Liquid-Liquid Extraction of Uranium from Nitric Acid Solution Using *N*, *N*-Di(2-ethyl- hexyl) hexanamide (DOHA) in Dodecane as Extractant

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Summary: This work describes the liquid-liquid extraction of uranium (U) from 3 mole L^{-1} nitric acid media by *N*, *N*-di(2-ethyl hexyl) hexanamide (DOHA) in dodecane and subsequent determination of uranium using arsenazo III as a chromogenic reagent. Different factors (nitric acid, DOHA, metal ion concentration, salting out effect, temperature and shaking time) affecting the extraction /stripping process, were investigated. The investigation showed that the extraction is independent of the conc. of metal ions in the range of 5 to 300 µg mL^{-1} and inversely dependent upon the temperature. The desired species so extracted were found to have a stoichiometric composition as $UO_2(NO_3)_2$.DOHA and $UO_2(NO_3)_2$.2DOHA at lower and higher concentration of extractant respectively. Uranium could be stripped back from organic phase by using 0.1 mole L^{-1} ammonium hydroxide (NH_4OH) and the extraction was found to be quantitative.

Keywords*:* Uranium; Extraction; DOHA; Nitric acid; Arsenazo III.

Introduction

Uranium has been found in ores and minerals and is a vital element for nuclear energy production [1, 2]. Some other uses of uranium are radiometric dating of metals, as target for producing high energy X-rays and in inertial guiding devices [3]. For these and other purposes, a high purity metal is required. In recent past, a large number of methods have been reported for the extraction of uranium from different ores and among them hydrometallurgical processes have received much attention due to their advantages: (a) simple apparatus (b) less time consuming (c) process operates at room temperature and does not need an energy input [4-9]. In addition to this, hydrometallurgical processes have large scale applications in synthetic, pharmaceutical and petrochemical industries [10]. However, most of these processes use tri-butylphosphine (TBP) as an extractant and encounter problems such as large random errors, low selectivity, time consuming, labor-intensive, involve multi-step procedures and use of large volumes of toxic organic solvents [11-18].

Monoamides *(N*, *N*-dialkylamides) have been proposed as alternative extractants to overcome the above mentioned problems. These monoamides have the advantages such as: (a) easy large scale synthesis (b) their better incinerability which reduces the volume of secondary nuclear waste (c) less harmful nature of their radiolytic degradation products (mainly *N*-alkylamide, *N*, *N*-dialkylamine, and carboxylic acids). In addition to this, amides contain the elements such as C, H, N and O and thus could easily be converted into respective gaseous products. Furthermore, monoamides afford better separation factors for the metals such as thorium and uranium. Also, the monoamides containing branched alkyl chains have been proved as excellent extractants for the extraction of uranium [19-36]. These attractive properties of monoamides make them better alternative extractants as compared to TBP which produces the dibutyl phosphate and monobutyl phosphate during radiolysis and these products are responsible for the formation of third phase or precipitation. Furthermore, monoamides could be diluted by aliphatic hydrocarbons used in commercial reprocessing plants.

Most of the solvent extraction procedures developed until now use the nitric acid as aqueous phase along with different extractants [35-38]. Uranium extraction with *N*, *N*, *N*, *N*-tetrabutyl-dipincamide (TB-AA) and *N*-alkyl carboxylic acid amides in nitric acid medium increased with increasing the acid concentration [39]. The amides such as *N*, *N*-dibutyldecanamide (DBDEA) and *N*, *N'*-dioctylsuccinyl-amide, *N*, *N*, *N*, *N'*-tetraoctyldiglycolamide (TODGA) have also been applied for the extraction of uranium from the nitric acid medium [39, 40]. A continuous current experiment has been made using *N*, *N*-di(2-

