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Modification on radiochemical procedure used for determination of uranium isotopes by alpha spectrometry in some stream sediment samples

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ABSTRACT

The present study aims to make modification on radiochemical procedure used for U-isotopes determination at Nuclear Materials Authority (NMA). A radiochemical technique for analysis of U-isotopes was carried out for stream sediment sample collected from Wadi El-Reddah and also for reference soil sample of International Atomic Energy Agency (IAEA). The uranium was extracted from the matrix elements with trioctylphosphine oxide (TOPO) and Di-2-ethylhexylphosphoric acid (D2EHPA) and stripped with 0.75M Na_2CO_3 solution. Modifications were performed for the procedure to give highest extraction. The obtained result of modified technique for a soil reference sample gave a greatly similar result to those obtained by the α-spectrometry method of IAEA for analysis of U-isotopes.

KEYWORDS

U-isotopes;
Alpha spectrometry;
Stream sediment samples;
Wadi El-Reddah.

INTRODUCTION

Uranium is widely distributed lithophile metallic element. It may be present as a significant component in some minerals (e.g. uraninite, uranophane, brannerite, uranophane and carnotite) or as an accessory element in others (e.g. zircon, apatite, allanite and monazite). The natural uranium consists of three isotopes; ^{238}U, ^{235}U and ^{234}U with abundances of approximately 99.27, 0.72 and 0.054%, respectively. The three isotopes are α-emitters as follows; i) ^{238}U decay by α-emission into ^{234}Th which decays by beta emission into protactinium (^{234}Pa). ^{234}Pa decays by beta emission into ^{234}U, ii) ^{234}U decays by α-emission into thorium (^{230}Th) and iii) ^{235}U decays by α-emission into thorium (^{231}Th) which is then decayed by beta emission into protactinium (^{231}Pa)[1].

The uranium concentration and U-isotopic ratios are usually detected and determined in various environmental, geological and biological samples by different non-destructive and destructive techniques. The non-destructive techniques are mostly achieved by γ-spectrometers (e.g. NaI- and HPGe- detectors). They are carried out on the bulk samples without the need for complicated and time consuming radiochemical methods[2]. On the other hand, the destructive techniques are carried out through several analytical methods (e.g. α-particle spectrometry, fluorimetry, kinetic phosphorescence, neutron activation analysis... etc). Among these techniques, α- particle spectrometry is the most common to obtain the isotopic composition and can detect low uranium levels (below ngl^{-1}). Its detection limit is 100- 1000 times lower than γ-spectrometry[3]. This technique is mostly used for detection and analysis of U- as well as Th-isotopes, particularly in the environmental and water samples which are characterized by low level activity concentration[2,4].

In addition, it is useful technique for geological studies[5-7] characterization of nuclear wastes[8-11], nuclear decay data measurments[12] as well as the studies related to health and security[13-16]. Also very useful to determine recent uranium deposits. Stream sediments remain the most important medium for geochemical exploration. According to IAEA [17], surficial deposits considered from the most important terms for uranium production, most deposits belonging to surficial deposits are of recent age and are found in surficial depression, solution cavities (Karst) or near-surface joints and fracture system. Uranium occurs almost exclusively as secondary minerals (hexavalent stage) or adsorbed on other materials. Because of their relatively young age, they are not easily detected by the conventional radiometric method of surveying[17]. So here come the importances of alpha spectrometry technique in which can determine the concentration of recent deposits which other techniques can't do it. In addition, it is important to make some modification on radiochemical procedure of alpha spectrometry to get better extraction for uranium.

SAMPLES AND METHODS

Reagents

TOPO and D2EHPA solvents were obtained from Merck Co., and were used without further purification. Uranium stock solution has a concentration of 1000 ppm and was prepared by dissolving uranyl nitrate $UO_2(NO_3)_2.6H_2O$ in 0.01M HNO_3. All other chemical reagents were of analytical grade (AR).

Samples

Three stream sediment samples were selected for the present study. The collected samples are representing sediment in horizontal scale from the upper layer of Wadi El-Reddah (about 30 cm depth). In addition, one standard soil sample of low U-activity concentration from IAEA (IAEA-312) was re-analyzed and used as reference sample to control the analytical procedure.

Methods

Method for modification

Spectrophotometric determination of uranium (VI) using Arsenazo (III) for the experiments of modification[18].

