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Physico chemical study on extraction of uranium

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In extraction technology, the sample pretreatment and particle disruption is an important aspects for analysis of extraction efficiency of any blended chemicals. This analytical study can be performed with the propagation of high energy ultrasound due to its characteristic property like high frequency and high wavelength comparable to atomic and subatomic dimension. This paper describes the effect of ultrasonic wave and sonication effect for extraction of uranyl nitrate with optimum blended solvent methyl iso-butyl ketone (MIBK). The result of physico-chemical properties well establishes the supporting environment for extraction of uranium (IV) from uranyl nitrate using 40% of MIBK at room temperature and for the low frequency ultrasonic wave.

Keywords: Ultrasonic velocity, Extractant, Diluents, Intermolecular interaction, Acoustic parameters

1 Introduction

In the past few decades due to the ever increasing demand of power sources, nuclear energy is going to be one of the most important resources of energy. Generally nuclear fuels containing fissile materials make them more useful operating reactors. Among all the nuclear reactor materials uranium is enrich of this fissile materials. Due to such reason it is necessary to recovery of uranium from various sources[1]. In the ground of separation and purification sciences, for the recovery of different metal ions, some commercial solvent extractants like tri-n-butyl phosphate (TBP), methyl-isobutyl ketone (MIBK), methyl ethyl ketone (MEK) and di-isobutyl ketone etc. have high utility in that particular field[2]. There are several methods which are used for the extraction of the nuclear reactor materials, among which we have chosen the ultrasonic assisted solvent extraction process[3-5] to achieve the task because it is a very economical way of getting uranium from the sources.

During solvent extraction process[6] splitting of the organic phase into two different phases of different viscosities and densities, encounters various major problems as a result of which the extraction efficiency reduces as shown in Fig. 1. During the process, occurrence of an intermediate phase generally known as third phase has been reported by many authors[7-11].

Removal of this third phase by adding appropriate diluents/modifiers to the solvent by obtaining a particular concentration of the mixture is a severe concern in this field. By using ultrasonic technique we can study different physico-chemical properties of the binary mixture. Sound wave parameters like compressibility, impedance, intermolecular free length and surface tension have been applied to understand the behavior of the intermolecular interactions present in the solvent mixture[12]. The transmission of sound wave of different frequency mostly ultrasonic wave in a liquid medium generates high energy due to cavitations process and this ultrasonic energy can be used to break chemical bonds to initiate new reactions. The basic acoustic forces occur in the treatment medium during propagation of ultrasonic wave that are well explained the fundamental concepts of interaction of a wave with a medium which may be a liquid, solid or a gas. In the current paper the basic mechanism of ultrasonic wave is applied to identify the different type of interactions present in a liquid medium due to which the extraction of nuclear material like uranium is well defined by variation of different sound properties[13]. Among all the extractant MIBK and toluene are the versatile solvents for extraction in the nuclear industry. The blend which was used in our extraction study (MIBK with toluene) is very much compatible and giving good results for extraction of uranium. MIBK is a mildly associated liquid[14,15]. Because of parallel dipolar alignments, the intramolecular alliance among MIBK molecules results in multimerization and in case of binary mixtures

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Fig. 1 — Precipitation of uranyl nitrate and third phase formation.

connecting two polar constituents, intermolecular alliance among distinct molecules results in formation of micro- heterogeneous clusters. The computation of acoustic parameters from the data of ultrasonic velocity enables to understand the physico- chemical natures of the blended solvent mixtures. Again the effect of sonication time, frequency and temperature are discussed in the extraction process of organo-aqueous mixture of uranyl nitrate.

2 Experimental Details

2.1 Reagents

Commercially existing products like methyl isobutyl ketone (MIBK) (AR,> 98%), toluene (AR,> 99%), nitric acid (HNO_3, AR, 15.5 mol/L) and uranyl nitrate (AR,> 99%) are obtained from CDH Chemicals. These are highly pure and analytical grade samples. All the chemicals were used as received without any further purification.

