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Ultrasonic Analysis of Solvent Extrancts for Nuclear Reactor Materials

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Abstract: ***The physical properties of a system basically affected by extraction power of a solvent.Tri butyl phosphate (TBP) has been extensively used as a solvent in nuclear chemistry of fuel reprocessing due to its excellent chemical resistance and physical properties which results in better separation than other solevents.Ultrasonic velocity and density measurement have been undertaken for polar extrants like tributyle phosphate (TBP) with non-polar diluents like kerosene, benzene and toluene over the entire mole fraction range of TBP at 303.15 K for frequency 2MHz. The study of different thermo acoustic parameters and their excess functions have been computed from the experimental data and used to shed light upon the molecular interactions present in the binary systems. The variation of other calculated parameters are agrees with such variation of velocity and execute their variation at same 0.3 mole fraction of TBP. A comparative study of TBP with kerosene, benzene and toluene shows kerosene is the better diluents in comparison to other diluents for processing of nuclear reactor material.***

Keywords: Solvent extraction, TBP, molecular interactions, thermoacoustic parameter

1. Introduction

The characterization and identification of different interaction in binary mixtures is an important step in liquid-liquid extraction technology. Liquid-liquid solvent extraction reagents used in several nuclear technological areas such as purification of reactor materials, radio nuclide production and the recovery of many nuclear material such as Thorium, Uranium and Plutonium etc. Solvent extraction technology is commonly used in many industrial processes and separates compound based on their relative solubilities in two different immiscible liquids. The knowledge of acoustic properties reveals the presence of different molecular interactions between the component molecules in the liquid mixture. The presence of molecular interaction in binary mixtures of nuclear extranets with non polar solvents identifies efficient modifier/diluents to be used in the solvent extraction process in the preparation of nuclear reactor materials.

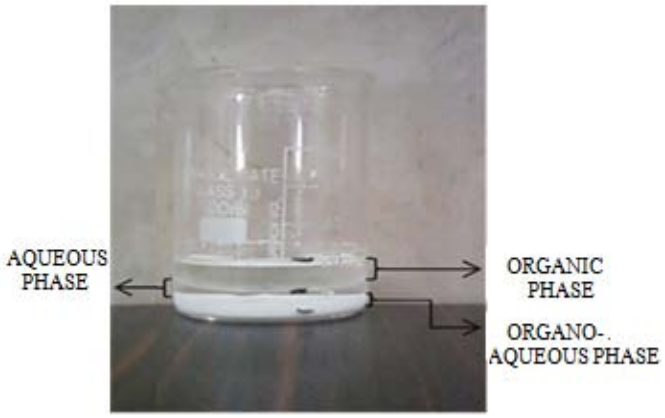

Figure 1

In extraction process it is generally observed that a third phase appears at the interface of organic and aqueous phase [1] as shown in Fig.1.which reduces the extraction efficiency of extracts in its pure form. Thus the mixing of the extracts with suitable diluents alters the physical properties of the entrants and helps to improve the extraction efficiency. As a part of our on going work, it is an attempt to study systematically the physicochemical behavior of TBP with Kerosene , benzene and toluene by evaluating different thermo-acoustic parameters like isentropic compressibility (β), intermolecular free length (L_f), acoustic impedance (Z) and the relevant excess data. An attempt has been made to study and characterize the different intermolecular interaction present in binary mixture system like TBP with kerosene.benzee and finally it is found that the extraction efficiency of REEs is also increases with mole fraction of extracts in diluents like kerosene with minimum optimum value of extranets which has been discussed in terms of intermolecular interactions.

2. Experimental Details

The ultrasonic velocity of the above liquids and their mixtures were measured using multi-frequency ultrasonic interferometer operating at different frequencies like 1MHz-5MHz (Mittal Enterprises, New Delhi,Model-MX-3).The accuracy in the measurement of ultrasonic velocity was within $\pm$ 0.01 ms^{-1}. The working principle used in the measurement of velocity of sound through medium was based on the accurate determination of the wave length of ultrasonic waves of known frequency produced by quartz crystal in the measuring cell. The temperature of the solution was controlled by circulating water at a desired temperature through the jacket of double walled cell within $\pm$0.01 K using a constant temperature bath and the temperature was monitored with a platinum resistance thermometer with an accuracy of $\pm$0.001 K.

