

Sorption of oxyanionic contaminants at the barite (001)–water interface[†]

PENG YANG¹, JACQUELYN N. BRACCO², PAUL FENTER¹, ANDREW G. STACK³ AND SANG SOO LEE¹

¹Argonne National Laboratory

²Queens College, City University of New York

³Oak Ridge National Laboratory

Presenting Author: pyang1@anl.gov

The interactions of contaminants with ionically bonded minerals strongly influence the fate and transport of these toxic chemicals in the environment. Because of the dynamically evolving surface chemistry of these minerals, the sorption mechanisms are not fully understood at the molecular scale. Arsenic is a carcinogen while selenium and molybdenum are essential nutrients but become toxic at high dosages. In this study, the sorption of arsenate, selenate, and molybdate on the barite (001) surface was examined at pH ~5 using in situ crystal truncation rod, resonant anomalous X-ray reflectivity, and atomic force microscopy. The results show that arsenate and selenate mainly incorporate into the top monolayers of barite, but arsenate shows a higher coverage. The protonation of hydrogen arsenate at pH 5 may enhance the formation of H-bonds between arsenate and oxygen atoms of neighboring sulfate and hence stabilizes hydrogen arsenate in the barite crystal. In contrast, almost no incorporated molybdate is observed presumably because it is too big to fit in the barite structure. Instead, most sorption of molybdate occurs above the surface presumably as surface-induced oligomers with a broader distribution. The distinct sorption mechanisms of arsenate, selenate, and molybdate to the barite surface manifest that the sizes, protonation states, and polymerization properties of the oxyanions control their interactions with ionically bonded minerals. This study provides important information on the influence of ionically bonded minerals on the fate and transport of contaminants and nutrients.

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