2.2 Instruments used for experiment

The binary mixture of MIBK and toluene were equipped on percentage basis (w/w) by adding appropriate concentration of MIBK with toluene. A high accuracy electronic balance (WENSAR, PGB 100(±0.001 gm)) was used to measure their masses. Using a bicapillary pyknometer the densities of the pure liquids as well as all the mixture were measured which was standardized with deionised double distilled water of density 0.9960×10^3 kg/m^3 for different temperature. The precision of density measurement was within ± 0.0001 kg/m^3. The sound velocity of the solvent mixtures were measured by a single crystal variable path multi frequency ultrasonic interferometer (Mittal Enterprises, New Delhi, Model-M-81S) for a constant frequency (2 MHz) and various temperature range 303.15 K, 308.15 K and 313.15 K. To control the temperature of the liquid mixture a thermostatically regulated constant temperature water bath (B-206) was connected with the interferometer which was kept constant within ± 0.01 K.

The measurement of velocity for the liquid mixture was fixed up to ± 0.01 m/s. Viscosities of the mixtures were evaluated using Redwood viscometer by obtaining the time of flow and the calculated values of viscosities were precise up to ± 0.0001 Ns.m^{-2}. During the extraction procedure of uranium, a magnetic stirrer (MAC, Model-#MSHP-2) and an ultrasonicator bath (EMCOLITE-Ultrasonics, 125 MHz) were used for preparation of organic and aqueous phase.

3 Methodology

A set of binary mixture was prepared of methyl isobutyl ketone (MIBK) and toluene for different mole fraction of MIBK ranging from 0.1 to 0.9 for every case and stored within ground-glass stopper bottles to avoid evaporation and contamination of the prepared samples. Using ultrasonic interferometer optimum concentration of the solvent mixture was found out in terms of studying different intermolecular interactions. Organic phase was equilibrated by adding required concentration of MIBK with toluene of various concentration and aqueous phase was equilibrated by putting uranyl nitrate in diluted nitric acid (2 mol/dm^3) with increasing concentration of uranium (10, 15, 25, 30, 45, 50) g/dm^3 at room temperature. Both the phases were subjected to a magnetic stirrer separately for 10 min to make sure that the two phases were fully mixed. Distribution studies were performed by adding equal volumes of aqueous phase with organic phase (phase ratio=1:1) in a beaker. Again the biphasic system was placed inside the ultrasonicator bath (125 kHz frequency) for dispersion. Then the mixture was put in a separatory funnel and allowed it to stand for 15 min at room temperature for phase settle. The extract was filtered through Whatman filter paper No. 42 from the aqueous phase. In Fig. 2 total experimental procedure is presented by a schematic diagram.

The mechanism of the uranium extraction by MIBK is given by the equation below:

$$UO_2^{2+}(aq.) + 2NO_3^{-}(aq.) + 2MIBK\ (org.)$$
$$\leftrightarrow UO_2(NO_3)_2(MIBK)_2(orq.) \qquad \ldots (1)$$

Uranium was determined in the organic phase solution $[U]_{org}$ by volumetric method and mass balance analysis was performed to measure the

