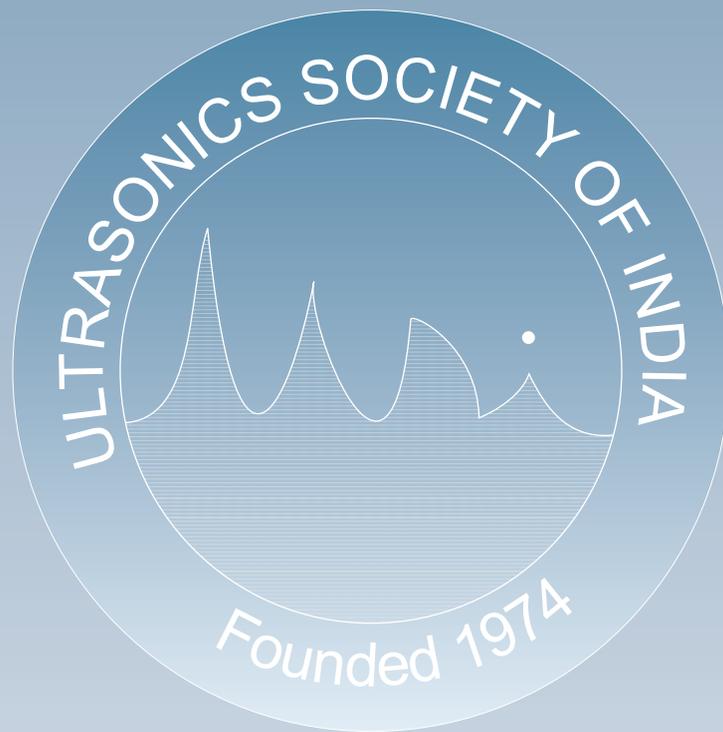


Journal of Pure and Applied
Ultrasonics



Website : www.ultrasonicsindia.org

A Publication of Ultrasonics Society of India



Ultrasonics Society of India

Ultrasonics Society of India established in 1974, is engaged in the promotion of research and diffusion of knowledge concerning the field of ultrasonics and allied areas.

Patron : Dr. V.N. Bindal
vnbindal@yahoo.co.in

Executive Council :

President Prof. Vikram Kumar
vkmr47@gmail.com

Vice-President Prof. R.R. Yadav
rryadav1@rediffmail.com

Dr. V.R. Singh
vrsingh@yahoo.com

General Secretary Dr. Yudhisther Kumar Yadav
ykyadav6659@gmail.com

Joint Secretary Mr. Gurmukh Singh
guru6850@gmail.com

Publication Secretary Dr. Devraj Singh
devraj2001@gmail.com

Treasurer Dr. (Mrs.) Kirti Soni
2006.kirti@gmail.com

Members Dr. N. R. Pawar
pawarsir1@gmail.com
Dr. K. Sakthipandi
sakthipandi@gmail.com
Dr. S. K. Jain
skjainnpl@yahoo.co.in
Dr. Ganeswar Nath
ganeswar.nath@gmail.com
Dr. J. Poongodi
poongodinagaraj@gmail.com
Dr. Janardan Singh
dr_janardansingh@yahoo.com
Dr. Punit Kumar Dhawan
pntdhawan@gmail.com
Dr. Giridhar Mishra
giridharmishra@rediffmail.com
Dr. Mukesh Chandra
mchandra1948@yahoo.in
Dr. Alok Kumar Gupta
alokphy@gmail.com

Immediate past president Dr. Krishan Lal
krish41ster@gmail.com

Past patrons Prof. A. R. Verma
Prof. E. S. R. Gopal

Membership of the Society is open to individuals without distinction of sex, race or nationality and to bodies who subscribe to the aims and objectives of the Society.

