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Accurate fast method with high chemical yield for determination of uranium isotopes (^{234}U , ^{235}U , ^{238}U) in granitic samples using alpha spectroscopy

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ABSTRACT

The present study aims to use the α -spectroscopy at Nuclear Materials Authority (NMA) of Egypt. A radiochemical technique for analysis uranium isotopes was carried out for ten mineralized granitic samples together with the International standards RGU-1 (IAEA) and St₄ (NMA). Several steps of sample preparation, radiochemical separation and source preparation were performed before analysis. Uranium was separated from sample matrix with 0.2 M TOPO in cyclohexane as an extracting agent with a chemical yield 98.95% then uranium was purified from lanthanides and actinides present with 0.2 M TOA in xylene as an extracting agent. The pure fraction was electrodeposited on a mirror-polished copper disc from buffer solution ($\text{NaHSO}_4 + \text{H}_2\text{SO}_4 + \text{NH}_4\text{OH}$). Rectangle pt-electrode with an anode-cathode distance of 2 cm was used. Current was 900 mA and the electrodeposition time reach up to 120 min. The achieved results show that the chemical yield ranged between 87.9 ± 6.8 and 98 ± 8.6 .

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1. Introduction

Alpha-spectrometric determination of uranium in geological samples requires thin, homogeneous and nearly weightless sample sources. Alpha-particle spectrometry is performed after a radiochemical procedure. In addition, the use of an alpha-emitting isotope ^{232}U (half-life of 70.6 a, $E_\alpha = 5320.24$ keV, probability = 69.1%) [1] as an internal tracer makes it a highly reliable technique. The main disadvantage of this method is that pure solution of actinide must be used as the presence of any trace of other elements results in reduction of both deposition yield and α -spectrum quality resolution [2]. For this reason, inactive elements such as iron, aluminum, calcium, etc. contained in samples, are added during the analytical method prior to pre-concentration and/or purification of actinides, are considered as undesirable impurities in electrolytic solution and treatments to remove them. This procedure have critical stages such as digestion of sample, chemical separation of uranium from sample matrix, uranium purification from present actinides and lanthanides to eliminate interfering elements and preparation of a thin homogeneous, virtually weightless source for high resolution spectrum by

deposition of the alpha-emitting radioisotopes onto a counting surface (source preparation). The prepared source introduced for high-resolution alpha-particle spectroscopy and determination of the activity by alpha particle counting in a vacuum chamber with the associated electronics. Accurate determination of alpha-emitting nuclide concentrations in geological samples depends heavily on the analyst's ability to prepare and mount the analyte in a form as pure as possible, free of any gravimetric or radiometric interference, which could lead to complications in the interpretation of the acquired alpha spectra, degradation of the resolution and reduced overall sensitivity. Lee et al., [3] say that the alpha source should be very thin, flat, and uniform to minimize source self-absorption and to obtain the best possible spectroscopic resolution; acid and solvent should be eliminated from the source to keep counting chambers and detectors from being damaged. Lozano et al., [4] the Th/U separation was performed using tri-*n*-butyl phosphate (TBP). Bonotto, et al., [5] determine ^{238}U and ^{234}U in the rock samples using standard alpha spectrometric techniques. After co-precipitation on $\text{Fe}(\text{OH})_3$ and purification by Biorad AG1-X8 100–200 meshes resin anion exchange. Electrodeposition of uranium on a stainless-steel planchete. Tomé, et al., [6] uranium isotopes were extracted with tri-*n*-butyl phosphate (TBP). The purified uranium solution was electroplated onto a stainless steel planchete and determines using standard alpha spectrometric techniques. Dos Santos, et al., [7] determine $^{234}\text{U}/^{238}\text{U}$ activity ratios in silicates volcanic rocks using alpha

Abbreviations: IAEA, International Atomic Energy Agency; NMA, Nuclear Material Authority

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spectrometry with chemical yield, in the range of 51–89%, after purification and electrodeposition on stainless steel disks. Elyahyaoui, et al., [8] uranium was extracted from travertine samples (geological materials) with diethyl ether in acetic acid with recovery yield of 86–94%. The alpha spectra were of good resolution. Numerous methods for radioactive source preparation for alpha spectrometry have been published [9,10] such as evaporation, electroplating, molecular plating, vacuum sublimation, electro-spraying, micro-precipitation, electrodeposition. However, the technique normally used is electrodeposition, since it permits the highest recovery with adequate energy resolution. Pimpl, et al., [11] uranium was separated from matrix elements by combine steps from (solvent extraction, co-precipitation and ion exchange) uranium isotopes were electroplated on stainless steel discs from HCl/oxalate solution by electrodeposition. The chemical yield was in the range of 50–80%, this method was used until now in radiochemistry laboratory of the Egyptian Nuclear Materials Authority [12], Egyptian Nuclear, and Radiological Regulatory Authority [13] for determination of uranium isotopes in granite. However, this method takes long time in preparation of uranium source in addition to its low chemical yield 50–80%. The aim of this work is to carry out a developed method that reduces the time required for alpha uranium source preparation and gives a high chemical yield using Liquid-Liquid Extraction (LLE) technique for separation of uranium from stable and radioactive elements.

