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Efficient separation of Re(VII) by radiation-induced reduction from aqueous solution

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Abstract: Rhenium (Re) is a good nonradioactive alternative of technetium-99 (^{99}Tc), which is a β emitter causing long-term radioactive toxicity due to its long half-life and high yield in nuclear reactors. TcO_4^-, the dominant form of ^{99}Tc, is very easy to migrate in the environment because of its high solubility. In this work, γ irradiation was applied to separate rhenium from aqueous solution in the presence of isopropanol. Re(VII) was reduced by hydrated electrons (e_{aq}^-) generated from γ-radiolysis of water, leading to the formation and precipitation of Re(0) and rhenium oxides (ReO_2 and ReO_3) nanoparticles. It was the first time that Re(0) was observed in the reduction products of Re(VII) by γ irradiation, which was confirmed by HR-TEM, EDX, XPS and XRD. The reduction of Re(VII) by γ irradiation was very fast and efficient in alkaline condition, whose separation ratio reached 93.6% after 2-hour irradiation and the final separation ratio was as high as 98.1%. Increase in absorbed dose rate, isopropanol concentration, and pH, was conducive to the separation of Re overall. Further studies on the transient species by pulse radiolysis revealed that OH^- might coordinate with the intermediate Re(VI) forming different complexes with different concentrations of OH^-, thus affecting the reduction of Re(VII). This work not only demonstrates that γ irradiation could be a promising method for the efficient separation of Re(VII), but also gives a better understanding in the reaction mechanism.

Key words: γ irradiation; separation; rhenium(VII); technetium(VII); pulse radiolysis