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ethylhexyl)butanamide (DEHBA) as an extractant for the extraction of uranium and plutonium. [12]. The extractant (DEHiBA) has been found advantageous for the selective extraction of uranium from spent nuclear fuels (GANEX process) [12]. The monoamide extractants such as *N*, *N*-di(2-ethylhexyl)butanamide (DEHBA) and *N*, *N*-di(2-ethylhexyl)hexanamide (DEHHA) having a linear alkyl chain on carbonyl carbon atom have been found efficient for the liquid-liquid extraction of uranium at low pH(1-3) in the presence of Pu(IV) [12]. Ahmad Hosseini *et al* have used a stable extractant impregnated resin (EIR) using a polymeric support (Amberlite XAD-2010). This EIR has been used for trace separation/preconcentration of U(VI) ion followed by spectrophotometric determination with arsenazo-III. Amberlite XAD-2010 exhibited good selectivity for U (VI) ions in the presence of other ions [41]. Recently R. S. Tegure *et al* have developed a novel method for the extraction of U (IV) using Amberlite XAD-4 with extractant *iso*-nitroso-4-methyl-2-petanone. They have made quantitative extraction of uranium at pH 4 [42].

The easy synthesis on bulk scale and inspired solvent extraction results of monoamides, attracted us to use *N*, *N*–di(2-ethyl hexyl) hexanamide (DOHA) for the liquid liquid extraction of uranium and to the best of our knowledge this extractant has not been applied for the solvent extraction of uranium so far. We have used the dodecane as a diluent which has the advantages such as low cost, environment friendly and maximum uranium extracting efficiency [43]. The basic purpose of our study was to develop an efficient and low cost method for the rapid extraction of uranium from spent over nuclear fuels and as well as from uranium ores. We also have investigated the different factors affecting the uranium extraction such as nitric acid concentration, DOHA concentration, metal ion concentration, salting out effect, temperature and shaking time.

Experimental

Reagents and Solutions

The analytical grade (Merck, Germany) chemicals were used for this research work. Technical grade *N*, *N*-di(2-ethyl hexyl) hexanamide (DOHA, 97%) was synthesized with a reported method [37]. Deionized water was used to conduct these experiments. Analar grade disodium salt of arsenazo- III and perchloric acid (70 to 72 %) were used. Stock solution (10 mole L^{-1}) of percholoric acid was prepared by dilution from 11.7 mole L^{-1} $HClO_4$. Uranium stock solution (1000 µg mL^{-1}) was prepared from analytical grade uranyl nitrate in deionized water and further dilutions were made when required. The solution of arsenazo-III (0.01% w/v) was prepared in $HClO_4$ (10 mole L^{-1}). The stock solution of HNO_3 (10 mole L^{-1}) was also prepared and further dilutions were made when required. DOHA solution (1 mole L^{-1}, 50 mL) was prepared by adding 17 g of DOHA in dodecane. Aluminium nitrate (salting out agent) solutions (0.25 , 0.5, 0.75, 1, 1.5, and 2 mole L^{-1} each) were prepared by adding its 1.5 g, 2.8 g, 4.2 g, 5.6 g, 8.7 g, and 11.3 g in 15 mL of HNO_3 (3 mole L^{-1}) respectively. The stripping agents (0.1 mole L^{-1} solutions of NaOH, $NaHCO_3$, NH_4OH) were prepared by adding 0.2 g, 0.4 g, 0.5 g of the respective compound in deionized water.

Apparatus and Instrumentation

Solvent extraction experiments were carried out in pyrex glass culture tubes (16x125mm). Griffin and George Wrist action Mechanical shaker (SF-1) of Stuart, UK and Gallenkamp Thermostirr 100, Model BKL, 235 water bath were used. UV/ Visible Spectrophotometer Model-1601 (Schmadzu, Japan) was employed for absorbance measurements. Bench top centrifuge machine centaur 2 (46000 rpm) was used for centrifugation.

