



On the shale-fluid reactions occurred in the hydraulic fracturing process of shale gas development: insights from lab simulation

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Hydraulic fracturing is a widely used technique for oil and gas extraction from the ultra-low porosity and permeability shale reservoirs. By injecting a large amount of fracturing fluids with specific chemical additives into shale reservoirs, porosity and permeability can be significantly improved, and thus enhances the recovery of oil and gas. However, hydraulic fracturing will not only bring economic benefits, but also cause a series of environmental issues, e.g., earth surface and water pollution. The Niutitang shale characterized by wide distribution, great thickness, high organic matter and brittle mineral contents is one of primary targets of shale gas development in South China. This research aims to simulate rock-fluid reaction and investigate the evolution of fluid composition and shale characteristics occurred during the hydraulic fracturing of the Niutitang shale.

Two sets of shale samples from different depths of a well located in Central Hunan Province were exposed to fracturing fluids with different pH values. Afterwards, changes in the fluids and the shale matrix were investigated through a series of geochemical, mineralogical, and textural analysis. By comparing results of different experiments at different scales, key geochemical rock-fluid reactions occurred during this process were found, and their potential influences on shale gas production were discussed.

Experimental results show obvious mineral dissolution, in particular the oxidation of pyrite. Pyrite oxidation significantly alters fracturing fluids and subsequently impacts on the dissolution of other minerals. During the process of pyrite dissolution, hydrogen ions release into the fluids, leading to obvious acidification. The acidified solution dissolves carbonate and feldspar minerals. Meanwhile, in the process of mineral dissolution, heavy metals or radioactive elements release as well, e.g., Ba, U, and Sr, which are all primary toxic elements of flowback fracturing fluids. The interaction between shale and fracturing fluid also causes changes in the shale matrix. Comparisons between shale samples before and after experiments clearly show density decrease while porosity increase. In addition, pore types change from ink bottle-shaped thin neck hole dominance to long and narrow plate-shaped hole dominance. Through the theoretical calculation of saturation index and observation by scanning electron microscope, we found that mineral dissolution is accompanied by secondary mineral precipitation, e.g., Fe-(oxy) hydroxide and gypsum. These precipitates, nevertheless, could potentially restrict the migration of metal elements by adsorption or co-precipitation, occlude the pore systems, and finally decrease the recovery of shale oil and gas. Overall, we conclude that mineral compositions and physical properties of the shale are among

primary factors controlling fluid-rock reactions. Therefore, mineral composition and textural analysis are critical to fracturing fluid design and important to lowering environmental risks caused by flowback fluids.