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Kinetics of Precipitation of Barium Chromate [$BaCrO_{4(s)}$] in Groundwater

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Water-soluble and carcinogenic hexavalent chromium [Cr(VI)] is often found as a pollutant in aquifers impacted by leaching from anthropogenic Cr-containing solid waste. In the subsurface environment, Cr(VI) has been reported [1] to interact with naturally present co-ions such as barium [Ba] to form relatively insoluble barium chromates [$BaCrO_{4(s)}$]. However, in aquifers with elevated levels of SO_4^{2-}, barite [$BaSO_{4(s)}$] precipitation might outcompete the precipitation of $BaCrO_{4(s)}$, as suggested by their relative thermodynamic stabilities. Although rapid precipitation of both $BaCrO_{4(s)}$ and barite in groundwater matrix has been reported previously [1], the relative rates of formation of these two solids have not been quantified. In this contribution, the relative precipitation of these solids was systematically investigated in a typical central Indo-Gangetic plain groundwater through continuous flow stirred tank reactor (CFSTR) experiments. Cr(VI)-free Ba-containing groundwater was introduced into a CFSTR, preloaded with Ba-free, Cr(VI)-spiked groundwater at a flow rate of 0.2 mL min^{-1}. Time-dependent concentration profiles of Cr(VI), SO_4, and Ba and pH were measured and mathematically modelled using component mass balances. The model accurately predicted the experimentally measured concentrations of Cr(VI) (Figure 1a). Geochemical analyses indicated rapid formation of $BaCrO_{4(s)}$ and barite, whose presence was also confirmed by XRD pattern of the precipitated solids (Figure 1b). A rise in pH of 0.6 log-units, and gradual decrease in the saturation indices with respect to $BaCrO_{4(s)}$ was observed after 7 h. High supersaturation of barite throughout the experiment suggested that barite might exert secondary control over $BaCrO_{4(s)}$. The surface area normalized rates of formation of $BaCrO_{4(s)}$ and barite were quantified as 2.61×10^{-4} moles m^{-2} min^{-1} and 7.29×10^{-4} moles m^{-2} min^{-1}, respectively. Even though barite precipitation was faster than of $BaCrO_{4(s)}$, Cr(VI) uptake still occurred which indicated that immobilization of Cr(VI) as $BaCrO_{4(s)}$ could still be achieved at a time-scale suitable for on-field application. These findings have implications for accurate prediction of Cr(VI) fate and transport in groundwater and also for the development of potential in-situ Cr(VI) remediation techniques.

[1] Bhattacharya M., Shriwastav A., Bhole S., Silori, R., Mansfeldt, T., Kretzschmar R., and Singh A. **(2020)** *ACS Earth and Space Chemistry* 4(1), 35-39.

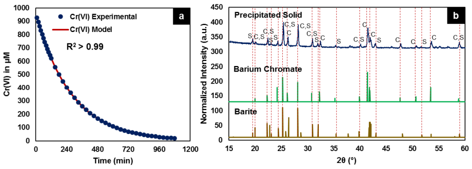

Figure 1. (a) Comparison between the experimentally measured and modelled concentrations of Cr(VI) due to flow of Ba-containing groundwater in the CFSTR; (b) X-Ray Diffraction analysis of solid residues after the CFSTR experiment. The peaks of $BaCrO_{4(s)}$ and barite in the sampled solid have been abbreviated as C and S, respectively. The XRD patterns of barium chromate [$BaCrO_{4(s)}$; PDF 00-035-0642] and barite [$BaSO_{4(s)}$; PDF 01-076-0217] have also been shown with distinct offsets for comparison with the precipitated solid.