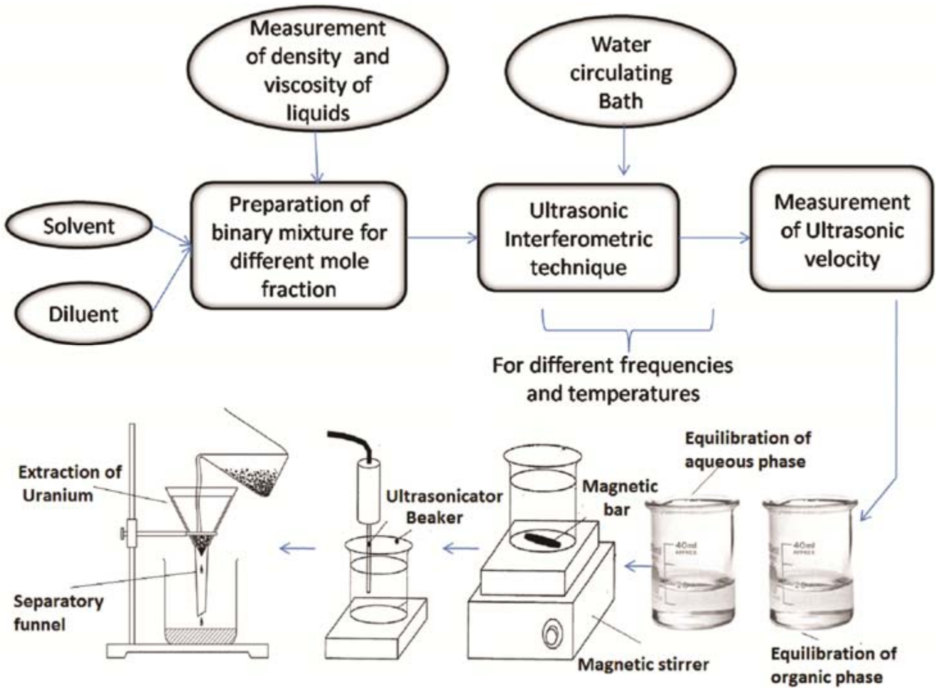

Fig. 2 — Schematic diagram for extraction procedure of uranium from uranyl nitrate.

concentration of uranium in the aqueous phase $[U]_{aq}$ and the determined distribution ratios was verified by comparing the literature of chemical equilibrium as stated by Nernst distribution law:

$$K_D = \frac{[U]_{org}}{[U]_{aqu}} \quad \ldots (2)$$

From the principle it is stated that the concentration extracted into organic and aqueous phase comes out in the numerator and denominator of the equation and the resulting value of K_D are large which indicates a high degree of extraction from aqueous phase into organic phase. On the other hand, if K_D is small, a smaller amount of chemical U is shifted from aqueous phase into organic phase[16]. If K_D is equal to unity then equivalent concentration exist in each phase.

The equilibrium constant of Eq. (3) can be expressed as:

$$K_U = \frac{[UO_2(NO_3)_2.2MIBK]}{[UO_2^{2+}][NO_3^-]^2[MIBK]^2} \quad \ldots (3)$$

4 Theory

When acoustical wave passes through the liquid sample or the treated medium, it behaves as an elastic medium. Due to this phenomenon liquid medium exerts a pressure wave caused by different variety of interactions between the molecules which depends on the distinctive properties of the medium. The acoustic wave produces due to bombardment of high intensity acoustic energy which is influenced by its velocity through the medium.

Therefore the force performing on the molecules of the system as stated by Hook's law is specified as:

$$F\alpha - x \quad \ldots (4)$$

$$F = -Kx$$

where F is the force, x is the amount of particle displacement and K is the spring constant depending on the property of the system on which the sound wave transmits and the molecular interactions occur.

If each atom or molecules of the medium assumes to be associated by a spring force then the differential equation for the acoustic wave is given as:

$$\frac{\partial^2\psi}{\partial x^2} = \frac{1}{C^2}\frac{\partial^2\psi}{\partial t^2} \quad \ldots (5)$$

where C denoted as velocity of the acoustic wave depends on the fundamental properties such as density, temperature, frequency and nature of the medium. Though the frequency is the characteristic of the medium but not the particles, so it may change when the density of the medium changes[16].

During the propagation of acoustic wave through a treated medium it may cause the molecules or atoms to interact with each other. When two molecules are not in close proximity, intermolecular forces may not significant. For the atoms or molecules which are much smaller than the half- wavelength of sound, the application of a one dimensional resonant acoustic field results in a time average force in the direction parallel to the propagation of the sound wave which can be expressed as:

$$F_{1,ac} = 4\pi a^3 k E_{ac} F \sin(2kx) \quad \ldots (6)$$

where k is wave number of the acoustical field, E_{ac} is average density of the acoustical field and x is position related to a pressure antinode,

F can be explain as the acoustical contrast factor, which evaluates the density and compressibility difference between the molecule and the continuous phase given as :

$$F = \frac{\hat{\rho} + (2/3)(\hat{\rho} - 1)}{(1 + 2\hat{\rho})} - \frac{1}{3\sigma^2 \hat{\rho}} \quad \ldots (7)$$

where $\hat{\rho}$ is ratio of density of MIBK to toluene.

