

Trace Analysis of Neptunium with Resonance Ionization Mass Spectrometry

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After a storage time of more than 1000 years, the radiotoxicity of spent nuclear fuel is mainly determined by long-lived isotopes of plutonium and the minor actinides neptunium, americium and curium. Hence, safety assessments of possible nuclear waste repositories must consider the geochemical behaviour of these elements.

Under natural conditions, neptunium can occur in the tetra- or pentavalent state. Depending on its oxidation state, neptunium has different migration properties. While Np(V) has a high mobility in aquatic systems, Np(IV) is much less soluble and has a strong tendency towards sorption on host materials. Speciation studies are therefore important to understand the migrational behaviour of neptunium. Very sensitive methods for the detection of neptunium are required, since the concentrations in aquatic systems of a nuclear waste repository are expected to be less than 10^{-10} mol/L.

We have developed a method for the sensitive detection of the long-lived isotope ^{237}Np ($T_{1/2} = 2.14 \cdot 10^6$ a) applying resonance ionization mass spectrometry (RIMS). In RIMS, laser light is used to produce neptunium ions by three-step resonant excitation and ionization of neutral neptunium atoms. Because of the uniqueness of optical transitions, this ionization process is element specific. In our setup, three titanium-sapphire lasers provide light for excitation and photo-ionization of neptunium atoms. The resulting neptunium ions are detected subsequently in a quadrupole or time-of-flight mass spectrometer.

Since only few energy levels for a multi-step excitation and ionization of ^{237}Np have been known, extensive spectroscopic studies had to be carried out. These studies led to the identification of suitable energy levels for a three-step excitation and ionization of ^{237}Np and will be presented in detail.

As a future application in speciation analysis, it is planned to use capillary electrophoresis coupled off-line with RIMS for the speciation of the different oxidation states of neptunium at ultratrace level.

Two of us (N.S. and S.R.) thank the Deutsche Forschungsgemeinschaft (Graduiertenkolleg 826) for financial support.