice Boltzmann method coupled fluid flow, mass transport, heterogeneous reaction, and structure evolution is proposed to quantitatively study the reaction transport of calcite dissolution near wellbores. A special solution included the general lattice Boltzmann concentration boundary condition and Volume of Pixel method is utilized to model the dissolution reaction. The influence of temperature on reaction dynamics in porous media is analyzed by exerting the Arrhenius expression, and the concentration distribution of H⁺ and Ca²⁺ is captured to reflect the dissolution front directly. Moreover, the evaluated hydraulic properties characterize the effect of the dissolution reaction on the brine injectivity.

Since the reaction rate of calcite dissolution increases with temperature, the dissolution node of calcite is proportional to the temperature at a constant pressure difference. And the dissolution node indicating the released volume of Ca²⁺ which is a key reactant for the mineral trapping is summed to estimate the sequestration result. Furthermore, the dissolution front observed from the pore-scale simulation is heterogeneous due to a higher velocity, and the dissolution pattern of all cases is identified as the wormholing dissolution, which means that the increasing porosity during the calcite dissolution has a great contribution to the permeability. Admittedly, the increasing temperature during sequestration is conducive to calcite dissolution and changes in hydraulic properties. For sequestration operation, the high-temperature formation is recommended for both the injectivity and the subsequent mineral trapping.

This work provides theoretical and pragmatic guidance to operators to design CO₂-saturated brine injection in saline aquifers. Furthermore, the results can also promote cognition of the calcite dissolution for sequestration at different temperatures.

Method

In this contribution, calcite dissolution near wellbore is investigated via multi-component lattice Boltzmann method. A single irreversible heterogeneous reaction is used according to the following chemical equation (Molins et al., 2020)



and the reaction rate of calcite dissolution can be described as

$$I_{H^+} = -k_{H^+} a_{H^+} = -k_{H^+} \gamma_{H^+} C_{H^+}$$

Based on the simple and popular Bhatnagar-Gross-Krook (BGK) collision operator (Bhatnagar et al., 1954), the single-phase flow in the D2Q9 model is depicted by the evolution function of the density distribution equation

$$f_i(\mathbf{x} + \mathbf{e}_i \Delta t, t + \Delta t) - f_i(\mathbf{x}, t) = -\frac{1}{\tau_\sigma} [f_i(\mathbf{x}, t) - f_i^{\text{eq}}(\mathbf{x}, t)]$$

The mass transport of H⁺ and Ca²⁺ can be described by the D2Q5 model. The corresponding evolution function of the concentration distribution equation is written as (Sullivan et al., 2005)

$$g_{k,i}(\mathbf{x} + \mathbf{e}_i \Delta t, t + \Delta t) - g_{k,i}(\mathbf{x}, t) = -\frac{1}{\tau_{k,g}} [g_{k,i}(\mathbf{x}, t) - g_{k,i}^{\text{eq}}(\mathbf{x}, t)]$$

In this work, heterogeneous reactions are solved by the lattice Boltzmann concentration boundary condition (Chen et al. (2013)) to solve reactions between CO₂-saturated brine and calcite (CaCO₃). The stationary solid-liquid concentration boundary condition of each component for the calcite dissolution reaction can be described by

$$D \frac{C_R - C_F}{c} = I_{H^+}$$

$$g_{H^+,2} - g_{H^+,4} = -\frac{D_{H^+}}{c} \frac{\partial C_{H^+}}{\partial y}$$

$$g_{\text{Ca}^{2+},2} - g_{\text{Ca}^{2+},4} = \frac{D_{\text{Ca}^{2+}}}{c} \frac{\partial C_{\text{Ca}^{2+}}}{\partial y}$$

The volume of pixel method (Kang et al. (2002)) is utilized to update solid nodes during heterogeneous reactions. The calcite volume fraction is updated at every time step