General Extraction Procedure

Three mL of nitric acid (3 mole L^{-1}) were taken in a pyrex glass culture tube. A known amount of uranyl nitrate was added into it and mixed thoroughly and then 1mL of this solution was pipetted out in a 50 mL pyrex glass beaker as reference before extraction. The remaining 2 mL were equilibrated with an equal volume of DOHA (1 mole L^{-1}) in dodecane for 10 min. Phase separation was achieved with centrifugation and 1 mL from aqueous portion was pipetted out in another beaker of the same size. Both solutions (before and after extraction) were heated to near dryness on an electric hot plate and the contents were dissolved in $HClO_4$ (2 mL) and again heated to near dryness to avoid organic traces. On cooling, 5 mL of chromogenic solution (0.01 % arsenazo-III in $HClO_4$, 3 mole L^{-1}) was added and mixed thoroughly for color development. Uranium was determined spectrophotometrically at 651 nm using the reagent as blank [32]. The distribution coefficient (D) and percent extraction were calculated using the following relationship:

$$\mathbf{D} = \frac{\textbf{Concentration in organic phase}}{\textbf{Concentration in aqueous phase}} \mathbf{x} \frac{\mathbf{Vaq}}{\mathbf{Vorg}}$$

$$\% \text{ Extn} = \frac{D \times 100}{D + \frac{V_{aq}}{V_{org}}}$$

where V_{aq}/V_{org}, refer to the volume of aqueous and organic phases respectively.

Results and Discussion

Optimization of Parameters

The extraction of uranium with DOHA (1 mole L^{-1}) in dodecane has been carried out from nitric acid (1 to7 mole L^{-1}) at room temperature (Fig. 1). The percentage extraction was found to increase with increasing acid concentration, giving a maximum value of 89.89 % with 7 mole L^{-1} of HNO_3. Such an increase in percentage extraction is possibly due to co-ion (nitrate ion) effect and salting out effect. On further increasing the acid concentration, the percentage extraction decreased. Such a decrease in the percentage extraction could also be rationalized that at lower acid concentration both acid (HNO_3) and extractant (DOHA) have nearly equal possibility to make the adduct/complex with the uranium metal center but in case of higher acid concentration, the probability of complex formation with the extractant is lower which results into decreased percentage extraction. This effect of nitric acid concentration on the extraction of uranium has a similar trend as already reported for this metal. In previous studies, maximum uranium extraction was noted with 5 mole L^{-1} HNO_3 [8]. Y. S. Wang *et al* have used *N*, *N*, *N*, *N*-tetrabutylsuccinylamide (TBSA) as extractant and found 1 to 6 mole L^{-1} HNO_3 concentration, beneficial for uranium extraction [32]. C. Yu *et al* have studied the extraction of U (VI) using DMDOSA as extractant and found that extraction of metal ions increases with an increase in nitric acid concentration [35].

The results of pre-equilibrated organic phase showed that extraction of uranium metal with DOHA (1 mole L^{-1} in dodecane) was higher (96.28 %) at 7 mole L^{-1} nitric acid as compared to the non-equilibrated phase (89.89 %) (Fig. 2). These findings are in close agreement with the general reported behavior of the neutral extractants [27]. In Purex Process 3 mole L^{-1} nitric acid has been used. Our attempt was to develop an alternative process keeping all the conditions of Purex process except we have used DOHA as extractant instead of TBP. Keeping in mind the economic use of nitric acid, we performed further extraction experiments with its 3 mole L^{-1}. In previous studies, it was noted that extraction of Hf with pre-equilibrated organic phase was higher as compared to the non-equilibrated one [37].

