Americium and Samarium Determination in Aqueous Solutions after Separation by Cation-Exchange

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Introduction

Analysis of man-made radionuclides in environmental samples is of particular interest with respect to radiation protection purposes and studies on the behavior and transport of radionuclides in natural environment. There is particular interest regarding actinides (e.g. U, Pu, Np, Am Cm), because of the amounts produced in fission reactors and their long-lived radionuclides, which are generally highly radiotoxic alpha radiation emitters. The trivalent state is one of the most common oxidation states of minor actinides (e.g., Am and Cm) in aqueous solutions under most environmental conditions [1].

In this study, we have investigated the applicability of Chelex-100 in the separation and pre-concentration of Am and Sm from aqueous solutions prior analysis. Results obtained from experimental investigations regarding chemical recovery and the effect of physicochemical parameters such us pH, salinity, and the presence of competitive cations (e.g. Fe^{3+} and Ca^{2+}), and HA and SiO_2 concentration, on the separation of the trivalent actinide/lanthanide from aqueous solutions by Chelex-100 has also been investigated. The investigation was performed to evaluate the applicability of cation exchange and liquid extraction as separation and pre-concentration procedures prior to the determination of Am in water samples.

Experimental

The radiometric analysis of Am-241 was performed using a high-resolution alpha-spectrometer, after pre-concentration [2] and separation of the radionuclide by cation exchange using Chelex- 100 resin and the quantitative analysis of samarium was performed by spectrophotometry using arsenazo-III [3].