Alpha particle spectrometry

The procedure of alpha particle spectrometry is carried out through several steps including sample preparation, radiochemical separation, source preparation and α-counting[19-22]. Sample preparation aims to convert the sample into a thin layered, chemically isolated form that can be placed into the spectrometer and counted with a minimum of interferences and self absorption. It is often an extensive process and requires several steps including; i) sample digestion (preliminary treatment), ii) uranium separation and purification and iii) source preparation. The uranium separation is usually carried out using different techniques such as co-precipitation, liquid-liquid extraction, ion exchange and extraction chromatography. Several methods and flow charts have been reported in different literatures for sample preparation of U-analysis using α-particle spectrometry[2,23-26].

The modification for radiochemical procedure for analysis of U-isotopes using α- particle spectrometry wasn't used before at Nuclear Materials Authority (NMA). Therefore, the present study is an approach - recorded for the first time at NMA-for establish the optimum conditions for preparation samples of interest to U-isotopes analysis using α- particle spectrometry.

Sample digestion

It is performed to homogenize the sample and to prepare it for the subsequent chemical processing. The crushed sample (> 2 mm) is dried at 110 ºC in an oven until constant. Then ashed with $HNO_3$to achieve oxidation of organic carbon at 550 ºC. A suitable weight of the ashed sample (about 5 g of soil ash and about 0.01 g of the stream ash) was analysed and then the ^{232}U tracer (100 mBq) was added. The sample was digested in a mixture of both nitric and hydrofluoric acids (40: 10 ml) at a temperature of (70-80ºC) giving white residue. The residue is dissolved in 8 M HNO_3 then the sample solution is boiled for 30 min after adjusting the solutions volume up to 100 ml. Two ml of H_2O_2 are added to change the uranium from the tetravalent state to the hexavalent one (steps 1&2, Figure13).

Source preparation

The electrodeposition technique was applied for source preparation in the present study. The electrodeposition cell was manufactured locally. Its wall is made of Teflon, which makes as a good insulating from electricity and as preventing adsorption of any radionuclide on the electrolyte along the wall. The anode of the cell is a spiral of platinum wire. The stainless steel screw cap serves as cathode and holds the stainless steel disk on which the deposition is taken place. The exposed cathode area is 3.14 cm^2 while the height of the cell is 34 mm, enough to contain about 10 ml of electroplating solution without losing any thing through progressive the electroplating. The electroplated uranium is in the form of ammonium format (ammonium oxalate, ammonium chloride or ammonium sulphate or combinations of these electrolytes[27].

The ammonium oxalate was preferred in our study because it's faster and less sensitive to experimental variations and impurities in the electrolyte. The eluted uranium is transferred into the electrolysis cell from the crystallization dish with 0.4 ml of 4 M HCl, three times by 1 ml of $(NH_4)_2C_2O_4$ (4%) and then once 0.6 ml distilled water. The electrolysis is carried out for 3 hours at 300 mA (0.3 A), then 1 ml of ammonia solution (25%) is added. After one minute, the electrolysis current is cut off. The ammonia increases the OH concentration which prevents redissolution of the hydroxide from the cathode surface (step 7, Figure13).

Alpha counting technique

The used alpha particle spectrometry system its α-detector (silicon barrier detector, model 576A with 450 mm^2, Ortec, USA). The silicon barrier detectors are characterized by high resolution performance, low background, excellent stability and high permissible counting rates. The detector efficiency is 23 % with no significant variation in the range interval 2.5-8.8 MeV.

The detection limit of the α- particle spectrometry is reported as 0.002 Bq kg^{-1} for sample[28,29].

EXPERIMENTAL

Effect of different concentrations of TOPO / cyclohexane

10 ml of the aqueous uranium solution (100 ppm U (VI)) and 10 ml of organic solution of TOPO / cyclohexane, with concentration ranging from 0.05 to 0.3 M, were shaken for 20 minutes at room temperature in an extraction funnel (apparatus that is used in mixing and shaking the two phases, aqueous phase and organic phase) to attain equilibrium state. It was found that percentage of extracted uranium increased gradually with increasing TOPO concentration from 0.05 to 0.2 M. After that the extraction was decreased probably due to an increase in viscosity of extractant TOPO that causes a decrease in the mass transfer of U (VI) from aqueous to organic phase. The optimum value of 0.2 M solution of TOPO was chosen Figure 1.