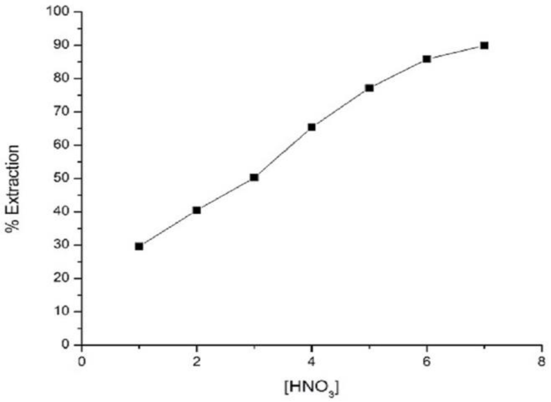

Fig. 1: Extraction of uranium (100 µg mL^{-1}) as a function of nitric acid concentration (1-7 mole L^{-1}) with DOHA (1 mole L^{-1}) in dodecane.

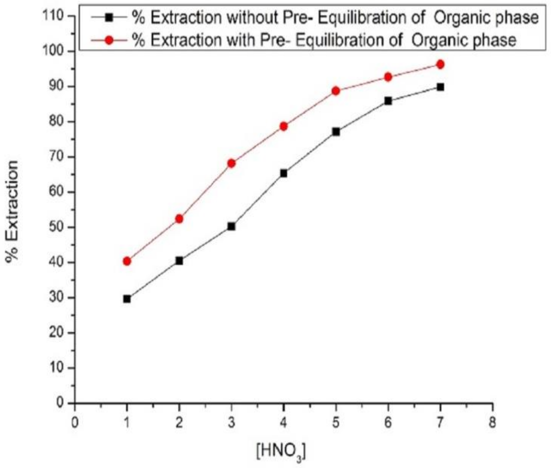

Fig. 2: Percentage extraction with pre-equiliberated organic phase and without pre-equiliberated organic phase (DOHA = 1 mole L^{-1}, uranium = 100 µg mL^{-1}, nitric acid = 3 to 7 mole L^{-1}).

Effect of Extractant Concentration

The effect of extractant concentration on fixed amount of uranium (100 µg mL^{-1}) has been studied by varying the concentration of DOHA (1 to 3 mole L^{-1}) using dodecane as diluent (Fig. 3). It was found that the extraction of uranium increased with increasing extractant concentration and was found maximum (93.3 %) with 3 mole L^{-1} extractant concentrations. Thus, considering cost aspect, DOHA (1 mole L^{-1}) was selected as optimum concentration for further experimental work; however, the extracting efficiency was enhanced by using salting out agent [$Al(NO_3)_3$]. In previous studies, extractant concentration effect was studied by using fixed amount of uranium and changing the concentration of dibutylsulfoxide (DBSO 0.3 mole L^{-1} to 1.54 mole L^{-1}) and it was noted that by increasing extractant concentration, the uranium extraction also increased [8]. Orabi has used TBP solution (10 to 90 % in kerocene) for the extraction of U(VI) [9]. The percentage extraction increased with increasing TBP concentration and maximum was observed with its 25 % solution [9]. As our method affords 93 % uranium extraction with 3 mol L^{-1} of DOHA. Keeping in mind the per mole cost of TBP and DOHA, the method we are reporting herein is cost effective as compared to the method developed by Orabi [9].

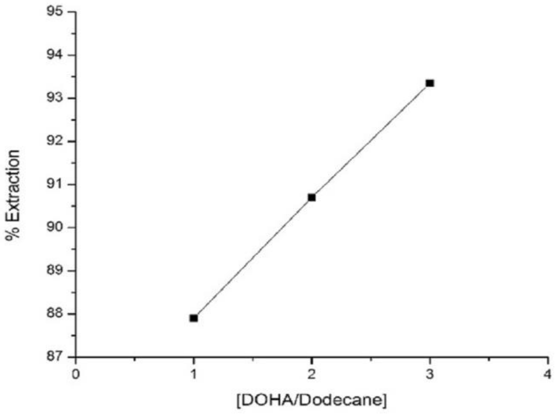

Fig. 3: Relationship between concentration of DOHA and percent extraction without pre-equilibrated phase (DOHA concentration = 1-4 mole L^{-1}, HNO_3 = 3 mole L^{-1}, metal ion concentration = 100 µg mL^{-1}).

