

Electronic Supplementary Information

Different Routes of Bismuth Mineral Transformation during Pertechetate and Perrhenate Uptake for Subsurface Remediation

Boglaienko D.^{*a}, Bowden M.E.^a, Escobedo N.M.^a, Collins Q.M.^a, Lawter A.R.^a, Levitskaia T.G.^{*a}, Pearce C.I.^{*a}

^a Pacific Northwest National Laboratory, 902 Battelle Boulevard, Richland, WA 99354

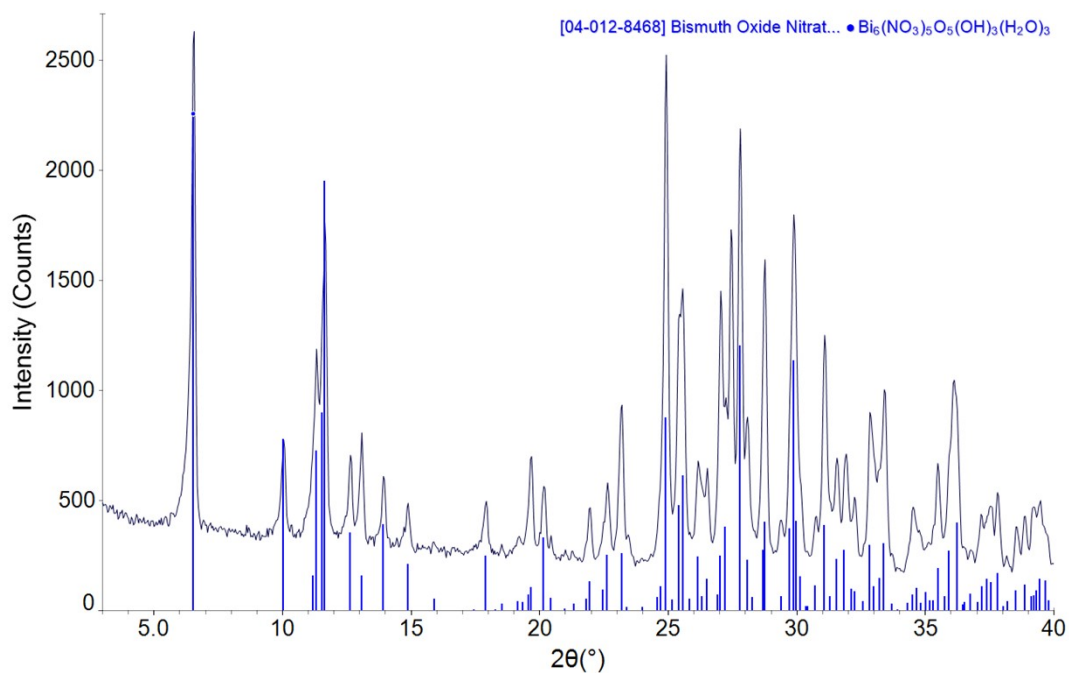


Figure S1. PXRD pattern for BSN starting material overlaid with the pattern for bismuth basic nitrate $[\text{Bi}_6\text{O}_5(\text{OH})_3]^{5+}$ clusters from Lazarini et al. 1978; here denoted as *clus*- $\text{Bi}_{12}\text{O}_{10}(\text{OH})_6(\text{NO}_3)_{10}\cdot 6\text{H}_2\text{O}$.