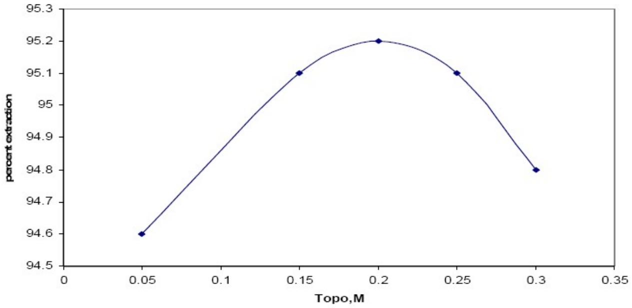

Figure 1: Effect of different concentrations of TOPO on the extraction of uranium (VI)

Effect of equilibration time

The effect of contact time on the attainment of an equilibrium state was studied at intervals between 3–30 minutes, while the other factors were maintained fixed at 1:1 (v/v) organic to aqueous phase ratio, 0.2M TOPO/Cyclohexane and room temperature. The results are shown in Figure 2 quite adequate for efficient uranium (VI) extraction.

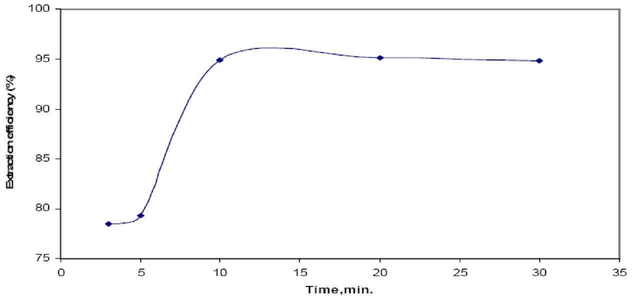

Figure 2 : The effect of shaking time on the extraction of uranium (VI)

Effect of pH on extraction of uranium (VI)

The extraction process has been studied by varying the pH. Other factors were maintained fixed at 1:1 (v/v) organic to aqueous phase ratio, 0.2 M TOPO / cyclohexane, contact time 20 minutes at room temperature. The results are shown in Figure 3. It can be observed that a pH of 1.5 can be taken as an optimum pH value.

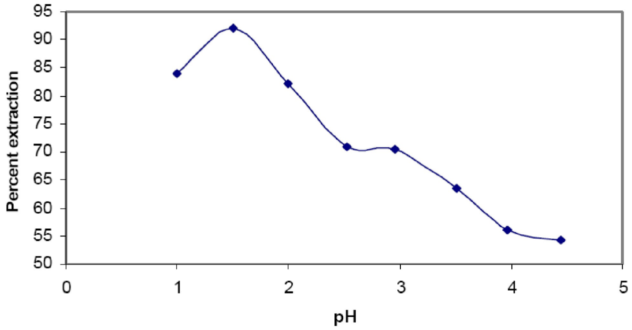

Figure 3: The effect of pH on the extraction of uranium (VI)

Effect of different TOPO diluents on uranium (VI) extraction

The extraction of uranium (VI) has been studied using different organic diluents for TOPO namely, carbon tetrachloride, benzene, toluene, o-xylene, kerosene, cyclohexane and chloroform. The other studied factors were maintained fixed i.e. 1:1 (v/v) organic to aqueous phase ratio, room temperature, 0.2 M TOPO in all diluents and contact time 20 minutes. It was found that although benzene and toluene have dielectric constants of the same order of magnitude as that of cyclohexane, both are less efficient at extracting uranium (VI). This is probably due to the lower solubility of the extracted species in these diluents as compared with cyclohexane. It is clear from Figure 4 that cyclohexane is the best diluents.

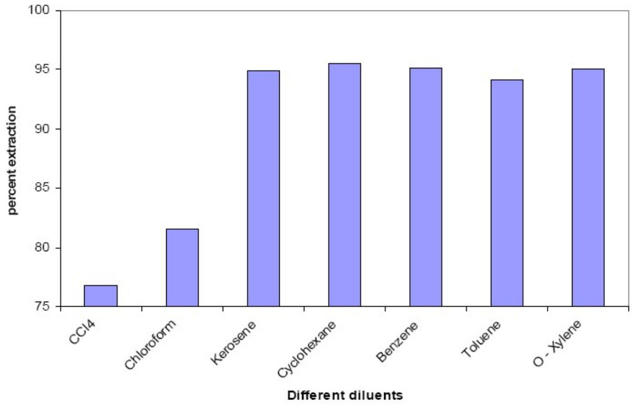

Figure 4: Effect of different diluents on the extraction of uranium (VI)

Effect of temperature on the extraction of uranium (VI)

For studying the effect of temperature on distribution coefficient and extraction percentage, a series of experiments were carried out, being the temperature range from 25 – 60ºC. The other factors were maintained constant at optimized conditions.