When two molecules are in close proximity, intermolecular forces become significant and subsequently the interaction between the liquid molecules also becomes high. If we take an example where a secondary acoustic force can be arise when molecules will scatter an acoustic field and the interaction of a molecule with the sound field scattered by neighboring molecule can give rise a secondary acoustical force. The attractive secondary force between the two molecules with identical compressibility is given by:

$$F_{2,ac} = \frac{k^2 E_{ac}}{2\pi}\left(1 - \frac{\gamma_{m1}}{\gamma_f}\right)\left(1 - \frac{\gamma_{m2}}{\gamma_f}\right)\frac{V_{m1} V_{m2}}{r^2} \quad \ldots (8)$$

Here V_{m1} and V_{m2} are volume of first and second molecule, γ_m is compressibility of toluene and γ_f is compressibility of MIBK. Atoms or molecules also gain Vander Waals attractive forces[17]. Our principal aim is to investigate the extraction behavior of methyl iso butyl ketone and toluene as a function of temperature and frequency and by explaining different intermolecular interaction present in the solvent mixture from the theory acoustical forces explained above.

5 Results and Discussion

5.1 Application of ultrasonic technique to study the physico-chemical properties of binary liquid mixture

The experimentally measured values of density, viscosity, ultrasonic velocity and the computed value of deviated isentropic compressibility (β_S^E), deviated molar volume (V_m^E) and deviated surface tension (σ^E) have been given in the Tables 1-4. Above mentioned all the sound wave parameters have been calculated using standard relations[18,19].

Figure 3 demonstrates the deviation of ultrasonic velocity with mole fraction of MIBK for the system of MIBK with toluene which is not linear. It is observed from the variation of graph that the ultrasonic velocity decreases up to 0.4 mole fraction of MIBK and then

Table 1 — Ultrasonic velocity (U), excess isentropic compressibility (β_s^E), excess surface tension (σ^E) and molar volume (V_m) of pure methyl iso-butyl ketone (MIBK), toluene and binary mixture of MIBK and toluene at constant frequency of 2 MHz and at temperature 303.15 K.

Mole fraction of MIBK (X_1)	U (m/s)	β^E_s (N^{-1}m^{-2})	σ^E (Nm^{-1})	V_m (m^{3}mol^{-1}) ×10^{-7}
0.000	1276	0	0	0
0.106	1268	-23.0698	-2149.6354	-1.36
0.215	1262	-40.1024	-3675.6765	-2.43
0.337	1257	-53.9341	-4858.7953	-3.25
0.419	1250	-74.1783	-6660.4488	-3.57
0.548	1254	-60.3424	-5264.3152	-3.67
0.603	1259	-44.0826	-3760.4701	-3.56
0.707	1263	-29.6685	-2449.4935	-3.10
0.841	1264	-23.4770	-1920.4672	-2.03
0.909	1267	-12.2077	-976.43930	-1.26
1.000	1270	0	0	0

Table 2 — Ultrasonic velocity (U), excess isentropic compressibility (β_s^E), excess surface tension (σ^E) and molar volume (V_m) of pure methyl iso-butyl ketone (MIBK), toluene and binary mixture of MIBK and toluene at constant frequency of 2 MHz and at temperature 308.15 K.

Mole fraction of MIBK (X_1)	U (m/s)	β^E_s (N^{-1}m^{-2})	σ^E (Nm^{-1})	V_m (m^{3}mol^{-1}) ×10^{-7}
0.000	1275	0	0	0
0.106	1267	-9.4516	-724.88290	-1.38
0.215	1261	-31.1893	-2542.0776	-2.39
0.337	1256	-49.6563	-4164.9499	-3.20
0.419	1249	-72.5525	-6222.2193	-3.41
0.548	1253	-62.0778	-5395.3609	-3.45
0.603	1258	-47.4572	-4117.7031	-3.32
0.707	1262	-36.5359	-3202.0301	-2.84
0.841	1263	-34.8206	-3152.4924	-1.89
0.909	1266	-26.3916	-2428.3754	-1.15
1.000	1269	0	0	0

