

Unravelling Controls on Marcellus Shale Weathering

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The development of Marcellus shale gas has significantly changed the energy landscape while drawn much public attention to potential environmental impacts. Here we develop a reactive transport model for shale–water interactions based on weathering data [1]. The model provides a powerful tool to reproduce the history of soil formation and to unravel key controls on shale weathering. With extension to relevant conditions, this model could be used to forecast reactions and evolution of mineralogical and hydrological properties during gas production at depth. Such prediction will be important for planning water management and addressing questions of societal significance.

The model was developed using the code CrunchFlow starting from measured parent rock composition and by matching soil and water composition profiles over 1.2 meter depth. Results show that the shale went through several major stages. Fast oxidative pyrite dissolution started first and was characterized by a quick loss of sulfur and precipitation of iron hydroxide, with porefluid pH around 5. This stage lasted for about 500 years while porosity increased from the original 5% to 15%. This was followed by stages lasting about 9,500 years that are dominated by chlorite and illite dissolution at much slower rates, continuously increasing the porosity from 15% to about 30% - 40%. Vermiculite precipitated along with the loss of Al, K, and Mg.

Parameter analysis shows that even small amounts of pyrite can produce sufficiently low pH conditions that can accelerate dissolution for a porosity increase. As such, porosity evolution is surprisingly not sensitive to the initial pyrite abundance (varied from 0.5 to 5%). The presence of CO₂ gas in the pore space was important in matching the observed low soil pH and weathering profile. Both solid and aqueous phases were sensitive to the porefluid flow velocity. Values of surface area have to be reduced by 2 orders of magnitude from measured BET values in order to match the data, suggesting much lower rock-fluid contact area that is effectively dissolving [2].

[1] Jin *et al* (2013) *Chem Geol* **356**, 50-63. [2] Li *et al* (2014) *Geochim. Cosmochim Acta*, **126**, 555-573.