From the obtained results shown in Figure 5 it is clear that room temperature can be considered as the best temperature. It is obvious from the data obtained, that percent extraction decreases with increasing temperature. This means that the extraction of uranium (VI) is exothermic. This behaviour was similar to that reported by[30].

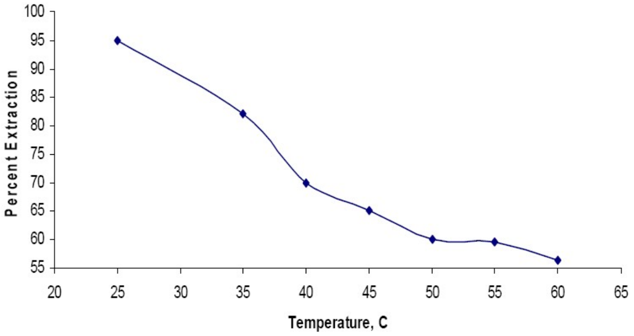

Figure 5: Effect of temperature on the extraction of uranium (VI)

Equilibrium line and construction of McCabe-Thiele diagram for uranium (VI) extraction

20 ml of each of the organic phase (0.2 M TOPO in cyclohexane) and aqueous phase were contacted for 20 minutes until equilibrium was obtained. The phases were allowed to separate and the aqueous phase was removed and analyzed. Fresh aqueous solution was then added to the organic phase to give the same phase ratio as that originally used.

The phases were again contacted until equilibrium was obtained, and the procedure repeated. This process was carried out until saturation of the solvent with

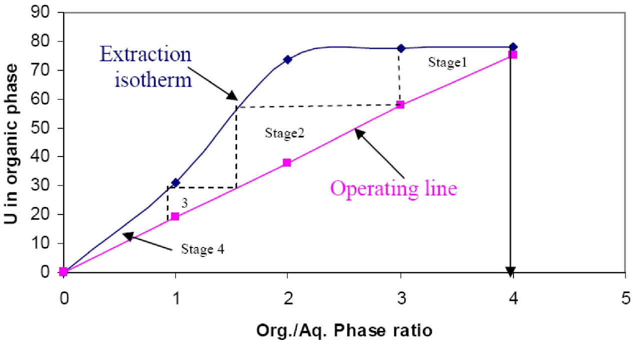

Figure 6: McCabe-Thiele diagram for uranium (VI) extraction

uranium was obtained. Care must be taken to keep the same pH value of 1.5 throughout the series of shake-outs. The data were used to construct the equilibrium curve by plotting the uranium (VI) concentration in the organic phase against its concentration in the aqueous phase. The next step is the construction of McCabe-Thiele diagram. From Figure 6 it can be seen that uranium (VI) extraction under these conditions needs only four extraction stages for nearly complete recovery.

Synergism and mixed extractants

The major objective in using mixed extractants for the extraction of metals is to take advantage of any synergism that may result. Synergism may be defined as “cooperative action of discrete agencies such that the total effect is greater than the sum of the effects taken independently. The reverse of this effect is known as antagonism. Many mixed extractant systems have been studied in attempts to take advantage of this effect, especially for the extraction of actinide elements in the nuclear industry. Many striking examples of synergism have been found. For example, the addition of only 0.003 M tributyl phosphate (TBP) to thenoyl trifluoroacetone (TTA) increases the extraction coefficient of U (V^2) by a factor of 5000 from 0.01 M nitric acid solution. During the last decades, other mixed systems have also been studied, including LIX 63-carboxylic acids, LIX 63-amines, LIX 63-D2EHPA, Kelex100-carboxylic acids, and many others[31].

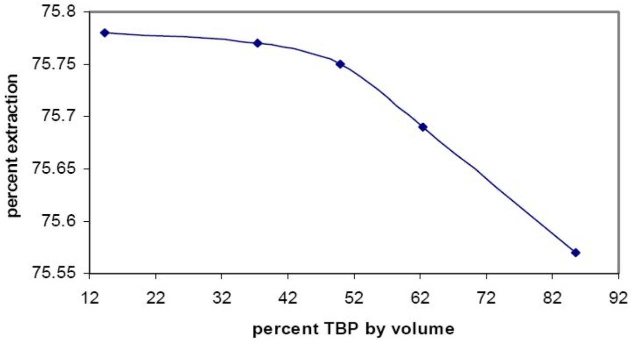

Figure 7: Effect of concentration of TBP on the extraction of uranium (VI)

