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Determination of uranium isotopes in environmental samples by anion exchange in sulfuric and hydrochloric acid media [☆]



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HIGHLIGHTS

- The method allows cost-effective determination of U isotopes.
- High amounts of environmental samples can be analyzed.
- High chemical yields, energy resolution and decontamination factors were achieved.
- Uranium isotope concentrations in mineral waters from Bulgaria are presented.

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ABSTRACT

Method for determination of uranium isotopes in various environmental samples is presented. The major advantages of the method are the low cost of the analysis, high radiochemical yields and good decontamination factors from the matrix elements, natural and man-made radionuclides. The separation and purification of uranium is attained by adsorption with strong base anion exchange resin in sulfuric and hydrochloric acid media. Uranium is electrodeposited on a stainless steel disk and measured by alpha spectrometry. The analytical method has been applied for the determination of concentrations of uranium isotopes in mineral, spring and tap waters from Bulgaria. The analytical quality was checked by analyzing reference materials.

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1. Introduction

Uranium is radioactive element with more than 27 characterized isotopes with mass numbers ranging from 215 to 242. It is one of the most important naturally occurring radioactive elements with two radioactive decay chains: most abundant ^{238}U (half-life of 4468.10^9 years, 99.27456% by mass in natural uranium, $12.2369 \text{ Bq mg}^{-1}$ in natural uranium; with its decay product ^{234}U – 2455.10^5 years, 0.00555% mass, $12.5624 \text{ Bq mg}^{-1}$) and fissile ^{235}U (7038.10^8 years, 0.720012% mass, $0.5685 \text{ Bq mg}^{-1}$) used for nuclear fuel (directly or after enrichment) (Laboratoire National Henry Becquerel, 2016). After the enrichment of ^{235}U the remaining ^{238}U (called depleted uranium - DU) is used mainly as counterweights in aircraft.

Most important man-made uranium isotopes are: ^{236}U (2342.10^7 years, found in spent or reprocessed nuclear fuel due to decay of ^{240}Pu), fissile ^{233}U (1592.10^5 years, produced by

irradiation of ^{232}Th and possibly reasonable alternative to ^{235}U nuclear fuel) and ^{232}U (70.6 years) used as radioactive yield tracer in radiochemical determinations of uranium isotopes (Laboratoire National Henry Becquerel, 2016).

Natural uranium is found at trace levels in all major environmental samples (water, soil, organic materials, etc.). Direct measurement of uranium (^{238}U and ^{235}U only) in environmental samples is possible by gamma spectrometry only when its concentrations are higher than its natural levels in the environment. Usually uranium is measured always after radiochemical separation procedures. Although mass-spectrometry (Avivar et al., 2012; Boulyga and Becker, 2002; Maxwell et al., 2014) has been gathering momentum in the recent years for measurements of long-lived alpha and beta radionuclides, alpha spectrometry is still the preferred instrument for measurements of alpha radionuclides due to the relatively lower price and maintenance costs of the equipment (Bojanowski et al., 2002; Boryło, 2013; Chen, 2001; Jia, 2016; Popov, 2013).

Nowadays separation of uranium is widely conducted by the use of extraction chromatography resins:

- UTEVA Resin[®] (Horwitz et al., 1992),

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- TRU resin[®] (Maxwell and Culligan, 2006; Maxwell et al., 2014),
- TOPO-Microthene resin (Jia et al., 2004; Jia, 2016)

as well as anion exchange resins (Bojanowski et al., 2002; Boryło, 2013; Chen et al., 2001;) and solvent extraction (Popov, 2012; 2013).

In dilute sulfuric acid (or sulfate) media U is quantitatively extracted by strong base anion exchange resins (Bojanowski et al., 2002; Boryło, 2013; Korkisch, 1969). Additional study on anion exchange behavior in sulfuric acid media of U, Pu, Np, Th, etc. was performed and based on the results a cost-effective radioanalytical method for determination of U in environmental samples containing high amounts of matrix elements has been developed.

2. Experimental

2.1. Reagents and equipment

All chemicals (Merck, Fluka) used were of analytical grade. Anion exchange resin AG1 × 4 (100–200 mesh) and borosilicate glass Econo-Column[®] chromatography columns (0.7 × 20 cm and 1.0 × 20 cm) were purchased from BioRad (USA). A Mars 5 (CEM, USA) microwave system with closed PTFE-vessels (EasyPrep[™]) was used for acid decomposition of the solid samples. A Hermle Z513 (Germany) centrifuge (with 100, 250 and 500 ml tubes) was used for separation of the precipitates. Uranium-232 (0.3633 Bq/g ± 1.6%, k=2) and plutonium-242 (0.1765 Bq/g ± 2%, k=2) tracer solutions were purchased from the Czech Metrological Institute (Czech Republic).

After their radiochemical separation / purification and electrodeposition on stainless steel disks ($\varnothing=20$ mm), isotopes of uranium were measured on the alpha spectrometer “Alpha Analyst” (Canberra, USA) with 8 vacuum chambers (PIPS detectors with a 450 mm² active area, 20 keV FWHM and 22% efficiency for a distance of 5 mm between planchet and detector surface).

2.2. Samples

Mineral, tap and spring water samples from Western and Southern Bulgaria were collected. Sampling locations are shown on Fig. 1.