3. Theory

The experimental measured values of ultrasonic velocity and computed values of density are used to compute acoustic parameters such as isentropic compressibility (β), intermolecular free length (Lf), acoustic impedance (Z) and surface tension and their excess values. The above acoustic parameters are determined with the help of the following relationship.

$$\beta_S = 1/\rho.C^2 \quad (1)$$

$$Lf = K.\beta^{1/2} \quad (2)$$

$$Z = \rho.C \quad (3)$$

$$\text{and } Y^E = Y_{mix} - (X_A \cdot Y_A + X_B \cdot Y_B) \quad (4)$$

where X_A, X_B, Y_A ,Y_B and Y_{mix} are mole fraction, isentropic compressibility, inter molecular free length ,acoustic impedance of TBP, Kerosene, benzene, toluene and mixture respectively. The constant k is temperature dependent which is given as [93.875+ (0.375T)] $\times 10^{-8}$ [2] and "T" being the absolute temperature.

4. Result and Discussion

From the observed values of density and ultrasonic velocity, isentropic compressibility, intermolecular free length, acoustic impedance and their deviated values were calculated. From the figure it is clear that kerosene has greater ultrasonic velocity compared to benzene and toluene as shown in Fig.2. As kerosene is a long chain hydrocarbon, so when the ultrasonic wave propagates through the mixture of kerosene and TBP there is fall up velocity takes place due to interaction of carbon molecules with the compression and rare fraction of ultrasonic waves. This is takes place up to 0.3 mole fraction of TBP as density of the medium is become more due to increase of concentration of TBP and thereafter increases gradually due to predominance of Vander walls forces between the TBP molecules.

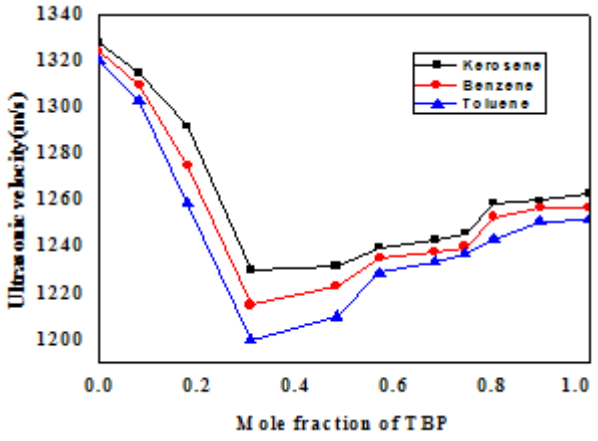

Figure 1

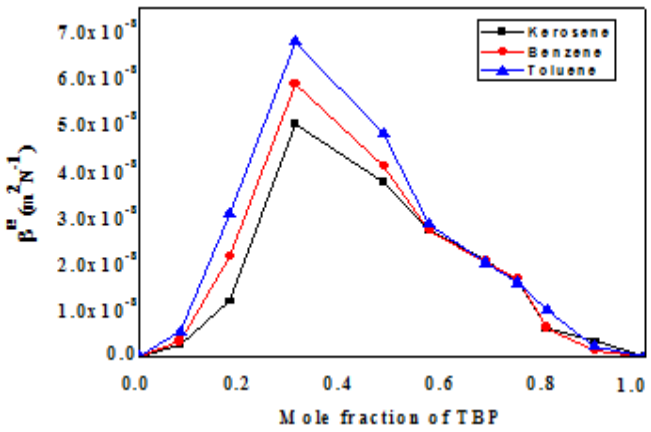

Figure 2

The variation of ultrasonic velocity in a solution depends upon the increase or decrease of intermolecular free length after mixing the components. The non-linear variation of excess properties with mole fraction of TBP is responsible for non-ideality in the binary mixture. The factors responsible for such departure from ideality may either be due to the presence of intermolecular forces between the constituents in the mixture, or to compound formation between solute and solvent, or as a result of association of either to form complex molecules [3]. These deviated parameters throw light upon the strength of interaction, and their variation with mole fraction finds application in characterizing the physico-chemical behavior[4] of liquid mixtures. The curves highlighted in figures show that the behaviour of β_s^E with composition of all the mixtures is reflected in that of L_f^E and both β_s^E and L_f^E curves are positive for all the systems under investigation. The nature of β_s^E and L_f^E play vital role in assessing the compactness due to molecular rearrangement. The extent of molecular interactions in liquid mixtures may be due to charge transfer, di- pole-induced-dipole, dipole-dipole interactions. The curve of Z^E in fig.5 shows the opposite trends as observed in figures 3 and 4 for β_s^E and L_f^E, which in fact supports the above view. The negative deviation of Z^E is attributed to specific interaction between the hetero-molecules. As these system is polar-non-polar types so the possible hetero-molecular association may be due to dipole-induced-dipole or dipole-dipole type. .