2. Experimental

2.1. Sampling

Ten mineralized granitic representative rock samples were collected from different locations, Eastern Desert, Egypt as shown in the Table 1, besides RGU-1 (IAEA) reference uranium material and St₄ (NMA):

2.2. Apparatus and reagents

An ORTEC model 576A-600UH Dual alpha spectrometer with (600 mm²) low-background, ion-implanted silicon detector and electrodeposition cell with copper discs of 22 mm diameter was used. ²³²U standard solution (tracer) was used for the calculation of chemical yield was supplied by NIST (National Institute of Standards and Technology). Tri-n-octylphosphineoxide (TOPO) and Tri-octylamine (TOA) were used. All other chemicals were used of analytical grade.

2.3. Analytical technique

The analytical technique was carried out through several steps including sample preparation and counting procedure. These steps are shown in Fig. 1 and can be summarized as follows:

Table 1
Samples Location.

Sample number	Location
1	From the entrance of the Western mine at Gatter II
2	Gatter II trench
3	95 meter Gatter II west tunnel to the left
4	Gatter V open pit
5	Pink granite Gatter
6	El Missikat
7	El Erediya
8	Abu Rushed
9	Umm Area
10	El Sella

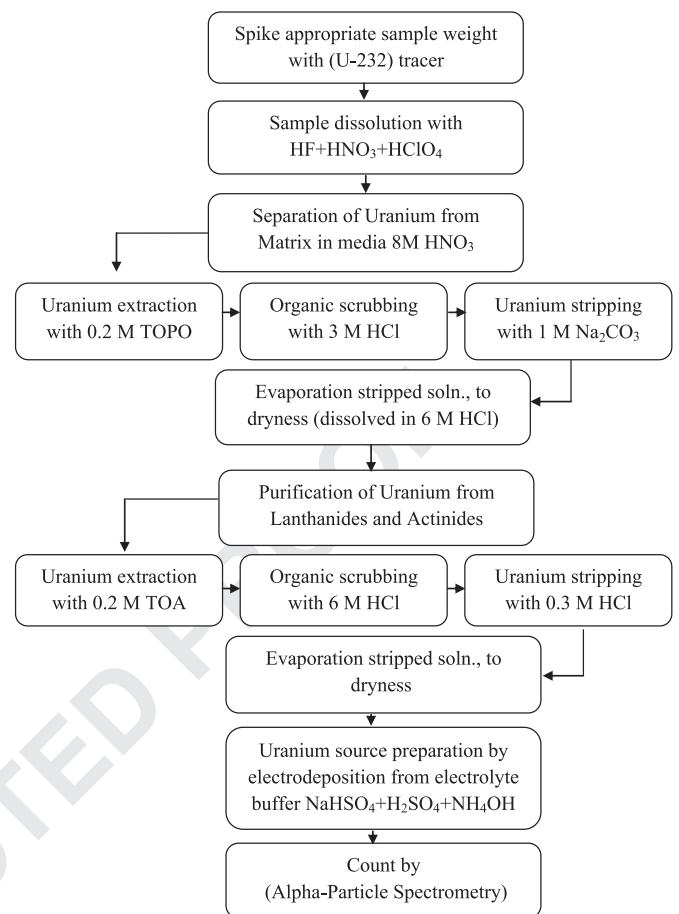


Fig. 1. Radio-Analytical Flow Sheet For Preparation of Ultra-Pure Alpha Uranium Source by The Present Technique.

i. Digestion of samples, separation and purification of uranium from stable and radioactive elements

Uranium was chemically separated from the bulk material of weight (0.0025–1 g) using LLE technique after addition of (13.5–30 Bq kg⁻¹) from ²³²U tracer for chemical yield calculation. Special attention in the chemical separation scheme was paid to remove stable elements also radioactive elements, which disturb the uranium isotopes determination via alpha-spectrometry. Best results for adopted chemical separation were achieved by digest granitic sample using mixture from mineral acids [HF (40%)+HNO₃ (69%)+HClO₄ (70%)] by ratio [5:3:1] and the last medium is 8 M HNO₃ media using (0.2 M TOPO/cyclohexane) phase ratio (O: A)=(1:4) in two stages and contact time (15 min). Organic phase was scrubbed using 3 M HCl phase ratio (O: A)=(1:1) in three stages and contact time (5 min). Finally, uranium was stripped by 1 M Na₂CO₃ phase ratio (O: A)=(2:1) in two stages and contact time (15 min). Then the media was changed from 1 M Na₂CO₃ to 6 M HCl for uranium purification from radioactive elements (lanthanides and actinides) using 0.2 M TOA/xylene phase ratio (O: A)=(2:1) in two stages and contact time (15 min). Organic phase was scrubbed using 6 M HCl phase ratio (O: A)=(1:1) in two stages and contact time (5 min). Finally, uranium was stripped by 0.3 M HCl phase ratio (O: A)=(1:1) in two stages and contact time (15 min). The aqueous solution was filtered through whatman (40) filter paper.

ii. Alpha uranium source preparation

The following relevant factors, effecting on electrodeposition

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