Journal of Radioanalytical and Nuclear Chemistry (2019) 321:973–975
https://doi.org/10.1007/s10967-019-06639-x

Alpha-spectroscopic analysis of uranium in ground- and seawater samples after EDTA-masking of interfering cations

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Received: 17 April 2019 / Published online: 5 July 2019

Abstract

The effect of EDTA-masking on the alpha-spectral characteristics corresponding to uranium analysis in ground- and seawater samples after cation-exchange and electrodeposition is systematically investigated. Addition of EDTA (1 mmol l^{-1}) to the investigated samples results in dramatic increase of the spectra quality, including improvement of the separation efficiency up to 75% and 85% for ground and seawaters, respectively. For EDTA concentrations lower than 1 mmol l^{-1} the spectral resolution and counting efficiencies decline, whereas for higher concentrations the resolution increases but the separation efficiency decreases due to the U(VI) stabilization in solution.

Keywords Uranium · Ground- and seawaters · EDTA-masking · *Chelex*®100 · Alpha-spectrometry

Introduction

Analysis of uranium in ground- and seawaters is of particular interest with respect to uranium monitoring in water resources for drinking purposes [1], environmental issues [2–6], and uranium recovery [7]. Uranium analysis by alpha-spectroscopy after cation-exchange separation using *Chelex*®100 has been extensively described [8–10] and used for the uranium separation from tap-, ground- and seawaters prior analysis by liquid scintillation counting [11] and alpha-spectroscopy [12].

Regarding alpha-spectroscopic analysis the corresponding measurements have shown that in waters with increased concentration of interfering cations (e.g. Ca^{2+}, Fe^{3+} etc.) the spectral resolution, as well as the counting efficiency and subsequently the detection limits were declined due to the deposition of interfering cations on the stainless steel disk along with the U(VI) cations [1–6, 13]. The deposition of interfering cations results in energy loss of the emitted alpha-particles by the deposited matrix materials and alpha-spectra of low resolution/quality, particularly for samples of increased salinity [14]. Usually organic acids (e.g. acetic, malonic, oxalic acid) are added to the studied waters in order to mask the interfering cations and avoid their transfer in the electrolyte solution and finally their deposition on the source disk [13]. In the present study the addition of EDTA in ground- and seawater samples to mask interfering cations (e.g. Ca^{2+}, Fe^{3+} etc.) has been systematically studied and the optimum conditions were identified.

Experimental

Ground- and seawater samples obtained from a local aquifer and coastal area have been repeatedly (seven times each) analysed with and without EDTA addition and the data obtained have been compared with one another. Prior to any treatment, all water samples were traced with 50 mBq ^{232}U tracer (NPL Laboratories). The uranium separation and pre-concentration were performed by cation-exchange using *Chelex*®100 as described elsewhere [1–5, 8–10]. However, in this study similar samples have been investigated also after adding different amounts of 0.1 M EDTA solution (1, 2, 5, 10 and 20 ml) to 200 ml of the water samples to be analysed. The procedure of the uranium electrodeposition on stainless steel disks was carried out similar to previous studies [1–5].

The alpha-spectroscopic analysis of uranium was performed after pre-concentration and separation of uranium using *Chelex*®100 resin as described elsewhere [1–5, 8–10]. Alpha-spectroscopic measurements were performed

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by means of an a-spectrometer (Alpha Analyst Integrated Alpha Spectrometer, Canberra) equipped with passivated implanted planar silicon (PIPS) detectors and analysed as described elsewhere [1–5].

Results and discussion

Figure 1a, b present alpha-spectra of uranium extracted from ground- and seawater samples, respectively, which have been obtained by alpha-spectroscopy after separation and pre-concentration of uranium by cation exchange from test solutions of varying EDTA concentrations.

From the spectra it is obvious that increasing EDTA concentrations results in significant improvement of the alpha-spectra quality. The spectral resolution increases dramatically, particularly in the case of the seawater samples, due to the masking effect of EDTA, which strongly complexes the interfering matrix cations (e.g. Ca^{2+} and Fe^{3+}) and stabilizes them in solution. This results in the preparation of uniform and almost massless sources, because the addition of EDTA to the solution minimizes the effect of interfering ions, present in environmental water samples [13, 15, 16]. The source discs are coated with a very thin layer of radionuclides and with no other material above to attenuate the alpha-radiation [14].

However, above a certain EDTA concentration (1 mmol l^{-1}) the chemical yield is decreasing significantly, because EDTA at relatively high levels (> 1 mmol l^{-1}) competes with the resin binding of U(VI), stabilizes U(VI) in solution and results in lower chemical/separation yields. The corresponding alpha-spectra have been quantitatively evaluated and the related data are graphically summarized in Fig. 2.