increases with mole fraction of MIBK, depending on the value of ultrasonic velocity of second constituent in the entire biphasic system. It gives highest deviation for low temperature 303.15 K. The outcome of adding a non-polar second element is primarily to dislocate the dipolar interactions of the first element. This phenomenon takes place due to self-association of the liquid molecules and a very weak dipole-induced dipole interaction among the constituent molecules, which is concentration dependent. This also indicates that the particular interaction exists between the constituent particles present within the solvent mixture which are used to study of structural difference[20].

The negative variation of β^E_s (Fig. 4) signifies that the extractant-diluent mixture is less compressible compared to the pure liquids which forms compound and molecules that are more compactly bound than in case of pure liquids. The β^E_s value decreases with the increase of concentration of MIBK and at low temperature it indicates maximum deviation and then

Table 3 — Ultrasonic velocity (U), excess isentropic compressibility (β_s^E), excess surface tension (σ^E) and molar volume (V_m) of pure methyl iso-butyl ketone (MIBK), toluene and binary mixture of MIBK and toluene at constant frequency of 2 MHz and at temperature 313.15 K.

Mole fraction of MIBK (X_1)	U (m/s)	β^E_s ($N^{-1}m^{-2}$)	σ^E (Nm^{-1})	V_m (m^3mol^{-1}) ×10^{-7}
0.000	1273	0	0	0
0.106	1266	-6.4461	-451.6346	-1.55
0.215	1260	-28.4889	-2251.5687	-2.69
0.337	1255	-47.1848	-3860.8784	-3.59
0.419	1248	-70.2507	-5902.6446	-3.83
0.548	1252	-59.6973	-5083.0944	-3.87
0.603	1257	-44.976	-3814.1599	-3.72
0.707	1261	-33.9425	-2903.6241	-3.18
0.841	1262	-32.1139	-2853.0105	-2.11
0.909	1265	-23.5712	-2131.5464	-1.29
1.000	1267	0	0	0

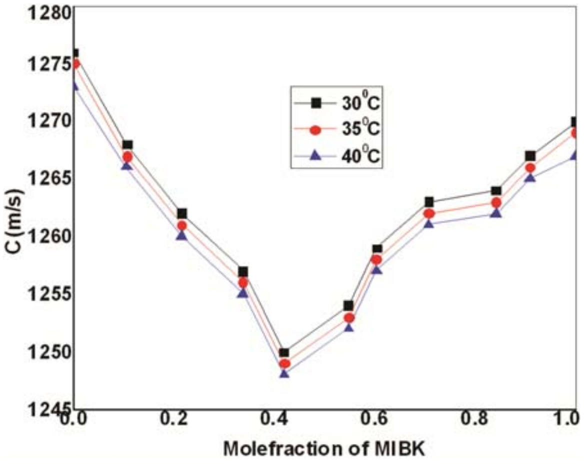

Fig. 3 — Variation of ultrasonic velocity with MIBK.

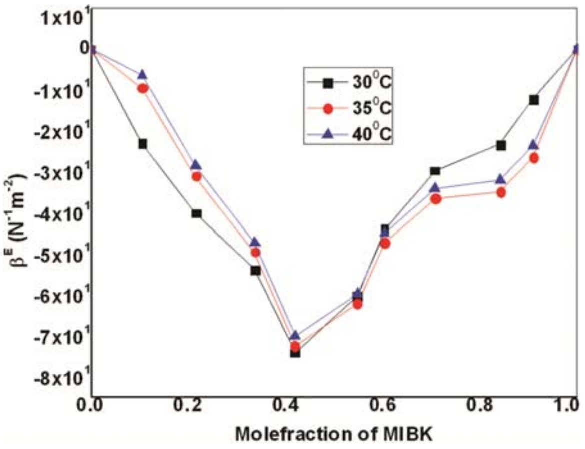

Fig. 4 — Variation of deviated isentropic compressibility with MIBK.