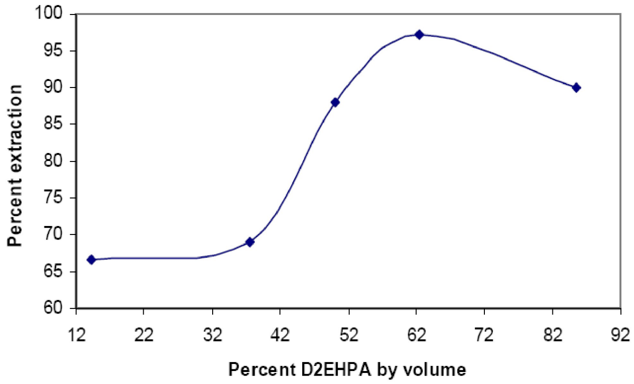

Figure 8: Effect of concentration of D2EHPA on the extraction of uranium (VI)

It certainly appears that mixed extractant systems can offer much in the way of enhanced separation of metals, and increase solvent loading. Another purpose for the use of mixed extractant systems is in the simultaneous extraction of both a cation and an anion.

Effect of TBP (tributyl phosphate) on the extraction of uranium (VI)

Series of solutions of TBP from 0.30 - 0.05 M in cyclohexane were added to series of TOPO/cyclohexane from 0.05 – 0.30 M respectively. Figure 7 shows that upon increasing the percentage of TBP added there was a decrease in the extraction of uranium (VI); indicating TBP is an antagonist in this system.

Effect of D2EHPA on the extraction of uranium (VI)

Series of solutions of D2EHPA from 0.30 - 0.05 M in cyclohexane were added to series of TOPO/cyclohexane from 0.05 – 0.30 M, respectively in different concentrations. Figure 8 shows that by increasing the percentage of D2EHPA added there was increase in the extraction of uranium (VI). This means that D2EHPA is synergist at lower concentrations (0.15M).

Effect of different stripping reagents

The loaded TOPO/D2EHPA /in cyclohexane was

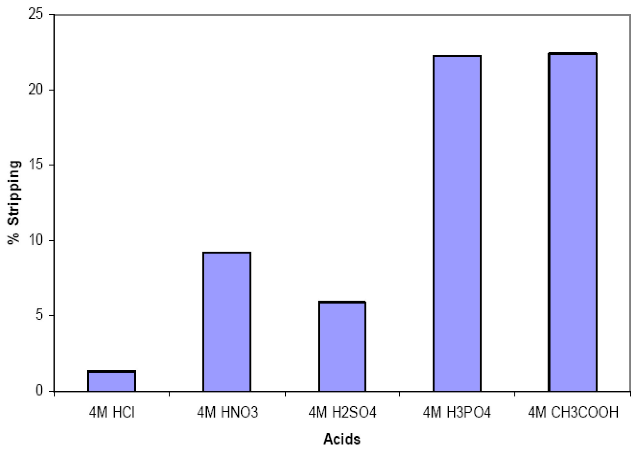

Figure 9 : Effect of different acidic reagents on the stripping of uranium (VI)

mixed with different stripping aqueous solutions, which may be acidic, basic, or even neutral to find out the most suitable reagent for re-extraction of uranium (VI).

Acidic stripping

Five different acidic reagents namely HCl, HNO_3, H_3PO_4, H_2SO_4 and CH_3COOH in the same concentrations were tried to find out the most suitable reagent for stripping uranium (VI) at an aqueous/organic phase ratio of 1:1.The obtained results are shown in Figure 9.

Alkaline stripping

Sodium hydroxide and sodium carbonate (0.5 M) were chosen as basic stripping reagents, being both more effective than any of the acidic reagents. The obtained results are shown in Figure 10.

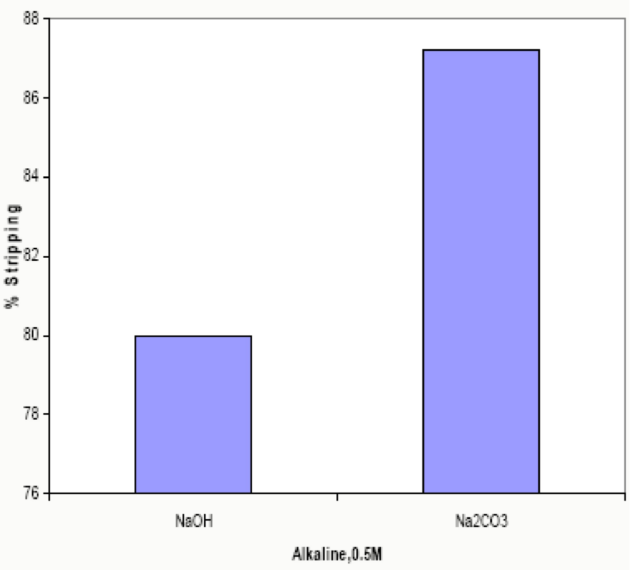

Figure10: Effect of different alkaline reagents on the stripping of uranium (VI).