The membership fee is as follows :

Class of Membership	Subscription (one time)
Honorary Fellow	Nil
Life Fellow / Member	Rs 3000/-
Associate Member	Rs 1000/- (for 5 yrs.)
Corporate Member	Rs 20000/-
Life Fellow / Member (Foreign)	US \$ 150
Corporate Member (Foreign)	US \$ 1000

Membership forms and the relevant information can be downloaded from the website or obtained from :

The General Secretary, Ultrasonics Society of India
CSIR-NPL, Dr. K.S. Krishnan Marg, New Delhi-110012
E-mail : ykyadav@nplindia.org

A Quarterly Publication of Ultrasonics Society of India

Journal of Pure and Applied *Ultrasonics*

No. 4	Volume 41	October-December 2019
-------	-----------	-----------------------

Chief Editor : Dr. S.K. Jain
Former Chief Scientist
CSIR-National Physical Laboratory, New Delhi
skjainnpl@yahoo.co.in

Editorial Board :

Prof. S.S. Mathur Formerly Prof., Indian Institute of Technology, New Delhi
sartajmathur@yahoo.co.in

Dr. P. Palanichamy Formerly Scientist, IGCAR, Kalapakkam, Tamil Nadu
ppc9854@gmail.com

Prof. R.R. Yadav Vice-Chancellor, Veer Bahadur Singh Purvanchal University, Jaunpur, U.P.

Publication Committee :

Dr. Devraj Singh AIAS, Amity University, Noida

Dr. Sanjay Yadav CSIR-National Physical Laboratory, New Delhi

Dr. K. Sakthipandi Sethu Institute of Technology, Pulloor Kariapatti, Tamil Nadu

Dr. Giridhar Mishra VBS Purvanchal University, Jaunpur, U.P.

Dr. Meher Wan CSIR-NISCAIR, New Delhi

Dr. Kirti Soni CSIR-National Physical Laboratory, New Delhi

Dr. Y. K. Yadav CSIR-National Physical Laboratory, New Delhi

SUBSCRIPTION (postage paid)

Single	Rs. 750/-	US\$ 60/-
Annual	Rs. 3000/-	US\$ 250/-
USI members	Free	

ADVERTISEMENT

The Journal offers opportunity of wide and effective publicity for the manufacturer, suppliers of ultrasonic equipment, devices and materials and also for scientific instruments and components. Tariff is as follows :

Back Cover	Rs. 5000/-	US \$ 200/-
Inside Cover	Rs. 3000/-	US \$ 150/-
Full page	Rs. 2000/-	US \$ 100/-
Half page	Rs. 1500/-	US \$ 60/-

Discount of 20% is admissible for 4 successive insertions.

The submission of papers and all other correspondence regarding the Journal may please be addressed to :

Publication Secretary
Journal of Pure and Applied Ultrasonics
C/o Ultrasonics Society of India
CSIR-National Physical Laboratory
Dr. K.S. Krishnan Road, New Delhi-110012
publicationsecretary.usi@gmail.com
www.ultrasonicsindia.org

Journal of Pure and Applied Ultrasonics

VOLUME 41

NUMBER 4

OCTOBER-DECEMBER 2019

CONTENTS

Thermo-acoustic studies of benzotriazole substituted 1, 2, 4-triazole derivatives Dinesh R. Godhani, Vishal B. Mulani and Jignasu P. Mehta	79
Acoustical parameters of binary mixtures of 1,4-dichlorobutane with some hydrocarbons at 308.15K, 313.15K and 318.15K Kanchan Yadav and S.S. Yadava	86
Estimation of Debye temperature for binary liquid mixtures using ultrasonic techniques A. Mathana Gopal, A. Moses Ezhil Raj and J. Poongodi	90
Study of highly viscous polyacryle measurements of viscosity and ultrasonic velocity S.V. Khangar, O.P. Chimankar, D.V. Nandanwar, J.N. Ramteke and Swapnil A Kale	94
Author Index	97

(Authors have stated that the papers have not been published elsewhere)

