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Selenium (–II, IV) interactions with Fe(II) species and phases under anoxic and reducing conditions

PAUL CLARENCE FRANCISCO[1], RYOSUKE KIKUCHI[2], DAIJU MATSUMURA[1], DR. HIDEAKI SHIWAKU, PHD[1], TAKAMITSU ISHIDERA[1] AND YUKIO TACHI[1]

[1]Japan Atomic Energy Agency

[2]Hokkaido University

Presenting Author: paul.francisco@jaea.go.jp

Selenium-79 (Se-79) will be an important contributor to the long-term radiologic hazards in geological repositories for high-level wastes due to its long half-life. Thus, understanding its behavior in anoxic and reducing subsurface environments prevalent in geological repositories is a key concern in repository safety assessments. There is limited information on primary reaction products arising from the interaction of aqueous Se species with either aqueous or solid Fe(II) phases, which are present in geological disposal repositories either as corrosion products of the steel overpack or as accessory phases in buffer materials or host rocks.

In this work, we investigated the interaction of aqueous Se(–II) and Se(IV), potentially the most mobile Se species in geological disposal repositories, with aqueous Fe(II) and solid $Fe(OH)_2$ at circumneutral and alkaline conditions, respectively. Using electron microscopy and XAFS spectroscopy, we identified β-FeSe as the primary reaction product at circumneutral and alkaline conditions between Se(–II) and either aqueous Fe(II) and $Fe(OH)_2$. For Se(IV), elemental Se(0) was the main reaction product at circumneutral conditions while iron selenides only became dominant at alkaline conditions; these results show rapid and efficient reduction of Se(IV). Aging the solids at 90°C for 1 week induced the transformation of β-FeSe and elemental Se(0) to $FeSe_2$ at circumneutral conditions, while a proportion of Se was released back into solution at alkaline conditions. These results provide constraints on the initial behavior of Se upon release from the waste, and may be useful in validating long-term thermodynamic predictions.

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