All experiments carried out at room temperature ($22\pm3°C$) under normal atmospheric conditions and pH 2.5. For the evaluation of the pre-analytical procedure (e.g. the ion-exchange efficiency) the present method was applied to laboratory solutions of constant Sm concentration (2×10^{-6} mol/l) and variable composition (0.1-1 M NaCl; 0.05-1 mM $FeCl_3$;) The solutions were prepared by dissolution of the appropriate amount of the corresponding salt (analytical grade, Aldrich) in de-ionized water and addition of a constant volume of the samarium standard solution.
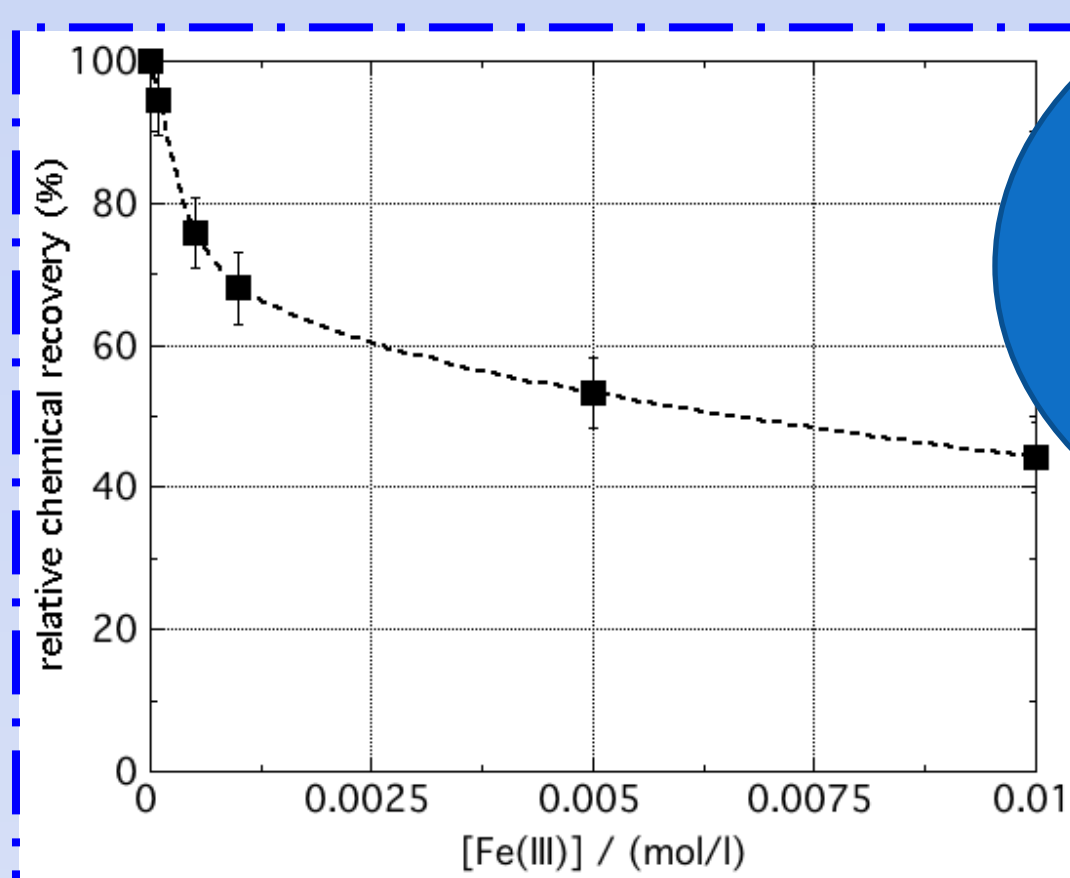
The effect of salinity and Fe^{3+}
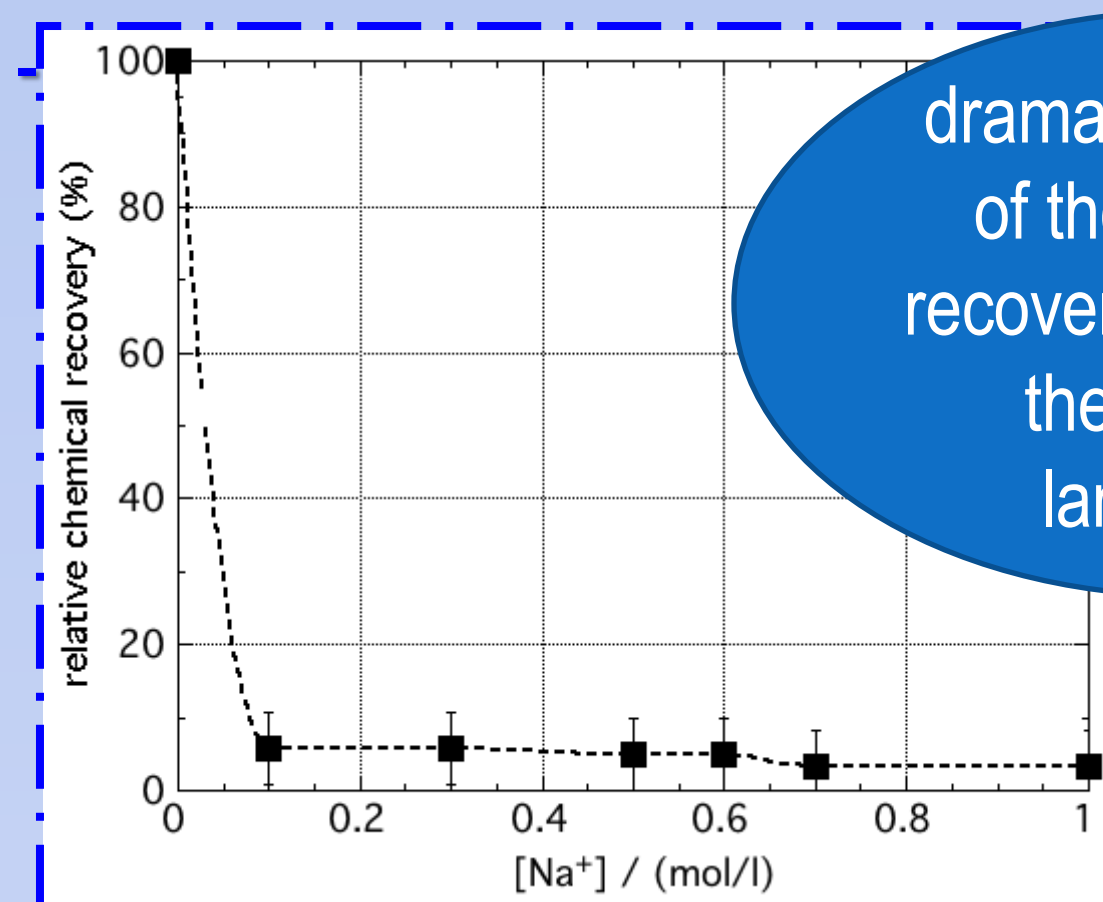
dramatic decrease of the chemical recovery (< 10%) of the trivalent lanthanide