EDITORIAL

Contemporary research in Physical Ultrasonics for relevance in industrial and basic applications

Dear Colleagues,

The Journal of Pure and Applied Ultrasonics has been publishing research papers, review articles, thesis summary, etc. on physical ultrasonics besides the other subjects such as non-destructive NDE, underwater acoustics, instrumentation, transducers, *etc.*. The studies on physical ultrasonics include thermo-acoustic parameters of (i) organic liquid mixtures and inorganic electrolytes, (ii) mechanical and thermal properties of single crystals of various elements and their inter-metallic compositions based on various series of periodic table. It is amazing to see how the subject has developed over the years and authors are looking various possibilities where this research can lead to potential applications in determination of properties such as effective blending of polymers, polymer chain lengths, ionic dissociation, thermal and refractory properties of industry relevant solids.

Subsequent to development of NRDC award winning innovation in ultrasonic interferometer at National Physical Laboratory, New Delhi during early 70's, it caught up the attention of physicists and chemists as an affordable and accurate device for ultrasonic velocity measurements in liquids at various temperatures. It led to a boom in research of thermo-acoustic properties of organic, organic-inorganic liquid mixtures, electrolytes, *etc.* Whereas most of such studies have relied in precision measurement of ultrasonic velocity, density and viscosity at a fixed frequency at temperatures in the vicinity of room temperature, it may be desired the conclusions on molecular associations, nature of molecular forces, *etc.* based on these which are mostly qualitative in nature are reaffirmed. Ultrasonic waves couple strongly to molecular volume and viscosity of liquids and as such ultrasonic absorption is strongly influenced by the relaxation processes and shows strong frequency dependence. The measurement of ultrasonic absorption over a wide range of frequency can yield quantitative conclusions about thermodynamic properties and hence molecular processes in liquids and in liquid systems. (Udo Kaatz, Broad-band acoustical spectroscopy of liquids: a review, *J. Pure Appl. Ultrason.*, **31** (2009) pp. 41-60). The determination of molecular relaxation processes, complex formations, concentration fluctuations, nature of molecular interactions, *etc.* can be possible through wide band measurement of ultrasonic absorption at various concentrations. I would like to emphasize that broad-band spectroscopy ("Ultrasonic measurement techniques for liquids, I Broad-band Spectroscopy, **30** (2008) pp. 1-12) of ultrasonic absorption has become a powerful tool in study of molecular processes in applied chemistry, biochemistry and materials science. Authors of research papers in physical ultrasonics can expand the scope of their research multi-fold by adopting techniques for broad band ultrasonic spectroscopy to reveal the molecular processes, kinetics, interactions, *etc.* in the large number of novel liquid systems waiting to be explored.

— **S. K. Jain**
Chief Editor

Thermo-acoustic studies of benzotriazole substituted 1, 2, 4-triazole derivatives

Dinesh R. Godhani*, Vishal B. Mulani and Jignasu P. Mehta

DST-FIST Sponsored, Department of Chemistry, Mahatma Gandhi Campus,
Maharaja Krishna Kumar Singh Ji, Bhavnagar University, Bhavnagar-364 022, India

*E-mail: drgodhani@yahoo.com

The thermo-acoustic properties of binary solutions of *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-bromophenyl)methanimine, (BT₄) and *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-methoxyphenyl)methanimine, (BT₁₁) in dimethyl sulfoxide (DMSO) and *N,N*-dimethyl formamide (DMF) were estimated. The thermodynamic and various acoustical properties were determined at three different temperatures *viz.* (298.15, 308.15 and 318.15 K) at atmospheric pressure. Likewise, the similarity between experimental results of the binary solutions was studied to analyze how the change in the structural modification and solvent change the values of Gibbs energy of activation (ΔG^*), enthalpy of activation (ΔH^*) and entropy of activation (ΔS^*).