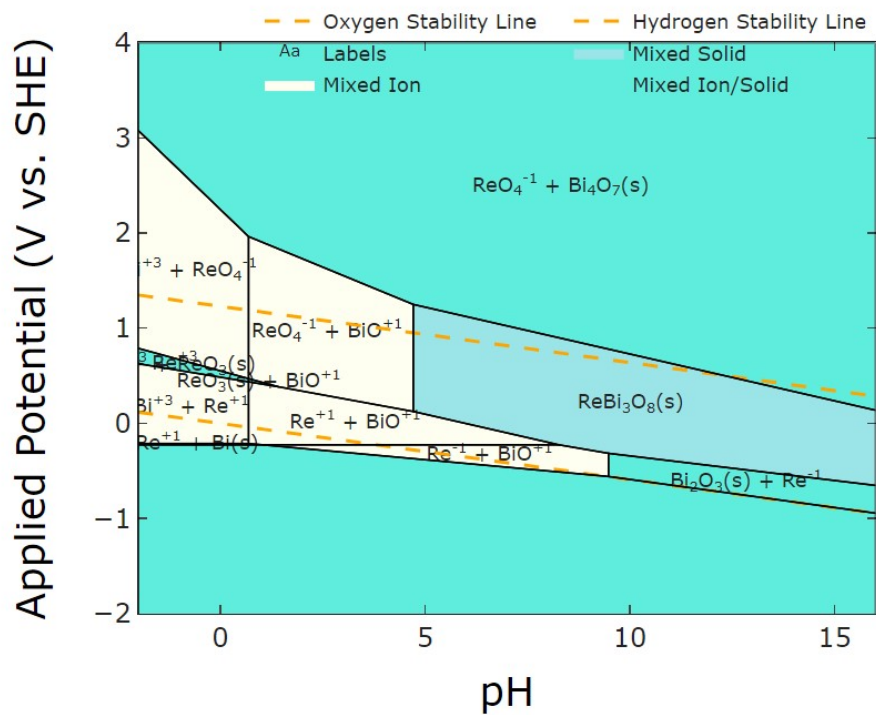


Figure S2. Pourbaix diagram, generated using materials project: Patel et al. (2019).