Effect of Pre-equilibrated Extractant

To check the influence of the pre-equilibration of extractant on the extraction, the organic phase (DOHA, 1 to 3 mole L^{-1}) was equilibrated with an equal volume of HNO_3 (3 mole L^{-1}) by mechanical shaking for 5 min. It was observed that extraction increased upto 96.07 % by increasing extractant (DOHA) concentration to 3 moles L^{-1} (Fig. 4). The data clearly indicated that after pre-equilibration, there is slight increase in percent extraction and such a behavior may be due to the extraction saturation of DOHA molecules with nitric acid and consequently lacking enough space to accommodate uranium ions more effectively. Extraction of uranium as a function of nitric acid concentration (1 to 6 moles L^{-1}) using 0.5 mole L^{-1} D2EHIBA and 5 % TBP as the extractant either pre-equilibrated or without pre-equilibration with respective acid solution showed that there was a minor increase in Kd value in the acidity range (1 to 3 mole L^{-1}) and there after a decrease was observed [20]. The effect of varying nitric acid concentration (1to 6 mole L^{-1}) using DBSO (1 mole L^{-1}) in petroleum ether by equilibrating equal volumes of both organic and aqueous solutions showed that Kd value increases sharply and attained a maximum value at 2 mole L^{-1} HNO_3 and decreased sharply up to 4 mole L^{-1} of HNO_3 concentration [10].

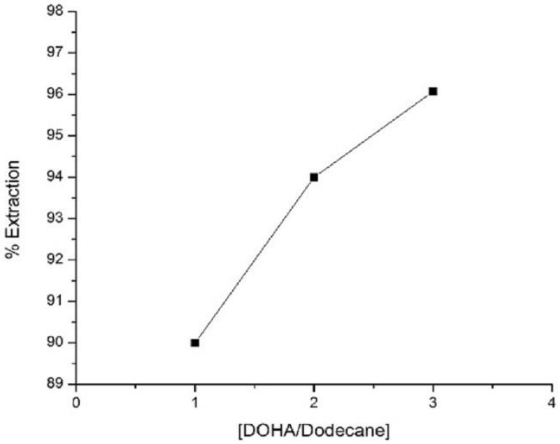

Fig. 4: Effect of DOHA concentration (1 to 3 mole L^{-1}) on the extraction of U(VI) (0.01 mol L^{-1}) from 3 mole L^{-1} HNO_3.

Effect of Salting-out Agent

Aluminium nitrate solutions (0.25, 0.5, 0.75, 1.0, 1.5 and 2.0 mole L^{-1}) as salting-out agent were used regarding the extraction of uranium with DOHA (1 mole L^{-1} in dodecane) from HNO_3 (3 mole L^{-1}). The maximum percentage extraction (96 %) was observed with 2 mole L^{-1} of $Al(NO_3)_3$ and similar result was obtained with its 3 mole L^{-1} solution. Based upon economic use of chemicals, 2 mole L^{-1} aluminum nitrate solution was selected as optimum concentration for further extraction experiments. The increase in percentage extraction with the salting out agent is probably due to participation of nitrate ion in the metal solvation process. However, in less acidic solutions, nitrates enhance extraction by salting out and common ion effect. In previous similar studies, Orabi has used sodium nitrate ($NaNO_3$, 0 to 0.8 moles L^{-1}) as salting out agent for uranium extraction by TBP and found that percent extraction was enhanced with increasing $NaNO_3$ concentration [9]. Aluminum nitrate (salting out agent) was used for uranium extraction using DBSO (1 mole L^{-1}) in petroleum ether from nitric acid (2 mole L^{-1}) and the results indicated a small influence of nitrate ion on the percentage extraction [10].

Table-1: Effect of salting-out agent on the extraction of uranium (100 µg mL^{-1}) with DOHA (1 mole L^{-1}) indodecane from HNO_3 (3 mole L^{-1}).

