Impact of Physical Heterogeneity on Effective Reaction Rates

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Abstract

Reactive transport in porous media is of key importance in contaminant hydrology, carbon storage, enhanced oil recovery and chemical reactors engineering. Despite progress made in recent decades, measurement and prediction of the effective reaction rates is still not well understood. In this study we show how flow and transport heterogeneity affects the effective reaction rate. Our approach is based on a reactive transport particle tracking model at the continuum scale, in contrast to the pore-scale models which were successfully used to study the impact of heterogeneity on non-Fickian transport (Bijeljic et al., 2011, 2013) and reactive transport (Pereira Nunes et al., 2016). We make use of our reactive continuous time random walk (CTRW) model that was previously validated using Nuclear Magnetic Resonance (NMR) experimental measurements during dissolution of a Ketton carbonate rock core sample (Oliveira et al., 2021). The CTRW model is defined using a truncated power-law distribution of transit-time, which contains diffusive time cut-off, mean advective time, and a parameter β characterising the domains unresolved physical heterogeneities. To systematically study the effects of flow and transport heterogeneities on the effective reaction rates, we construct three domains with increasing physical and transport heterogeneity, and subject each domain to three different advective regimes with $Pe = 20$, 200 and 2000. This strategy allowed us to examine nine initial states. For transport, we characterize signatures of physical heterogeneity in the three porous media using velocity distributions and show how these imprints on the signatures of particle displacement, namely particle propagators distributions. In addition, we demonstrate the ability of our CTRW model to capture the impact of physical heterogeneity on the longitudinal dispersion coefficient over several orders of magnitude for a wide range of Pe defining transport regimes. Reactive transport simulations indicate that the effective reaction rates depend on (i) initial physical heterogeneity and (ii) transport conditions. We show that the higher the initial heterogeneity, the lower the reaction rate. Finally, a decrease in Pe would promote mixing by diffusion over advection, resulting in the higher reaction rates. Overall, we establish a framework to demonstrate and quantify the impact of physical heterogeneity on transport and effective reaction rates in porous media.

keywords — Continuous Time Random Walks, Physical Heterogeneity, Reactive Transport Modelling, Effective Reaction Rates

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