Neutral stripping

Sodium chloride and sodium sulphate (0.5M) were chosen as neutral stripping reagents, being both less effective than any of the acidic and alkaline reagents. The obtained results are shown in Figure 11.

It is clear from the obtained results that Na_2CO_3 is the best reagent for stripping uranium (VI), moreover it is cheapest.

Effect of different sodium carbonate concentrations on the stripping of uranium (VI)

Series of stripping experiments were carried out, using Na_2CO_3 solution of concentration ranging from 0.1 to 1 M. The obtained results are shown in Figure 12. It is clear from the obtained results that stripping by 0.75M Na_2CO_3 gives the best uranium (VI) re-extraction percent. The contact time was 20 minutes and phase ratio (4:1) (v/v) organic / aqueous phase ratio.

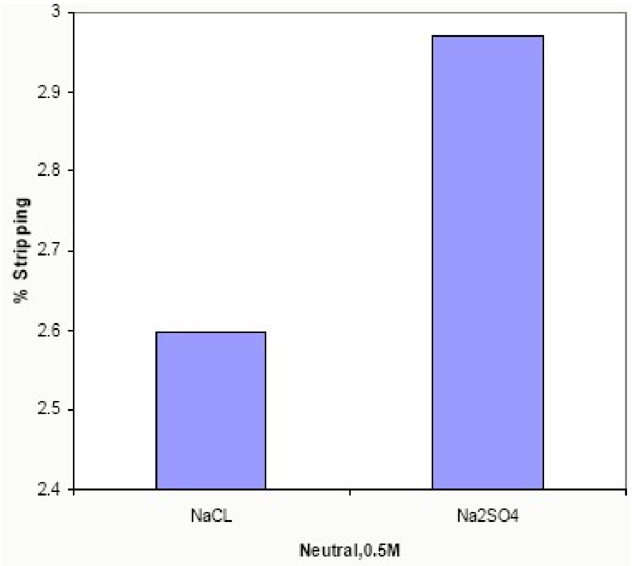

Figure 11: Effect of different neutral reagents on the stripping of uranium(VI)

So the next step is applying these new modifications on the chemical procedure of alpha particle spectrometer.

Description of the complete radiochemical separation

Uranium separation and purification

The uranium was separated from the matrix elements using TOPO and D2EHPA as extracting agents. Firstly, the aqueous sample solution is transferred into a separator funnel and shaken for 20 minutes with 100 ml of 0.25M TOPO and 0.15M D2EHPA dissolved in cyclohexane and this step gives two phases; organic (contains most of U-content) and aqueous phase. After separation of two phases, repeat shaking for another 20 minutes. After separation, the organic phase is transferred into another separating funnel. By these steps, Np, Pa, U and Th elements are extracted in the TOPO and D2EHPA phase while other matrix elements like Na, K, Mg, Ca and Al were not extracted and remain in the aqueous phase (step 3, Figure 13).

The uranium of the organic phase is re-extracted to the aqueous phase by using 0.75M Na_2CO_3 and then purified by washing this phase using $CHCl_3$ (step 4,