Salting out agent: mole L^{-1}	Distribution ratio(D)	Extraction (%)
0.25	2.6	72.5
0.50	3.3	77.0
0.75	4.9	83.0
1.00	7.7	88.6
1.50	13.0	93.0
2.00	24.6	96.0

Effect of Metal Ion Concentration

Uranium was extracted in the range of its concentration from 5 µg mL^{-1} to 300 µg mL^{-1} under optimum conditions (HNO_3 = 3 mole L^{-1}, DOHA = 1 mole L^{-1}, T=25 °C). At low metal ion concentration, the extraction was found to be lower due to more competition of extractant for metal and nitric acid. However, it increased with increasing metal concentration upto100 µg mL^{-1} beyond which it then decreased (Fig. 5).This decrease in the extraction may be due to the change in activity coefficient of metal ions and probably due to self salting out effect. These investigations show a high loading potential of extractant and the extraction system reported herein could be applied for bulk separation of uranium in the range studied. Previously, uranium was extracted in the range of its concentration 7 x 10^{-5} to 1x 10 $^{-3}$ µg mL^{-1} under optimum conditions and it was found that at lower concentration, the extraction was lower, however, it initially increased and after that started decreasing due to change in activity coefficient [10].

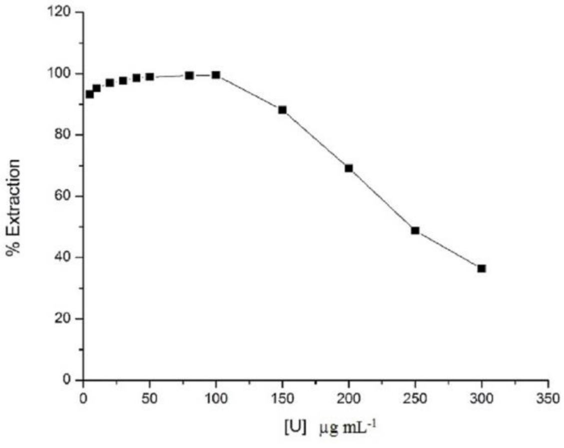

Fig. 5: Effect of metal ion concentration on extraction of uranium (DOHA =1 mole L^{-1}, HNO_3 = 3 mole L^{-1}).

Nature of the Extracted Complex

A graph was drawn between log [DOHA] and log D and two slopes were obtained for DOHA indicating that at its lower concentration, one water molecule was replaced while at higher concentration, both water molecules were replaced with DOHA resulting into two different complexes. At lower DOHA concentration, two molecules of DOHA coordinate with $UO_2(NO_3)_2$ forming $UO_2(NO_3)_2$. 2DOHA, complex (Fig. 6). At higher concentration of DOHA, we get a mixture of two types of complexes; one [$UO_2(NO_3)_2$. 2DOHA] containing two DOHA molecules while other [$UO_2(NO_3)_2$. 3DOHA] containing three molecules of DOHA (Fig. 7).

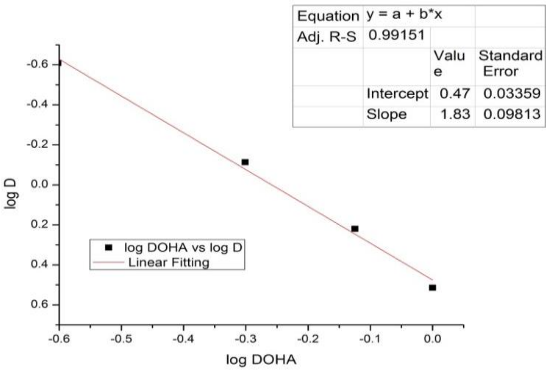

Fig. 6: Uranium complex formation at lower concentration of DOHA.