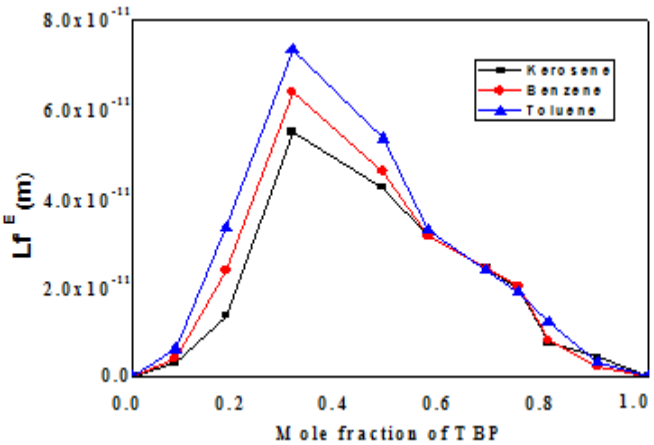

Figure 3

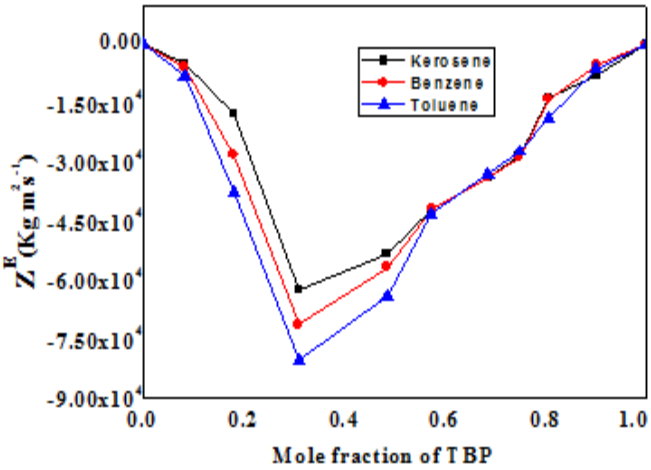

Figure 4

TBP molecules can remain either in "wood-pile" or as "head-tail configuration. Head-tail structure results in α-multimers and wood-pile structure results in β-multimers. In the binary mixtures of TBP-kerosene, TBP-benzene and TBP-toluene, TBP-TBP interaction is present throughout and no significant change in cluster formation involving dissimilar molecules is expected. The increase in the value of L_f^E on dilution may be attributed to the solvent effect. Due to less polarizability kerosene molecules away from the TBP molecules and high intermolecular free length.But due to high polarizability benzene molecules develop induced dipoles which interact with the dipolar molecules of TBP. As a result benzene molecules come closer to each other forming planar stacks sandwiching (clustering) TBP molecules. This leads to a slight increase in intermolecular free length in the mixtures but less than the kerosene. Similar observation is also observed in toluene and TBP mixture. Ultrasonic velocity is governed by the combined effect of intermolecular free length and density.

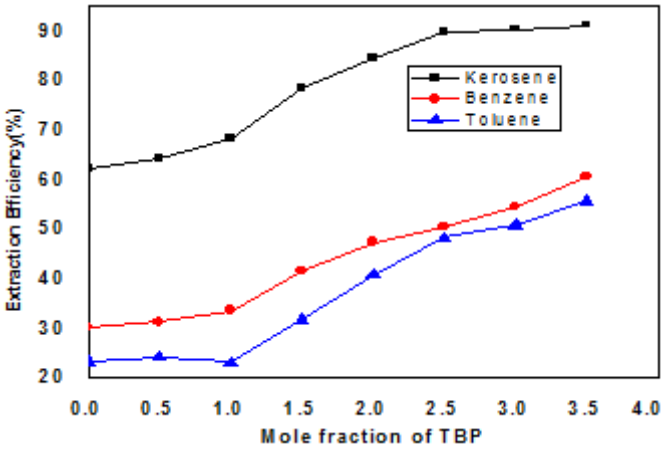

Figure 6: Extraction of nuclear material vs Concentration of TBP with different diluents

On mixing of components, specially for non-polar types of components there is the probabilities of various type of forces like H-bonding, dipole-dipole, dipole-induced dipole, dispersion forces ,charge transfer etc. operating in them. The pure kerosene has low density and low viscosity. This property of kerosene makes it becomes non-polar for which it is immiscible with other liquids. It can acts as better diluents. As seen from the discussion of the different thermo acoustic parameter the behavior of kerosene with TBP though similar to that of benzene and toluene but the dilution capacity of kerosene is better than benzene and toluene [5]. In pure kerosene the ultrasonic velocity is high, so the extraction efficiency of kerosene is higher than the other diluents like benzene and toluene.

5. Conclusions

Thus it can be concluded that the interaction of TBP kerosene is stronger than the other diluents like benzene and toluene in low frequency as is observed in different acoustic parameters. When the concentration of kerosene decreases and TBP concentration increases there is more than one type interaction like dipole-dipole ,dipole-induced dipole, dispersion force may appears in the mixture which basically responsible explain the suitable diluents behavior of kerosene as compared to benzene and toluene.

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