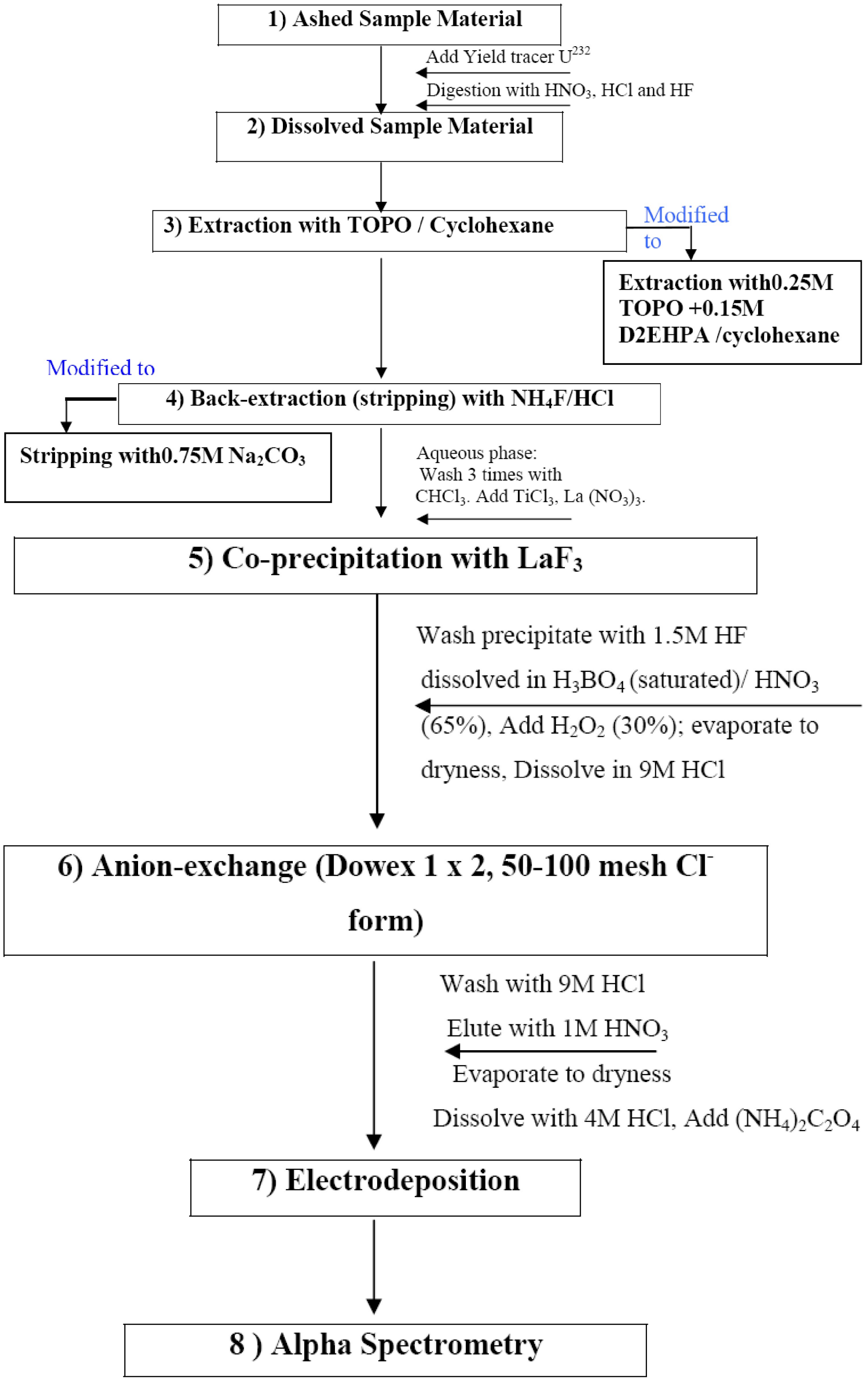

Figure 13 : Pre-determination flowchart of uranium isotope radiochemical analysis

Figure13). The obtained uranium U (VI) is reduced again to the tetravalent state U (IV) by adding a reducing agent ($TiCl_3$). The solution is co-precipitated by LaF_3 (25 mg/ml of La $(NO_3)_3$ with HF 40%), (step 5, Figure13). Then, the solution is centrifuged and the formed precipitate is dissolved in hot boric acid (saturated solution) and HNO_3. The uranium is re-oxidized to the hexavalent state by adding H_2O_2. This is followed by evaporation of the solution to dryness and the obtained residue is dissolved in 9M HCl, and then passed through a conditioned Dowex 1x2, 50-100 mesh, anion exchange column (15 cm long; 8 mm inner diameter) at a rate of 1 ml/ minute. To elute U of the column, 0.5M of HNO_3 is passed through the column and adding 1 ml of concentrated HCl to the eluted U, finally the evaporation process was carried out until dryness in a crystallizing dish (step 6, Figure13).