Table 4 — Density (ρ) and viscosity (η) of pure methyl iso-butyl ketone (MIBK) and toluene at constant frequency of 2 MHz and different temperature.

	303.15 K				308.15 K				313.15 K			
	Density (ρ) (Kgm^{-3})		Viscosity (η) (Nm^{-2})		Density (ρ) (Kgm^{-3})		Viscosity (η) (Nm^{-2})		Density (ρ) (Kgm^{-3})		Viscosity (η) (Nm^{-2})	
	Experiment	Literature	Experiment	Literature	Experiment	Literature	Experiment	Literature	Experiment	Literature	Experiment	Literature
MIBK	790.312	792.000[a]	0.5875	0.5430[d]	782.629	786.800 [b]	0.5783	--	775.068	--	0.5579	--
Toluene	854.456	--	0.5362	0.5272[e]	843.301	834.800[c]	0.5241	--	839.873	--	0.5189	--

[a,b]Ref.[28]; [c]Ref.[29]; [d]Ref.[30] and [e]Ref.[31]

increases with the increase of composition range of MIBK. This is due to the existence of comparatively stronger molecular interaction arises through hydrogen bonding[21-23] between the constituent elements. Addition of MIBK with toluene will stimulate the infringement of the connected clusters of MIBK discharging a number of dipoles which in turn can provoke a dipole moment in the molecules of toluene, following dipole-induced dipole interactions. Moreover toluene works as a structure breaker of MIBK. When the frequency of the ultrasonic interferometer increases the interaction between the components molecules gradually decreases and the breaking of associated configuration of MIBK dominates, resulting in increase[24] of $\beta^E{}_s$.

From Fig. 5 it is observed that the excess molar volumes ($V_m{}^E$) is negative and decreases with the increase of the mole fraction and temperature, which can be explained by the predominance of shrinkage in volume, caused by self association and difference in size and shape of component molecules, due to the dipole-dipole and dipole-induced dipole interactions[25,26].

Figure 6 shows that the surface tension of the liquid mixture decreases with the mole fraction of the liquid mixture. But at low temperature it shows maximum deviation. This means the interaction is not strong. It has also been observed that rise in the σ^E value occurs when mole fraction of the binary mixture increases[27].

The intense discussion on physico chemical parameters of extractants and diluent makes it suitable for their compatibility up to a particular concentration (0.4 - 0.5 mol%) as confirms from the variation of each parameter.

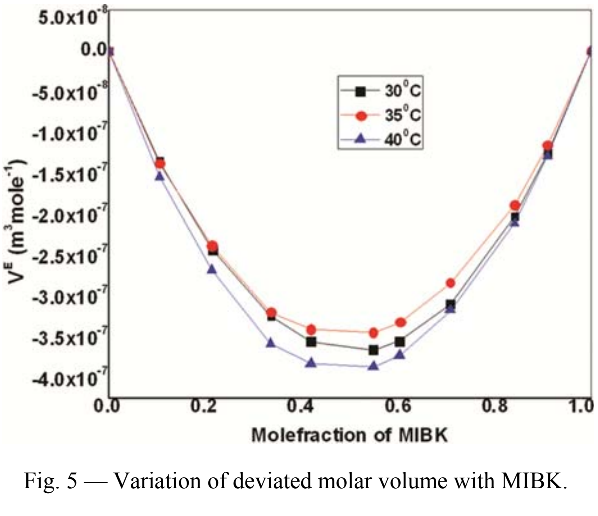

Fig. 5 — Variation of deviated molar volume with MIBK.

5.2 Effect of nitric acid Concentration

Different solutions of 0.5, 1.0, 1.5, 2.0 and 3.0 mol/dm^3 of nitric acid including 50 g/dm^3 uranium were prepared and added wit 40% of MIBK and toluene. The extraction isotherm curves for the nitric acid solution with 50 g/dm^3 uranium are shown in the figure below.

Figure 7 points out that the loading capacity of the organic phase rises significantly with increasing initial concentration of nitric acid up to 2 mol/dm^3 and then remains nearly constant with further increase in nitric acid concentration above 2 mol/dm^3.