Keywords: Thermodynamic parameters; density; molecular interaction; ultrasonic sound velocity; acoustical parameter.

Introduction

The thermo-acoustic study is essential for active pharmaceutical ingredient¹. Which is used in the formulation so the drug has better stability for long duration with the same mechanism of action². This study is also useful for the mass production of the drug by applying better engineering³. The scientists developed many types of studies for different development phases⁴. In primary development phase the data of melting point, solubility, viscosity, density, conductance, ultrasonic sound velocity, refractive index and the thermal study provides valuable guidance about molecular interactions taken places in the solution⁵.

The joint study of density, ultrasonic sound velocity and viscosity provides very important data about molecular interactions taken places in the solution which suggest complex formation is taken places in the solution or not⁶. Furthermore, thermodynamic parameters such as ΔG^* , ΔH^* , ΔS^* were determined from viscous relaxation time data from various concentration and temperature. Which provides data of reaction condition and chemical kinetics⁷.

In the present study, we have selected 1,2,4-Triazoles, which are act as a destroyer of the fungal cell wall and reproduction system⁸. Additionally, they show good antimicrobial activities. In the extension of our previous work⁹, the present analysis defines the influence of temperature and solvents on the molecular interactions on the same molecule with a different functional group. Like, *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-bromophenyl)methanimine, (BT₄) and *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-methoxyphenyl)methanimine, (BT₁₁) and thereby effect on thermo-acoustical considerations at atmospheric pressure. The results are interpreted in terms of molecular interaction evolving in the solution.

Materials and Methods

The *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-bromophenyl)methanimine, (BT₄) and *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-methoxyphenyl)methanimine, (BT₁₁) used in this analysis was manufactured in our laboratory⁹. The

structure of the present synthesized compounds BT₄ and BT₁₁ is shown in Fig. 1.

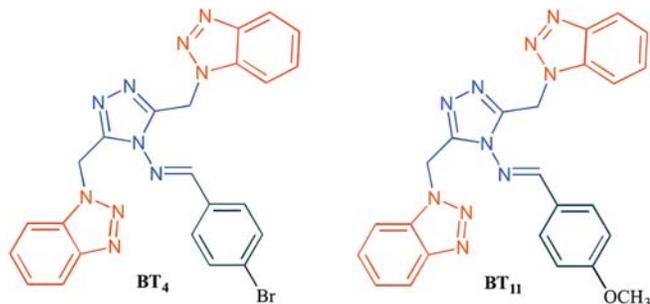


Fig. 1. Structure of *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-bromophenyl)methanimine, (BT₄) and *N*-(3,5-bis((1*H*-benzo[*d*][1,2,3]triazol-1-yl)methyl)-4*H*-1,2,4-triazol-4-yl)-1-(4-methoxyphenyl)methanimine, (BT₁₁).

The solvents dimethyl sulfoxide (DMSO) and *N,N*-dimethyl formamide (DMF) used in the current study were of AR grade supplied by Spectrochem Pvt. Ltd. and was purified according to literature method^{5,10}. The estimated purity of solvents was more than 99.5% and was confirmed by GC-MS (Shimadzu model no. QP2010) technique. The moisture content was measured by Karl Fischer titration as shown in Table 1. The Benzotriazole substituted 1,2,4-triazole derivatives were synthesized according to standard protocols.

Ultrasonic sound velocity measurements in pure solvents (DMSO and DMF) and solutions of both the compound (0.001-0.010 mol kg⁻¹) in DMSO and DMF were made at three different temperatures *viz.* (298.15, 308.15 and 318.15 K). The temperature was controlled by the digital temperature controller of the Instrucare solution viscosity bath with an accuracy of 0.5°C. For density measurement, Rudolf research analytical's automatic density meter (DDM2909) was used. Fungilabviscolead advance viscometer was used for the viscosity measurement. Ultrasonic sound velocity (*U*), density (ρ) and viscosity (η) measurements were accurate to $\pm 0.03\%$, $\pm 0.0002 \text{ g cm}^{-3}$ and $\pm 0.40\%$

respectively. The uncertainty of temperature is $\pm 0.1\text{K}$ and that of concentration measured is $\pm 0.001 \text{ mol dm}^{-3}$. All the samples were prepared freshly using Sonixvibracell (VCX500) and retained at the required temperature for 24h to settle their solubility at the temperature. Samples were preserved in bottles under vacuum until further use⁶.