RESULTS

The radiochemical procedure has been validated using a soil reference material (IAEA-312). Once checked its proper operation, the procedure has been applied to the three sediment samples above described and the obtained results in terms ^{238}U and ^{234}U isotopic activity concentration (Bq kg^{-1}) for the studied samples are listed in TABLE 1. Also, the table shows the comparison between the old obtained results of α-spectrometry of NMA (Nuclear Materials Authority) by khattab 2011[32] and the new obtained result of α- particle spectrometry of NMA on one hand and the reported result given by IAEA (International Atomic Energy Agency)[33]. In which the obtained results in this work show harmony and good agreements with those certified by IAEA for the same reference soil sample, in which the results revealed that the activity concentration of ^{238}U in the soil sample (S-3) given by α- particle spectrometry of NMA in this work is 112 ± 1.3 Bq kg-1 and that reported by IAEA for the same U isotope in the same sample is 105.4 Bq kg-1. The activity concentration of ^{234}U in the same sample given by NMA α- particle spectrometry is 97.85±1.9 Bq kg-1 and that reported by IAEA for the same U isotope is 99.2Bq kg-1. This harmony in the results between the new modification that has been done in this work and the results of IAEA is due to the improvement has been done in the uranium extraction stage due to the presence of a little amount of 0.15M D2EHPA as a synergist which is acidic extractant and has highly loading capacity which help in the increase in the extraction than TOPO alone and also the extraction was carried out four stages for complete recovery and using 0.75M Na_2CO_3 as stripping agent and also changing of mixing and stripping time of the process, all these help in the improvement which leads to this harmony in the results between the results reported by IAEA and the new results reported by NMA, in which the whole procedure controlled by adding ^{232}U tracer for chemical yield and activity calculation. After the insurance of the validation of the new procedure, it was applied to three stream sediment samples which give results for JJ/600E (U) sample an activity concentration of ^{238}U is 11000±2.2 Bq kg-1 and for ^{234}U is 4621.8±1.6 Bq kg-1and the results of ^{238}U for the S/300E (U) sample is 7557.0±1.5 Bq kg-1 and for ^{234}U is 6058.3±1.7 Bq kg-1and the results of ^{238}U for DD/300W sample is 35402.3±2.3 Bq kg-1and for ^{234}U is 38850.27±2.5 Bq kg^{-1}. The results arises after modification gave a much better values than done before in the old method either done by NMA or by EAEA in the soil reference sample (IAEA-312), in which the sample was complete dissolved and the same dissolution procedure that is used in old measurements

TABLE 1 : Activity concentration of U-isotopes (Bq kg^{-1}) in the analyzed samples

Sample No	U- isotopes	Present Work at NMA	EAEA result	The Old NMA result	IAEA result
S-3	^{238}U	112.0 ±1.3	88.96 ± 2.1	75.64 ± 3.4	105.4
	^{234}U	97.85 ±1.9	60.22 ± 1.7	55.8 ± 2.8	99.2
JJ/600E (U)	^{238}U	11000 ±2.2	--	-	--
	^{234}U	4621.8 ±1.6	--	-	--
S/300E (U)	^{238}U	7557.0 ±1.5	--	--	--
	^{234}U	±1.76058.6	--	--	--
DD/300W	^{238}U	±2.335402.3	--	--	--
	^{234}U	2.5± 38850.27	--	--	--

Note: NMA result means the obtained result using α- particle spectrometry of Nuclear Materials Authority; EAEA result means the obtained result using α- particle spectrometry of Egyptian Atomic Energy Authority; IAEA result means the recorded data for the standard samples; JJ/600E (U), S/300E (U) and DD/300W are stream samples from Wadi El-Reddah area, and S-3 (IAEA 312) is soil sample from IAEA.

except the solvent extraction stage has been changed in the new method which increase the efficiency of the whole procedure and this reflected in the results arises after modification in a good agreement up to 98%, in which old method of both of NMA and EAEA results revealed that activity concentrations of ^{238}U in the soil reference sample are 75.64±3.4, 88.96±2.1 Bq kg-1 respectively and the activity concentration for ^{234}U in the same sample is 85.8±2.8, 60.22±1.7 Bq kg-1 respectively.

CONCLUSION

- The extraction for uranium increased after the modification that has been done on radiochemical procedure for alpha spectrometry. Where the main differences between the old method and the new one are summarized in:
  1)0.25 M TOPO + 0.15 M D2EHPA/cyclohexane while in the old method was 0.2MTOPO/ cyclohexane.
  2)pH 1.5
  3)Contact time 20 min while in the old method was 15 min.
  4)Aq/Org. phase ratio 1:1
  5)Four stages for complete recovery while in the old method were only two stages.
  6)0.75 M Na2CO3 as stripping agent while in the old method was 1M NH4F/0.1MHCl.
- The reasons for the improvement in the uranium extraction were mainly due to the presence of a little amount of 0.15M D2EHPA which is acidic extractant that is characterized by highly loading capacity that help in the increase in the extraction percent than TOPO alone. After validation of the modification using soil reference material which gives a good agreement between the results obtained in this work and those certified by IAEA.

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