5.3 Effect of sonication time

It was found that in ultrasonic interferometer the high frequency ultrasound 1 to 4 MHz did not have enough power to cause mixing of organic and aqueous phases. The absence of cavitations was responsible for the poor performance of the high

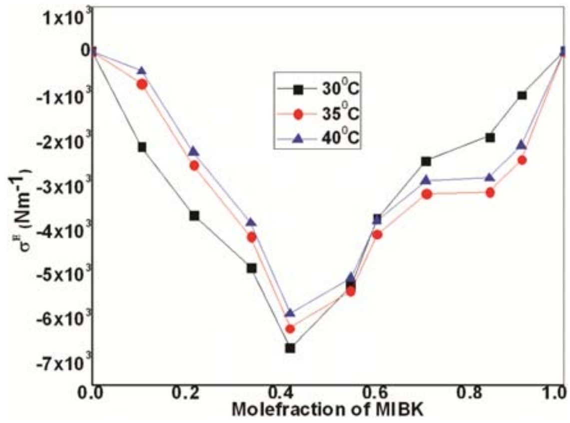

Fig. 6 — Variation of deviated surface tension for different mole fraction of MIBK.

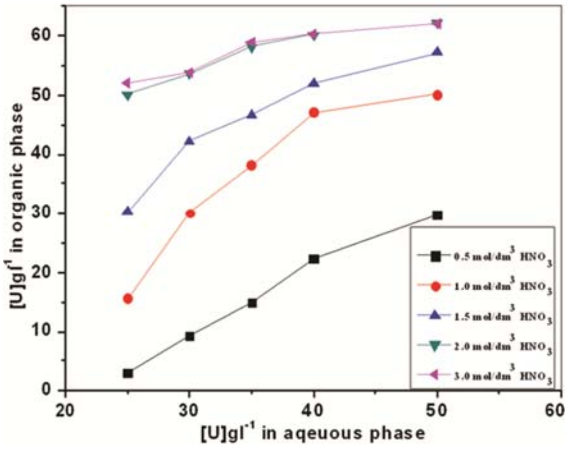

Fig. 7 — Uranium extraction isotherm for various nitric acid concentrations using 40% MIBK/toluene.

frequency ultrasound. Cavitations effects can be produced only at low frequencies[13]. For that reason, the extraction of uranium with the extractants was subjected to low frequency ultrasonicator of 125 kHz for mixing of the two phases and settlement of the phase. The higher frequency ultrasonic interferometer was used only for physico-chemical study of the liquid mixtures of MIBK and toluene.

5.4 Effect of temperature

To study the effect of temperature in the extraction process, the phenomena of uranium extraction was carried out by 40% MIBK in toluene which was pre-equilibrated with 0.5, 1.0, 1.5, 2.0 and 3 mol/dm^3 nitric acid at different temperatures. The mixture for extraction for 2 mol/dm^3 nitric acid solution was put into a magnetic stirrer, and then the mixture was stirred for 10 min at *U*org./*U*aq. = 1 at different temperatures from 25 to 50 °C. For different temperatures the variation of distribution co-efficient for nitric acid (2 mol/dm^3) medium is shown in Fig. 8. The distribution coefficient decreases with increase of temperature. Thus the extraction reaction is exothermic. So in low temperature the extraction process of uranium can be done easily.

5.5 Extraction of uranium on percentage basis

For the extraction process each concentration of MIBK /toluene was treated with uranium in addition of HNO_3 and the distribution coefficient recorded as per Nernst distribution law. The extraction of uranium increased steadily with an increase in MIBK concentration up to 0.5 mole fraction and then plateaued as shown in Fig. 9. When ultrasonic wave propagates in the treatment medium, it generates compressions and rarefactions. The compression cycle exerts a positive pressure on the medium by pushing molecules together. The rarefaction cycle exerts a negative pressure by pulling molecules from each other, and micro bubbles can be generated and grow due to such negative pressure. When these micro bubbles grow to an unstable dimension, they can collapse violently and generate shock waves, resulting in very high temperature and pressure in a few microseconds creating separation of metallic part from uranyl nitrate. Such cavitations phenomenon can increase the temperature of the emulsion system and decrease its viscosity, increase the mass transfer of liquid phase, and thus lead to destabilization of system. Ultrasonic irradiation cannot only clean the surface of solid particles but also penetrate into different regions of a multiphase system. This mechanism is called ultrasonic leaching which enables solvents to more readily enter the interior of solids pores, and increases the mass transfer of contaminants through the solids matrix.