Results and Discussion

The standard data of density (ρ), viscosity (η) and ultrasonic sound velocity (*U*) of dimethyl sulfoxide (DMSO) and *N,N*-dimethylformamide (DMF) used in the current analysis are given in Table 2 and compared with the accessible literature data, which suggest acceptable agreement between theoretical and practical data. The outcomes were also suggesting that our experiments were perfectly calibrated and standardized.

The ρ , η and *U* of pure solvents and solutions of BT₄ and BT₁₁ in DMSO and DMF were measured at $T = (298.15, 308.15 \text{ and } 318.15 \text{ K})$ and are given Figs. 2-4. The density and viscosity of medium, pressure, temperature etc. effect on the ultrasonic sound velocity. With a view to understand the influence of concentration, temperature, nature of solvents and the structures of BT₄ and BT₁₁ on structure forming or structure-breaking tendency, several parameters¹¹ like isentropic compressibility (k_S), Van der Waals constant (*b*), internal pressure (π), Solvation number (S_n), free volume (V_f), intermolecular free path length (L_f), viscous relaxation time (τ), acoustical impedance (*Z*), Gibbs energy of activation (ΔG^*), enthalpy of activation (ΔH^*) and entropy of activation (ΔS^*)¹² were determined by using the experimental data of ρ , η and *U* of BT₄ and BT₁₁ in DMSO and DMF solutions at three temperatures according to standard equations. The concentration and temperature dependence of acoustical parameters provide a wealth of information concerning the strength of molecular interaction appearing in the solutions of BT₄ and BT₁₁.

Table 1 – Chemicals used during the experiment.

Chemicals	Source	CAS number	% moisture	Molar mass
DMSO	Spectrochem Pvt. Ltd.	67-68-5	0.2	78.13
DMF	Spectrochem Pvt. Ltd.	68-12-2	0.1	73.09
BT ₄	Synthesis	-	0.2	512.08
BT ₁₁	Synthesis	-	0.2	464.18

Table 2 – Comparison of measured density, ρ , viscosity, η , and ultrasonic sound velocity, U data for pure DMSO and DMF with literature values at $T = (298.15, 308.15 \text{ and } 318.15 \text{ K})$.

Organic liquids	This work			Literature		
	$T = 298.15 \text{ K}$	$T = 308.15 \text{ K}$	$T = 318.15 \text{ K}$	$T = 298.15 \text{ K}$	$T = 308.15 \text{ K}$	$T = 318.15 \text{ K}$
	$\rho / (\text{kg} \cdot \text{m}^{-3})$			$\rho / (\text{kg} \cdot \text{m}^{-3})$		
DMSO	1095.34	1085.16	1075.19	1095.29 ¹³	1085.25 ¹³	1075.21 ¹³
DMF	943.95	934.39	924.78	943.94 ¹⁶	934.64 ¹⁶	925.80 ¹⁷
	$\eta / (\text{mPa} \cdot \text{s})$			$\eta / (\text{mPa} \cdot \text{s})$		
DMSO	1.8613	1.4748	1.2581	1.8480 ¹³	1.4980 ¹³	1.310 ¹⁴
DMF	0.8072	0.7064	0.6174	0.8040 ¹⁸	0.7103 ¹⁷	0.6348 ¹⁷
	$U / (\text{m} \cdot \text{s}^{-1})$			$U / (\text{m} \cdot \text{s}^{-1})$		
DMSO	1484.4	1450.8	1416.8	1485.1 ¹⁵	1451.3 ¹⁵	1417.7 ¹⁵
DMF	1457.11	1418.04	1379.81	1457.49 ¹⁶	1418.95 ¹⁶	1379.60 ¹⁹