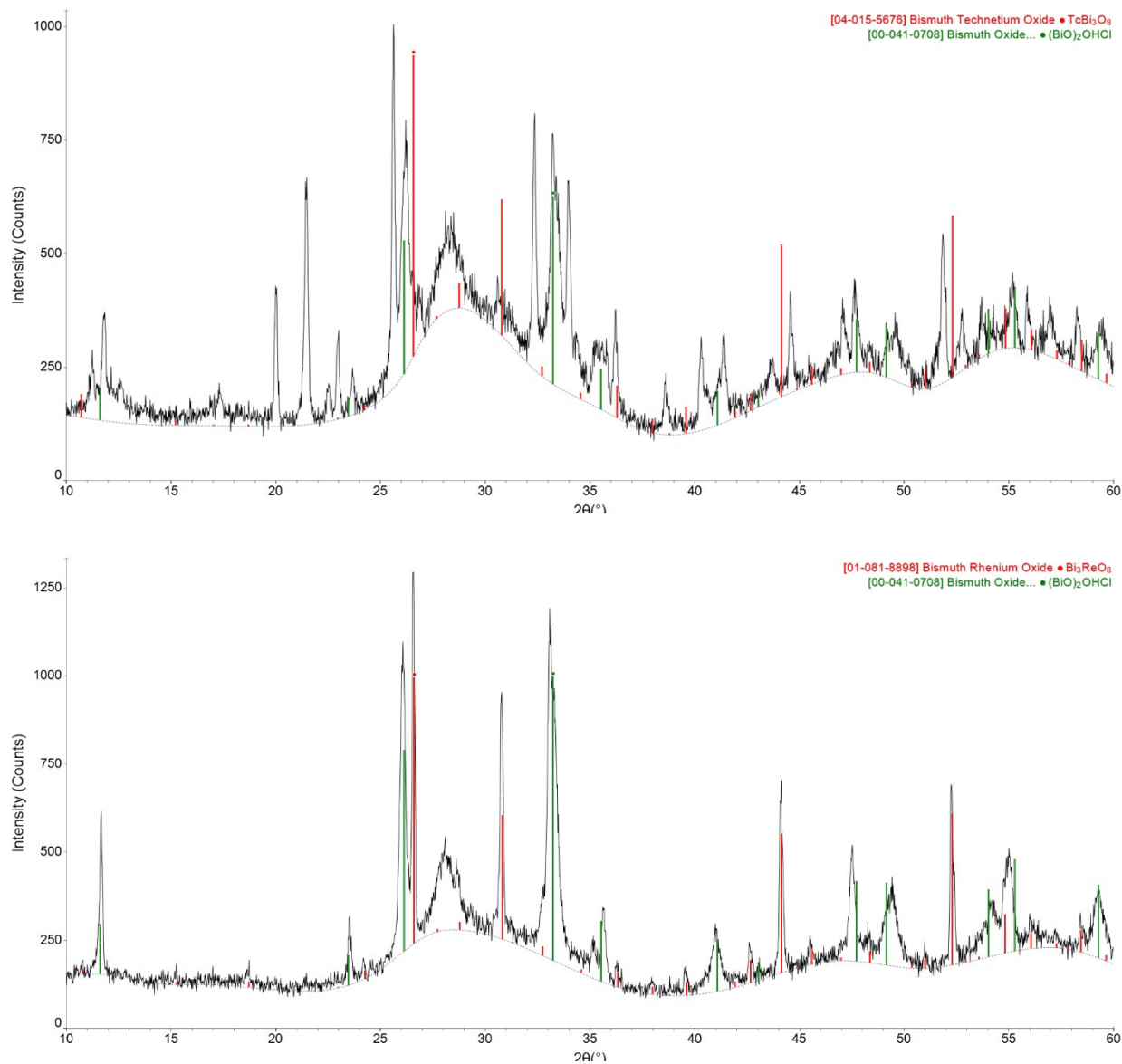


Figure S3. PXRD patterns for BSN with 0.17 mM Tc 24 days (top) and 0.17 mM Re 28 days (bottom) matched with daubreite related structure $(\text{BiO})_2\text{OH,Cl}$ (in green) present in both samples, and demonstration of the absence of Bi_3TcO_8 phase in 0.17 mM Tc sample (top) and the presence of Bi_3ReO_8 phase in 0.17 mM Re sample (bottom).

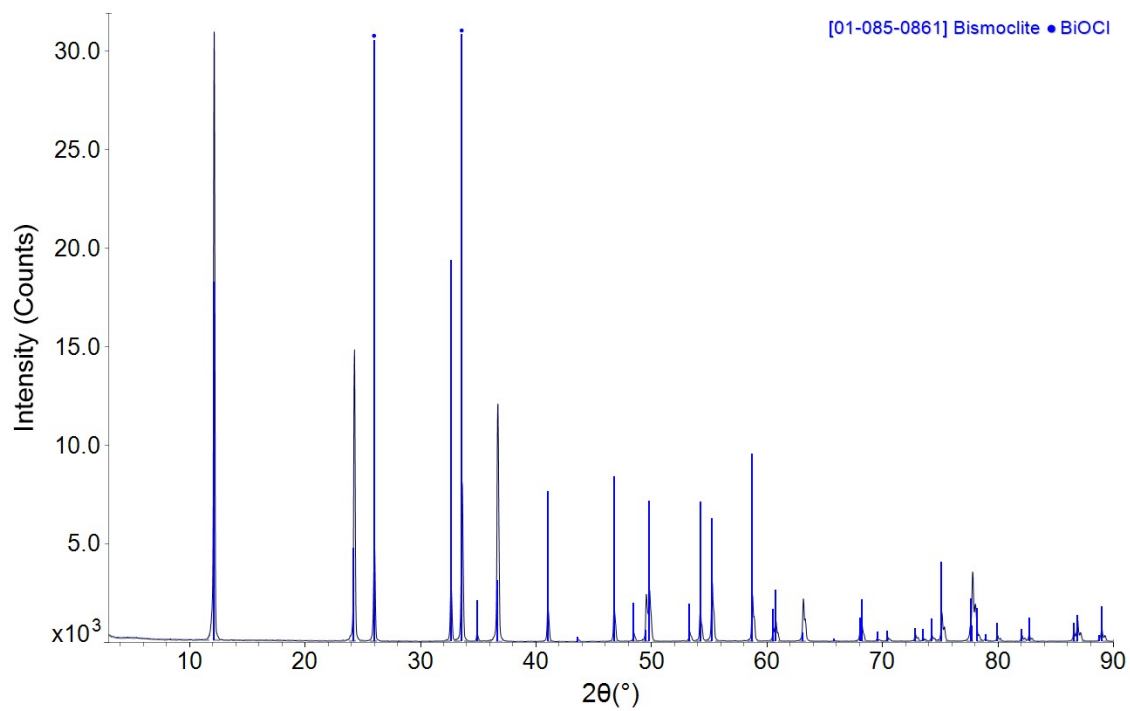
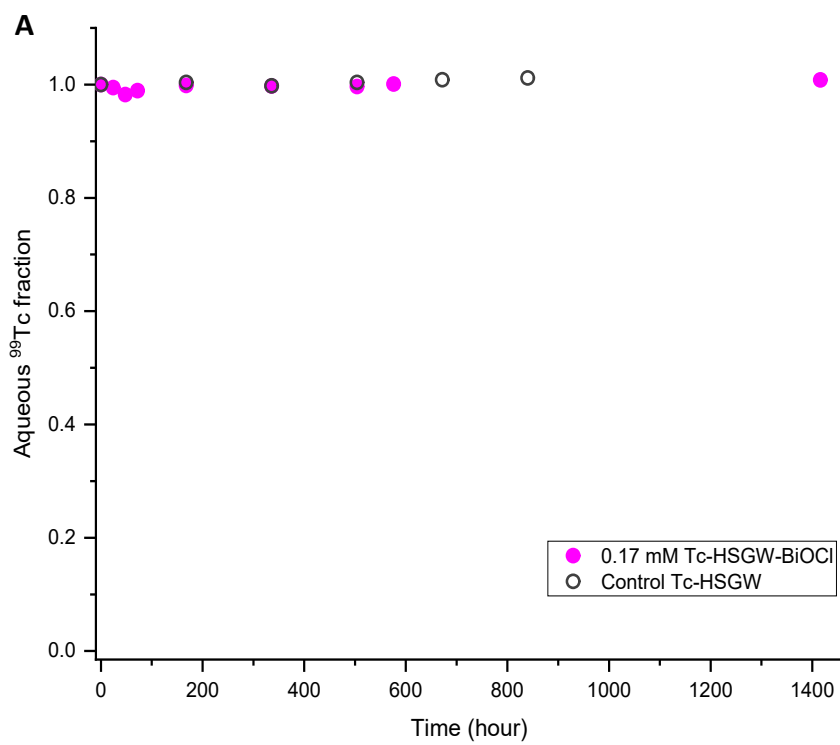


