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Mechanisms of Np(V) interaction with soil horizons: reduction and complexation

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Nuclear energy production and nuclear legacy sites management raise concerns about radioactive waste, of which ^{237}Np is a significant component due to being a long-lived alpha-emitter with activity increasing over time as a result of ^{241}Am decay. Neptunium is mostly present in oxic environments in a highly mobile form, NpO_2^+. Understanding its interaction with complex environmental matrices, such as soils, is crucial for modeling potential migration. This study aimed to investigate the mechanisms of neptunium sorption onto individual peaty-podzolic-gleyic soil horizons with varying characteristics.

Np(V) interaction with soil horizons was studied in two variations: sorption and incubation. Sorption focused on determining Np fixation features in solutions with varying pH (1-8) and ionic strength (0.01-0.5M) values over time. Solutions were additionally analyzed by spectrophotometry and redox potential measurements. The objective of incubation was to observe sorption in conditions resembling natural ones, alternating wetting and drying in two-week cycles over a period of 3 months at 8°C. Np speciation in the samples was examined by X-ray absorption spectroscopy (XANES) and sequential extraction following modified Tessier procedure.

It was established that two lowest soil horizons of the five studied in this work sorb Np(V) in a similar way: sorption is pH-dependent, gradually increasing with increasing pH, steady-state is reached within a week. This is consistent with Np(V) forming inner-sphere complexes with illite and iron (hydro)oxide minerals, which are more prevalent in lower horizons. Absence of sorption dependence on solution ionic strength indicates absence of outer-sphere complex formation. Sequential extraction results demonstrate that neptunium is predominantly associated with readily available fractions, maintaining the mobility characteristic of the NpO_2^+ cation.

In the case of two upper horizons containing more organic matter, neptunium sorption is distinctly different. At pH<4, atypical for pentavalent neptunium species sorption values up to 90-95% are observed. For these horizons Np(V) reduction to Np(IV) was confirmed by XANES and sequential extraction results[1]. However, at higher pH values, sorption values decrease to 40-50%, which may indicate sorption competing with complexation to organic matter in solution.

[1]Skryleva P. et al.//Chemosphere. 2025,T. 374, C.144196.

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