

Nanogeochemistry of Gas Disposition and Release in Shale Matrix

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Shale is characterized by the predominant presence of nanometer-scale (1-100 nm) pores. The behavior of fluids in those pores directly controls shale gas storage and release in shale matrix and ultimately the wellbore production in unconventional reservoirs. Recently, it has been recognized that a fluid confined in nanopores can behave dramatically differently from the corresponding bulk phase due to nanopore confinement. CO₂ and H₂O, either preexisting or introduced, are two major components that coexist with shale gas (predominately CH₄) during hydrofracturing and gas extraction. Liquid or supercritical CO₂ has been suggested as an alternative fluid for subsurface fracturing such that CO₂ enhanced gas recovery can also serve as a CO₂ sequestration process. Limited data indicate that CO₂ may preferentially adsorb in nanopores (particularly those in kerogen) and therefore displace CH₄ in shale. Similarly, the presence of water moisture seems able to displace or trap CH₄ in shale matrix. Therefore, fundamental understanding of CH₄-CO₂-H₂O behavior and their interactions in shale nanopores is of great importance for gas production and the related CO₂ sequestration. The work presented here focuses on the systematic study of CH₄-CO₂-H₂O interactions in shale nanopores under high-pressure and high temperature reservoir conditions. Using molecular simulations, we show that methane release in nanoporous kerogen matrix is characterized by fast release of pressurized free gas (accounting for ~ 30 - 47% recovery) followed by slow release of adsorbed gas as the gas pressure decreases. The first stage is driven by the gas pressure gradient while the second stage is controlled by gas desorption and diffusion. We further show that diffusion of all methane in nanoporous kerogen behaves differently from the bulk phase, with much smaller diffusion coefficients. The MD simulations also indicate that a significant fraction (3 - 35%) of methane deposited in kerogen can potentially become trapped in isolated nanopores and thus not recoverable. Our results shed a new light on mechanistic understanding gas release and production decline in unconventional reservoirs. The long-term production decline appears controlled by the second stage of gas release. The MD simulations also show that nanoporous kerogen tends to adsorb CO₂ preferentially over CH₄ and H₂O sorption is minimal.

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