According to the data shown in Fig. 2 the optimum concentration regarding the chemical yield is given when the EDTA concentration is 1 mmol l^{-1} for both ground and seawater samples, which is significantly below the concentration of the interfering ions (e.g. Ca^{2+}) in the respective waters [16]. Nevertheless, EDTA at the given concentration seems to effectively mask the interfering ions, without affecting dramatically the uranium binding by the iminodiacetic resin moieties [9].

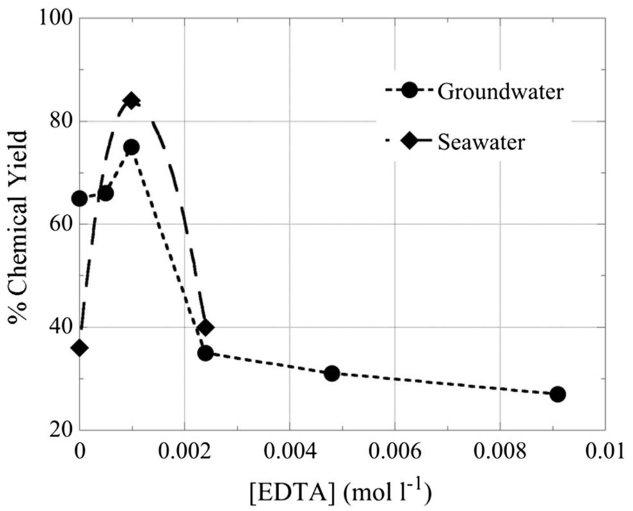

Fig. 2 Relative chemical yield for the uranium separation by cation exchange using Chelex@100 from ground- and seawater samples as a function of the added EDTA concentration

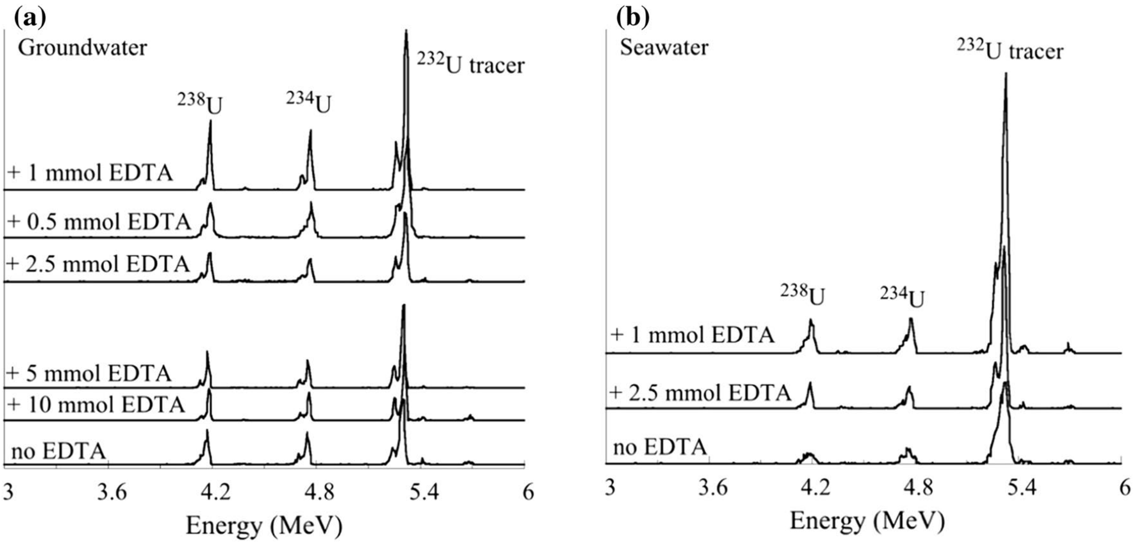

Fig. 1 Alpha-spectra of uranium extracted from **a** ground- and **b** seawater samples obtained by alpha-spectroscopy after cation-exchange separation of uranium by cation exchange from test solutions containing varying concentrations of EDTA

Conclusion

Addition of EDTA at concentration levels around 1 mmol l^{-1} to environmental water samples results in dramatic increase of the alpha-spectra quality and chemical yields up to 75% and 85% for ground and seawaters, respectively.

For EDTA concentrations lower than 1 mmol l^{-1} the spectral resolution and counting efficiencies decline, whereas for higher concentrations the resolution increases but separation efficiency decreases due to the U(VI) stabilization in solution.

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