▪ Binding of M^{3+} by the Chelex-100 resin at pH 2.5 is based on electrostatic interactions and on a cation exchange mechanism

The method could be restricted only to low-salinity waters

[Fe^{3+}] affects significantly the lanthanide recovery ⟶ almost 45% separation recovery at [Fe^{3+}]= 0.01 mol/l

Higher affinity of the resin for the Fe^{3+} cations at pH=2.5

Fe^{3+} and Ac^{3+} /Ln^{3+} cations present similar charges

Competing effectively the binding by the resin through its iminodiacetic moieties

A possible interaction between Am^{3+} and the iminodiacetic moiety
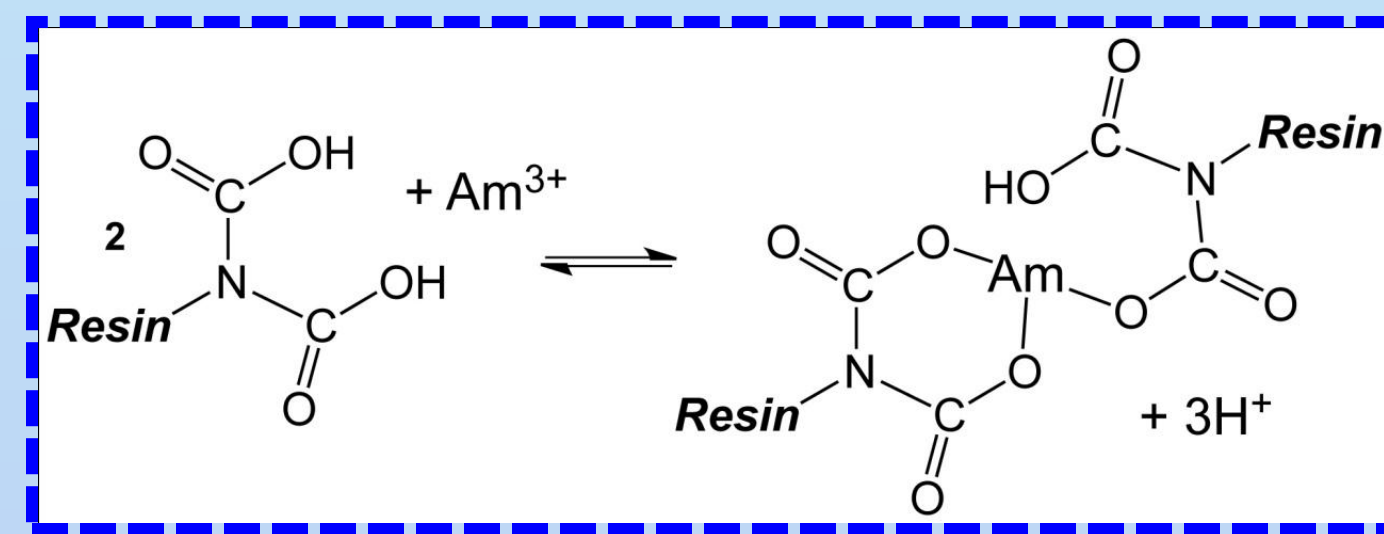
[H^+] —› key factor governing the H^+ dissociation of the iminodiacetic moieties and Am(III) or trivalent lanthanide hydrolysis [4]
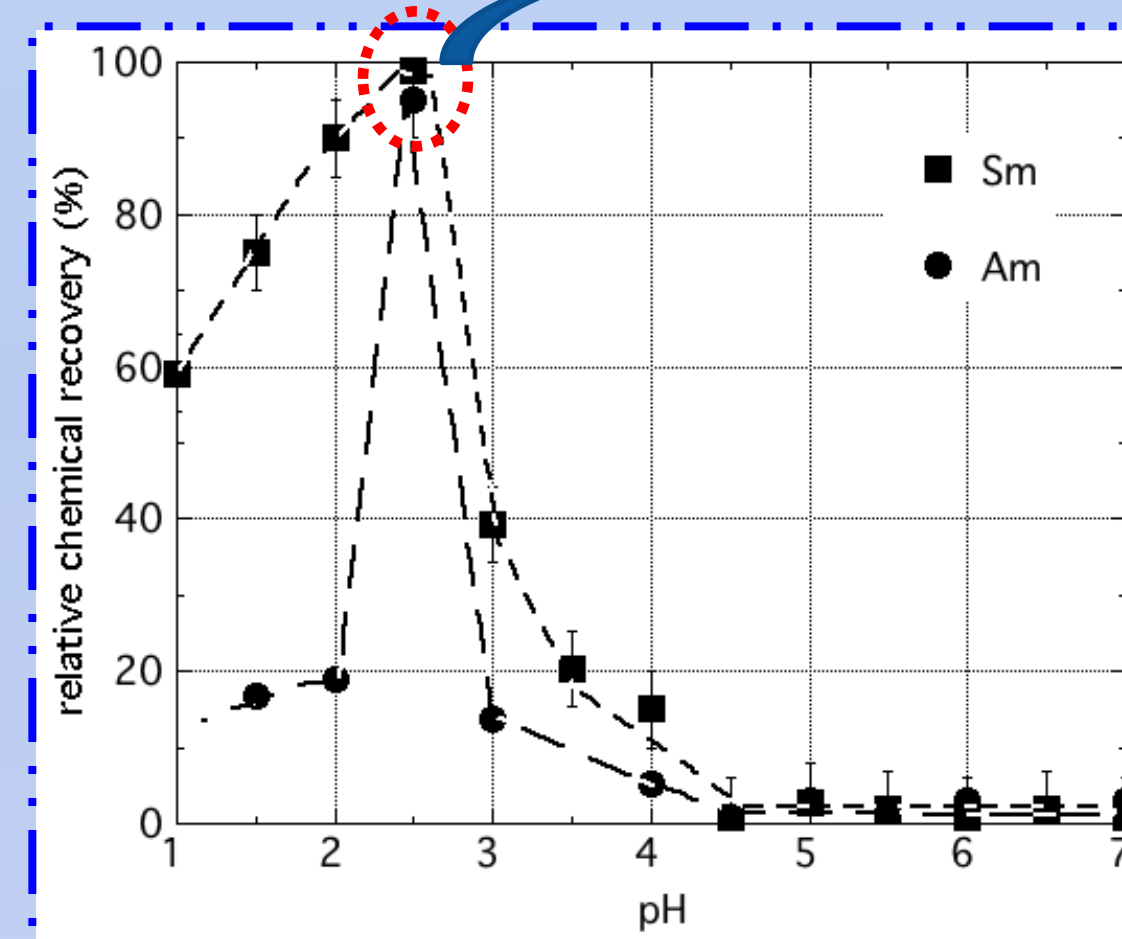
optimum pH for the separation efficiency of Am and Sm

similar chemical behavior of the +III actinide and lanthanide in aqueous solutions.

Below and above pH 2 the affinity decrease

▪ Concentration of the competing protons increases dramatically
▪ M(III) starts forming hydroxo complexes species

Stabilize the metal ions in the solution

Conclusions

The results obtained from this study lead to following conclusions:

▪ The maximum separation efficiency of Chelex-100 for Ac^{3+} /Ln^3 is given at pH 2.5 .

▪ Increasing salinity (e.g. [Na^+]) and iron ([Fe^{3+}] >10^{-4} mol/l) concentration in solution results generally in decreasing chemical recovery.

▪ The method could be successfully applied for americium determination in waters of low-salinity and metal-ion contamination.

References

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