$$V_s(t + \Delta t) = V_s(t) + V_m I_{H^+} \Delta t A$$

Results

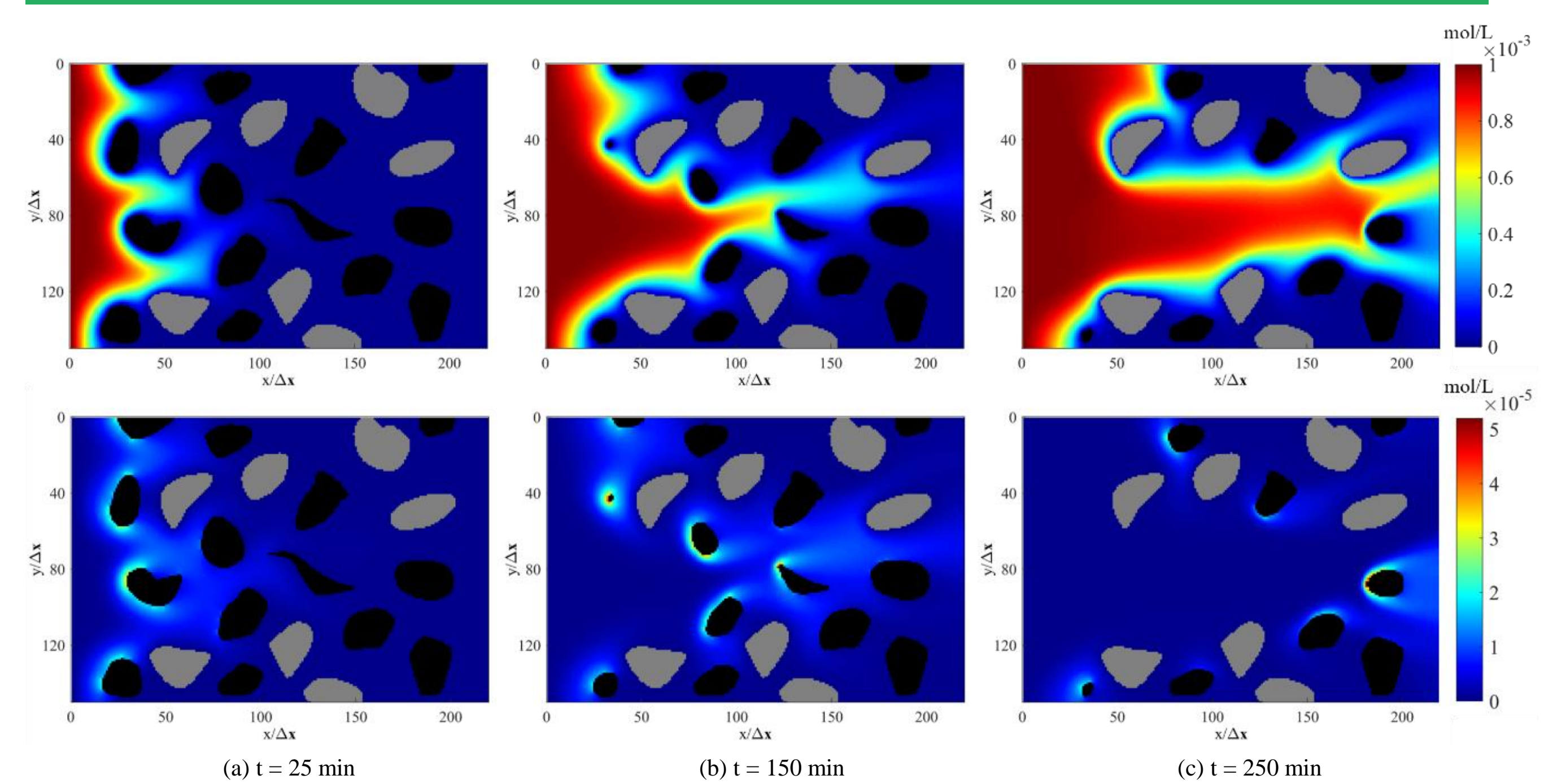


Fig. 1 Temporal evolution of H⁺ concentration (top) and Ca²⁺ concentration (bottom) during the reaction under the high *Pe* condition at 85 °C.

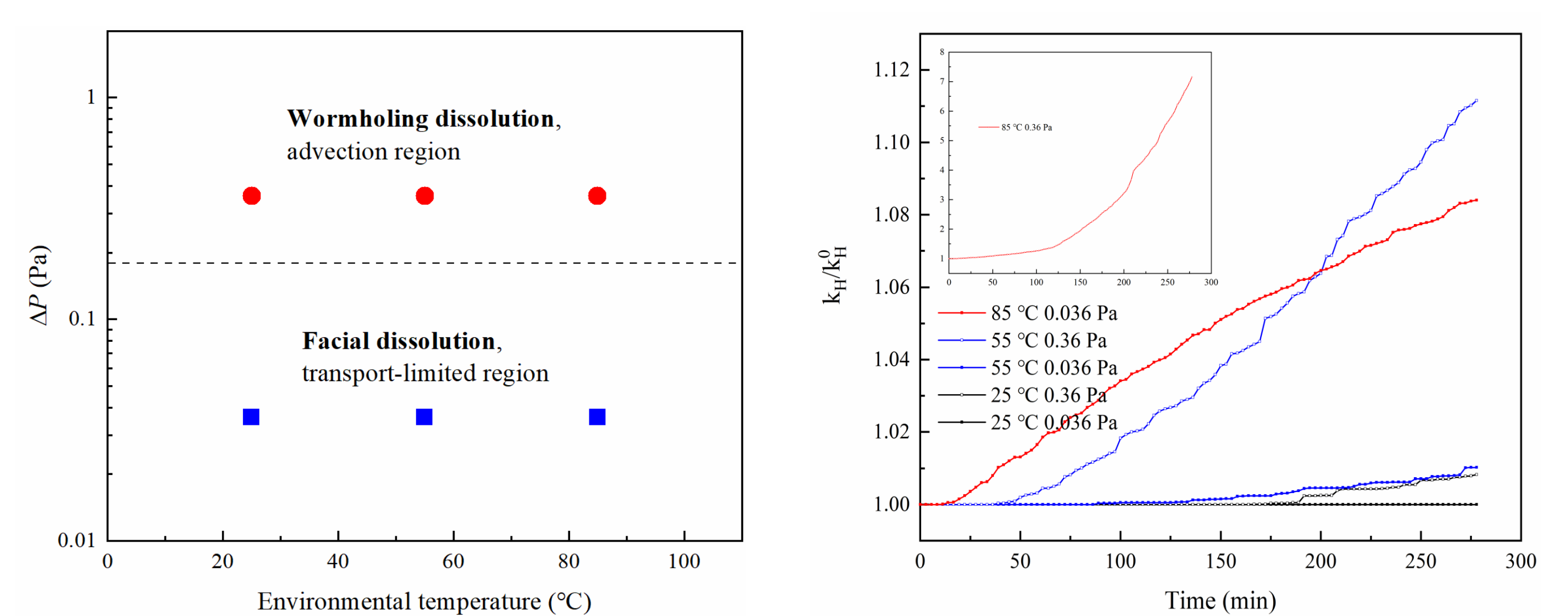


Fig. 2 Regime diagram of calcite dissolution in different cases.

Fig. 3 Permeability and porosity relationship in different cases.

Reference

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Relevant work

Interested one can refer to our recent work for details. (<https://doi.org/10.1016/j.jgsce.2023.204978>)