2.2.1. A. Uranium, plutonium and thorium behavior on anion exchange resin in sulfuric acid media

2.2.1.1. Elution curves of U, Pu and Th with anion exchange resin in 0.2 M to 4 M H₂SO₄ acid media (without reducing/oxidizing

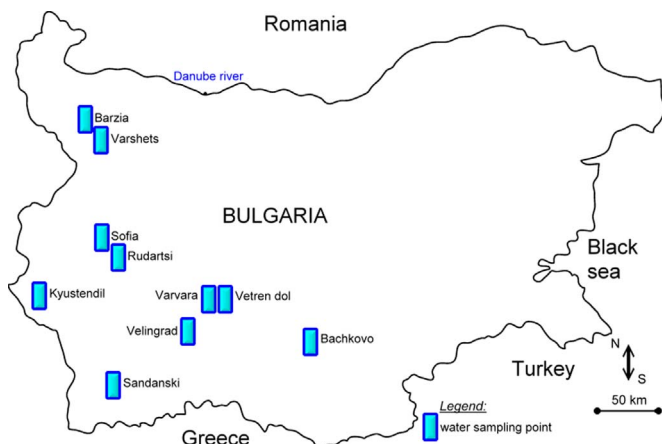


Fig. 1. Map of the sampling locations.

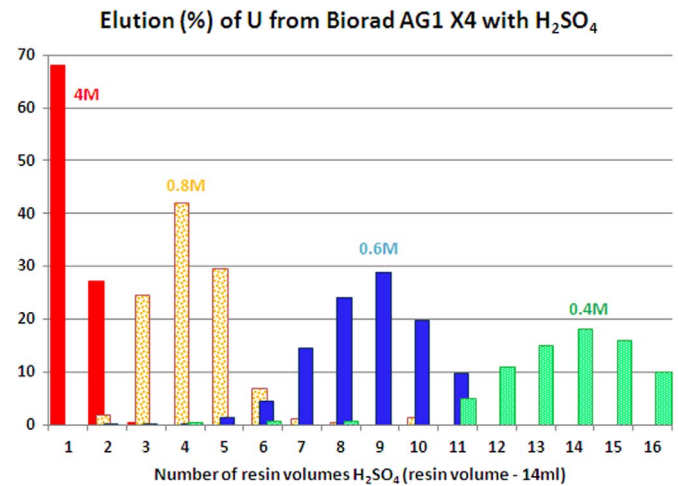


Fig. 2. Elution curve of U from Biorad AG1 × 4 with H₂SO₄.

agents). Ten portions of 14 ml (V_R , resin volume) Biorad anion exchange resin were transferred in 10 glass columns (i.d. × column length = 10 × 200 mm, 14 mm resin bed length). The resin in each column was preconditioned with 84 ml ($6V_R$) solutions of 0.2 M, 0.4 M, 0.6 M, 0.8 M, 1.0 M, 1.2 M, 1.4 M, 1.6 M, 2.0 M and 4.0 M H₂SO₄ correspondingly. Ten beakers containing 14 ml 0.2 M to 4 M H₂SO₄ were spiked with ²³²U and ²⁴²Pu tracers. The solution in each beaker was passed through the corresponding column. The columns were washed with $17V_R$ (~240 ml) 0.2 M to 4 M H₂SO₄ solutions by portions of 14 ml (V_R). Each portion was evaporated to dryness. It was continued with Section A.3 – Electrodeposition. The results are presented in Figs. 2–4.

In dilute sulfuric acid media U is quantitatively extracted by strong base anion exchange resins (Korkisch, 1969). Depending of the concentration of H₂SO₄ different elution curves for BioRad AG1 × 4 anion exchange resin were built for U, Pu and Th (Figs. 2–4). Elution curves of U widen with decreasing concentrations of H₂SO₄ and its quantitative adsorption is possible only at concentrations of 0.2 M H₂SO₄ and lower (for very large samples / elution volumes concentrations even lower than 0.1 M H₂SO₄ will be required). At 0.2 M H₂SO₄ except U, Pu, Th, Po and some rare earth elements like Zr, Nb, Mo, Pa, etc. are partly (Pu, Th, Po, etc.) or fully (Pa, Zr, Nb, Mo) co-adsorbed by the resin and additional techniques should be applied for their separation from U.

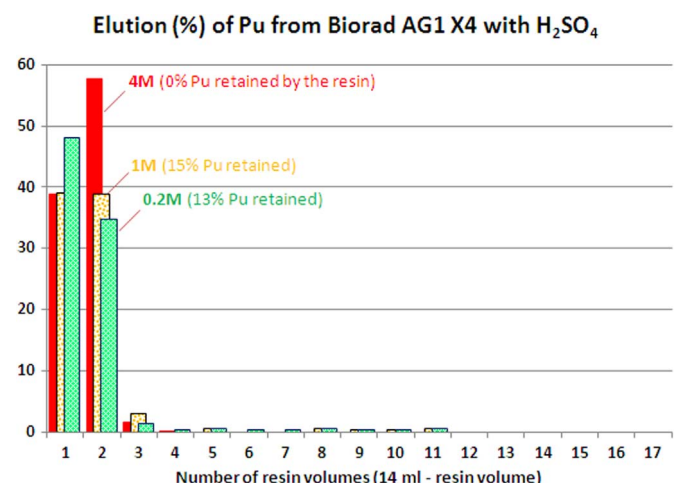


Fig. 3. Elution curve of Pu from Biorad AG1 × 4 with H₂SO₄.

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