Figure S4. PXRD pattern for pristine BiOCl material.



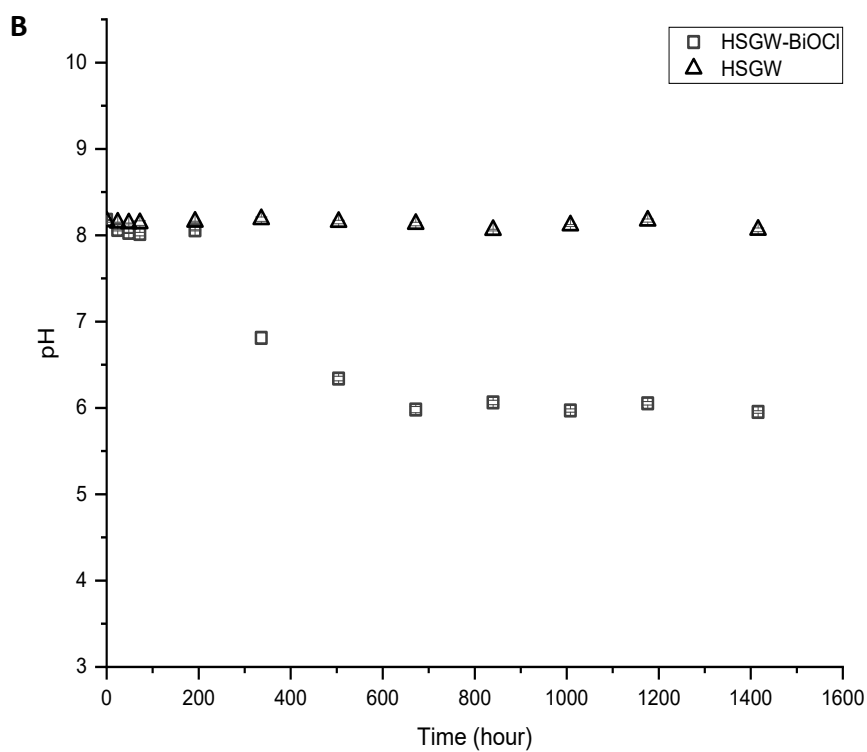


Figure S5. A. Removal of 0.17 mM TcO_4^- with BiOCl starting material (0.1 g) in HSGW simulant (100 mL). Control series: 0.17 mM TcO_4^- in HSGW simulant. **B.** PH measurements for control series: HSGW only and HSGW with BiOCl .

