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1,10-phenantroline complex of iron(II) nitrate: the challenging salt for the technetium precipitation from nitric acid solutions

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Tc-99 is a long-lived radioactive isotope; its main source is a spent nuclear fuel (SNF). Thus, it is necessary to localize Tc either into the glass or in the other solid matrix with a slight leaching or into the matrixes fitted for transmutation to prevent technetium escape into the environment. Due to the widely used hydrometallurgy flowsheet of the SNF reprocessing, Tc occurs as pertechnetate (TcO_4^-) ion in the nitric acid solutions.

The possibility of pertechnetate precipitation with organic complexes of Fe(II) was studied earlier. Among these compounds, o-phenantroline complex of iron(II) (Ferroine nitrate) provided most complete precipitation of pertechnetate ion.[1] The effect of the precipitant amount and the temperature of the initial and the precipitant solutions on the technetium precipitation from 1 M nitric acid solutions was studied.[2] The conditions providing maximal precipitation of Tc (up to $93 \pm 5\%$) were determined.

The main goal of this work was to study the mechanism of the precipitate formation under the conditions providing maximal technetium recovery from the solutions with the aim to obtain stable metal-technetium matrix.

The precipitation of TcO_4^- with Ferroine sulfate from the sulfuric acid solutions (with increasing H_2SO_4 concentration up to 3 M) was studied. Under these conditions the degree of Tc precipitation was $97 \pm 3\%$. On changing sulfate to nitrate media the degree of technetium precipitation decreased to $93 \pm 5\%$. The oxidation of Fe (II) to Fe (III) in the nitric acid solutions and its negative effect on Tc precipitation was demonstrated. Absorption (UV-vis) spectra of Tc in the course of its precipitation with Ferroine nitrate in 1 M nitric acid solutions were registered. It was found that no TcO_4^- reduction in the presence of iron(II) in nitric acid media was observed.

To confirm the composition of the precipitates formed under the conditions providing maximal recovery of Tc (up to $93 \pm 5\%$), their IR spectra were recorded. The IR band at 1384 cm^{-1} belonging to coordinated nitrate ion and the IR band at 900 cm^{-1} of TcO_4^- ion were clearly observed. The IR bands at $300\text{-}600 \text{ cm}^{-1}$ of crystal water were also detected. We proposed the following possible formula of the precipitating compound $\text{Fe}(\text{phen})_3(\text{TcO}_4)_x(\text{NO}_3)_y \cdot n\text{H}_2\text{O}$, where x and y changed from 0 to 2 and $x+y=2$.

Based on the precipitate with the highest degree of technetium precipitation of $93 \pm 5\%$, which contained maximal amount of Tc and minimal amount of nitrate-ion, the metal-technetium matrix was prepared.

1 Бойцова Т.А., Мурзин А.А., Бабаин В.А., Осаждение технеция из азотнокислых растворов органическими катионами и комплексами металлов. «Радиохимия-2012», Тезисы докладов, с. 200.

2 T.A. Boytsova, A.A. Murzin, B.A. Babain, The pertechnetate-ion precipitation in nitric acid solutions with o-phen complex of iron (II) nitrate., «Russian-nordic Symposium on radiochemistry», Abstracts, p. 5, <http://rnsr.org/images/7%20boytsova.p>

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