The variation of all parameters explaining physico-chemical study exhibits its inflexion point where the interaction of ultrasonic wave with molecular environment takes place. As this particular mole fraction of blend (i.e., 0.4-0.5 mole fraction of MIBK) in almost all profiles, it suggests a good reaction of the functional group of acidic HNO_3 with extractant MIBK for extraction of metal part from uranyl nitrate in presence of low frequency ultrasonic wave. Up to 0.5 mole fraction of MIBK, the molecular environment of extractant-diluent is sufficient to react directly with the metallic part of uranyl nitrate. When the concentration of MIBK increases, reaction between the functional group of the diluent-extractant with the metallic part of uranium is weak due to high Van-der Walls force of interaction. As a result of which the removal of the metallic part decreases even if the ultrasonic wave penetrates through the

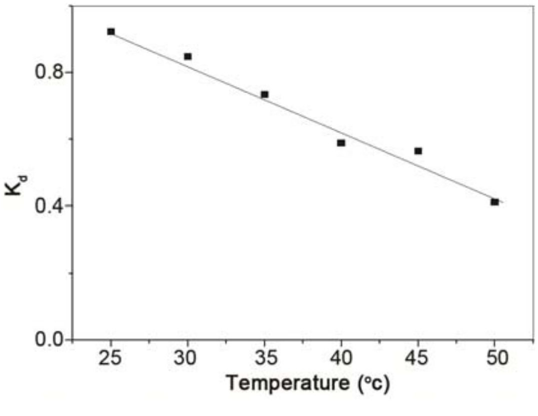

Fig. 8 — Variation of K_d against temperature, $[HNO_3]$ = 2 mol/dm^3.

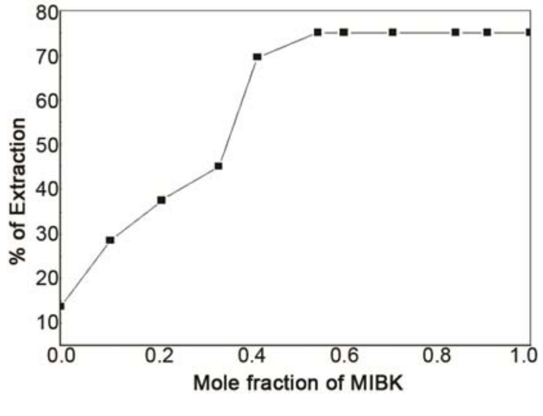

Fig. 9 — % of extraction of uranium.

molecular range. Again, the presence of more concentration of diluent-extractant reduces the interaction of ultrasonic wave with metallic part of uranyl nitrate for which the extraction of uranium is remaining constant even if the concentration of MIBK increases. Though diluents are unable to extract metal ion from the aqueous phase, but they greatly affect the extraction behavior of extractants[25].

6 Conclusions

The transmission of ultrasonic wave through the liquid medium explains the presence of the different acoustic forces which fundamentally explains the basic mechanism present in solvent mixtures to calculate the extraction efficiency for uranium extraction. Presence of various acoustic forces due to transmission of ultrasonic wave has confirmed its importance in extraction technology in terms of measurement of sound wave parameters. The variation of different acoustic parameters confirms the presence of different mutual interactions between the molecules of the solvent mixtures from which the optimum concentration and suitable parameters for extraction are evaluated. From the above analysis it is confirmed that MIBK with toluene may be used as efficient diluents/modifiers and the extraction efficiency of uranium is found to be (70-75)% in presence the optimum blend at 40% of MIBK and 60% of toluene for low frequency and low temperature.

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