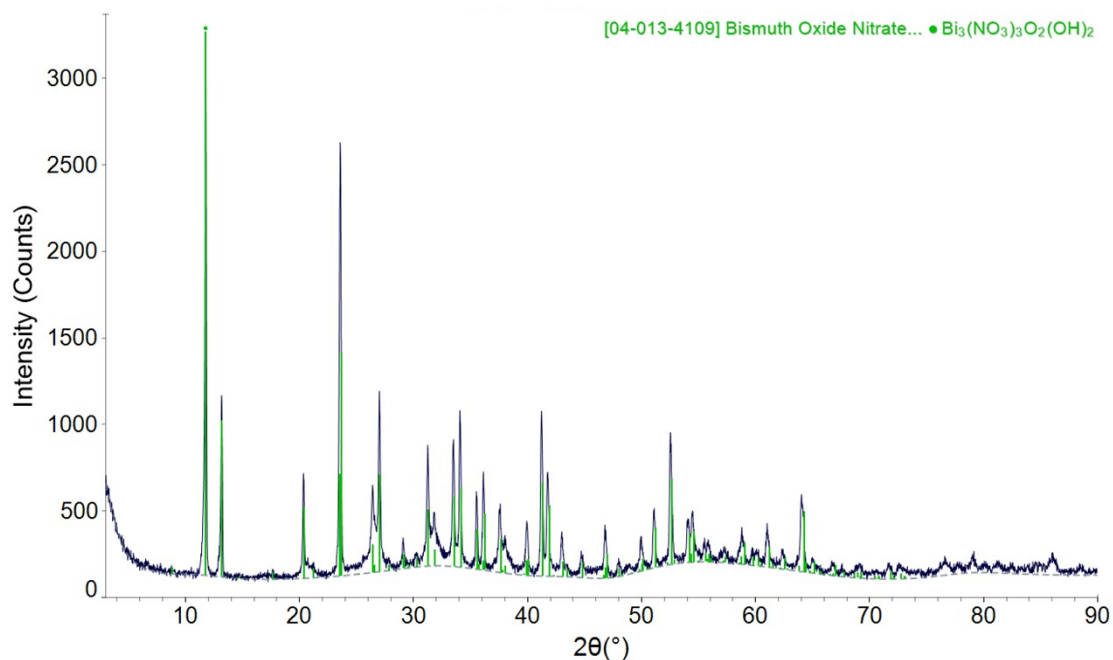


Figure S6. PXRD pattern for *clus*-Bi₆O₄(OH)₄(NO₃)₆ synthesized material (Henry et al., 2006) overlaid with Bi oxide nitrate, Bi₃(NO₃)₃O₂(OH)₂.

References

Lazarini, F., 1978. The crystal structure of a bismuth basic nitrate, [Bi₆O₅(OH)₃](NO₃)₅·3 H₂O. *Acta Crystallographica Section B: Structural Crystallography and Crystal Chemistry*, 34(11), pp.3169-3173.

Patel, A.M., Nørskov, J.K., Persson, K.A. and Montoya, J.H., 2019. Efficient Pourbaix diagrams of many-element compounds. *Physical Chemistry Chemical Physics*, 21(45), pp.25323-25327.

Henry, N., Mentré, O., Abraham, F., MacLean, E.J. and Roussel, P., 2006. Polycationic disorder in [Bi₆O₄(OH)₄](NO₃)₆: structure determination using synchrotron radiation and microcrystal X-ray diffraction. *Journal of Solid State Chemistry*, 179(10), pp.3087-3094.