$UO_2(NO_3)_{2(aq)}$+$DOHA_{(org)}$→$UO_2(NO_3)_2$.2DOHA
(at lower concentration of DOHA)
$UO_2(NO_3)_{2(aq)}$+$2DOHA_{(org)}$→$UO_2(NO_3)_2$ 2DOHA + $UO_2(NO_3)_2$ 3DOHA
(at higher concentration of DOHA)

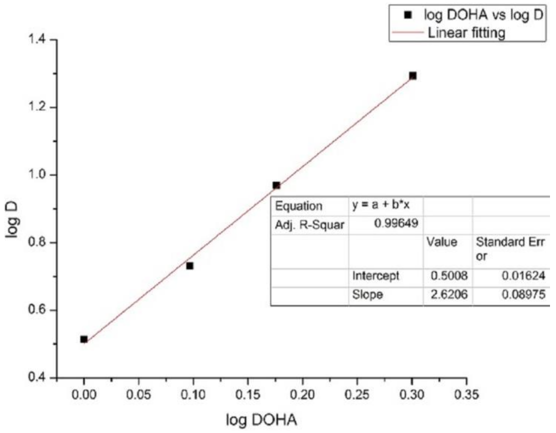

Fig. 7: Uranium complex formation at higher concentration of DOHA.

Y.S. Wang *et al* have used TBSA as an extractant for extraction of U(VI) and found that extractant and metal ion formed a 1:1 complex [32]. L. Nigond *et al* have used the amides such as *N, N, N, N*-tetraalkyl-2-alkyl propane-1, 3-diamides for the extraction of uranium and plutonium and found the extracted complexes ($L.UO_2(NO_3)_2$, $L.Pu(NO_3)_4$ and $L_2.Pu(NO_3)_4$) respectively [39]. K. K. Gupta *et al* have extracted the U(VI) and Pu(IV) from nitric acid medium using *N, N*-dihexylamide (extractant) and found the extracted complexes as $UO_2(NO_3)_2.2DHOA$ and $Pu(NO_3)_4.2DHOA$ in moderately acidic solutions [26].

Shaking Time Effect

The shaking time duration affects the extraction of uranium and has been investigated under optimized conditions. In order to optimize the equilibration time of uranium metal with DOHA (in dodecane) 1 to 10 min shaking time was studied. Quantitative extraction was observed within first 3 min. and later on by increasing the shaking time to 10 min, no significant change in extraction was observed (Fig. 8). This behavior could be explained on the basis of the fact that equilibration between uranium and DOHA attains saturation with a higher rate constant that no further increase in the extraction was found after 5 min shaking time. Consequently, 5 min shaking time was selected for further studies. Orabi have studied the effect of shaking time on the extraction of U(IV) using 25 % TBP in kerosene and found that 10 min. shaking time was adequate for extracting uranium (VI) efficiently. P. N. Pathak *et al* have used the extractant such as D2EHAA, D2EHPRA, and D2EHPVA for the extraction of U(VI), and Th(IV) and 2 to 60 min equilibration time was investigated and found that 15 min was sufficient for equilibration of organic and aqueous phases [5].

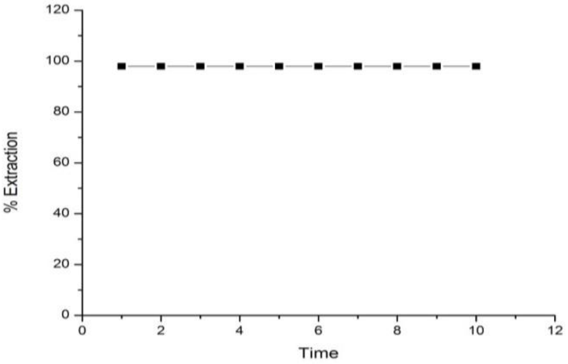

Fig. 8: Effect of shaking time on the extraction of uranium (DOHA =1 mole L^{-1}, HNO_3 = 1 mole L^{-1}, metal ion concentration = 100 µg mL^{-1}).

Effect of Temperature on the Extraction

The effect of temperature on the extraction of uranium was studied in the temperature range (15 to 45ºC) under optimized conditions (DOHA = 1 mole L^{-1}, HNO_3 = 3 mole L^{-1}, metal ion concentration = 100 µg mL^{-1}). The graph indicated that the extraction decreased with increasing temperature in the range studied (Fig. 9). The reaction was spontaneous and uranium complex formation was an exothermic process hence the extraction experiments were made at room temperature. In previous studies, the extraction of uranium decreased from 99 % (at 25 ºC) to 88 % (at 55ºC) [10]. The effect of temperature (15 to 45 ºC) on the extraction of uranium showed that extraction decreased by increasing temperature [37]. Y. S. Wang *et al* have used TBSA as an extractant for extraction of U(II) and found the interaction of extractant with the U(II) ion was an exothermic process [32].

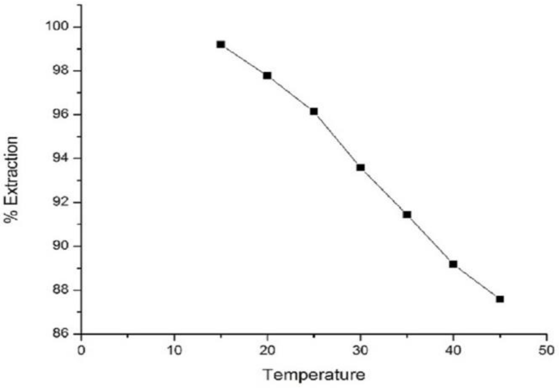

Fig. 9: Effect of temperature on the extraction of uranium (DOHA = 1 mole L^{-1}, HNO_3 = 1 mole L^{-1}, metal ion concentration = 100 µg mL^{-1}).

Study of Stripping Agents

The extracted uranium was stripped from organic phase by using different solvents/solutions such as deionized water and 0.1 mole L^{-1} (HNO_3, NH_4OH, $NaHCO_3$ and NaOH) solutions. Equilibration of organic phase with equal volumes of striping agents gave the recovery of uranium (Table-2). Maximum recovery of uranium (96.5 %) was achieved with NH_4OH. The NH_4OH solution has a higher pH which resulted into hydrolysis of UO_2. However, deionized water (percentage extraction= 93.2 %) was selected as stripping agent due to its extraction efficiency and economic consideration. The previous research results showed that equilibration of organic phase with an equal volume of NH_4OH, HNO_3 and deionized water for three min. ensures 97.8, 86.2 and 93.0 % recovery of uranium [10]. To recover uranium from organic phase, some stripping agents were used such as HCl, H_2SO_4, HNO_3, CH_3COOH and results showed that CH_3COOH and H_2SO_4 were more effective than HNO_3 and HCl [9].

Table-2: Stripping agents and respective % extraction.

Strippants	% Extraction
Deionized water	**93.2**
HNO_3	**87.6**
$NaHCO_3$	**92.4**
NaOH	**82.3**
NH_4OH	**96.5**

Conclusion

Quantitative extraction of uranium from 3 mole L^{-1} HNO_3 solution with DOHA (1 mole L^{-1} in dodecane) could be achieved. The extracted uranium could be stripped back in 2 steps using NH_4OH as stripping agent. The diluent (dodecane) is quite stable with high loading capacity and could be recycled without any degradation. The monoamides are better extractants due to their certain advantages such as their large scale synthesis and easy degradation into gaseous products. The extractant (DOHA) could easily be synthesized in a good yield. The extraction of uranium with DOHA is advantageous and economic as compared to its extraction with TBP. Furthermore, clean separation of uranium from ores is possible by employing this method. The method is simple, short step, convenient, rapid and economical. This method could be applied for extraction of uranium from ores and other materials when present as nitrate medium.

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