It is clear from Tables 3 that ρ , η and U increased with concentration (C) and decreased with temperature (T). The concentration and temperature dependence of these data were tested by least square analysis. The experimental correlation between ρ and C are $R^2 = 0.832-0.991$, η and C are $R^2 = 0.990-0.999$ and U and C are $R^2 = 0.990-0.999$ which suggest a fairly good to an outstanding linear relationship between studied

parameters as shown in Figs. 2-4. The increased ρ , η and U with C indicate that the increasing of cohesive forces was due to powerful molecular interactions, while the decreasing of these parameters with T indicates that cohesive forces were decreased. The increasing temperature has two opposite effects namely structure formation (intermolecular association) and structure destruction. When the thermal energy is higher than the

Table 3 – The least-square equations and regression coefficients for BT₄ solutions in DMSO at $T = (298.15, 308.15 \text{ and } 318.15 \text{ K})$.

Parameter	Least square equations (regression coefficients, R^2)		
	$T = 298.15 \text{ K}$	$T = 308.15 \text{ K}$	$T = 318.15 \text{ K}$
$\rho / (\text{kg} \cdot \text{m}^{-3})$	$y = 343.12x + 1096.6$ $R^2 = 0.8863$	$y = 301.1x + 1086.7$ $R^2 = 0.9444$	$y = 379.42x + 1077.7$ $R^2 = 0.8320$
$\eta / (\text{mPa} \cdot \text{s})$	$y = 29.281x + 1.8755$ $R^2 = 0.9967$	$y = 21.429x + 1.5091$ $R^2 = 0.9994$	$y = 15.384x + 1.3078$ $R^2 = 0.9929$
$U / (\text{m} \cdot \text{s}^{-1})$	$y = 680.41x + 1485.8$ $R^2 = 0.9991$	$y = 604.3x + 1452.4$ $R^2 = 0.9966$	$y = 655.56x + 1418.8$ $R^2 = 0.9939$
$Z \cdot 10^6 /$ $(\text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1})$	$y = 1.2583x + 1.6293$ $R^2 = 0.9770$	$y = 1.0958x + 1.5782$ $R^2 = 0.9843$	$y = 1.2473x + 1.529$ $R^2 = 0.9471$
$k_s \cdot 10^{-10} / (\text{Pa}^{-1})$	$y = -5.0322x + 4.1308$ $R^2 = 0.9892$	$y = -4.8019x + 4.3626$ $R^2 = 0.9899$	$y = -5.8323x + 4.6094$ $R^2 = 0.9683$
$L_f \cdot 10^{-11} / (\text{m})$	$y = -2.6003x + 4.2554$ $R^2 = 0.9893$	$y = -2.4137x + 4.3732$ $R^2 = 0.9901$	$y = -2.853x + 4.4952$ $R^2 = 0.9686$
$b \cdot 10^{-5} / (\text{m}^3)$	$y = -1.7988x + 7.0246$ $R^2 = 0.8394$	$y = -1.5621x + 7.0856$ $R^2 = 0.9155$	$y = -2.1026x + 7.1416$ $R^2 = 0.7769$
$\pi \cdot 10^8 / (\text{Pa})$	$y = 56.255x + 7.5859$ $R^2 = 0.9942$	$y = 47.724x + 7.0701$ $R^2 = 0.9996$	$y = 38.611x + 6.8361$ $R^2 = 0.9889$
$V_f \cdot 10^{-7} / (\text{m}^3)$	$y = -10.286x + 0.5465$ $R^2 = 0.9894$	$y = -12.658x + 0.7321$ $R^2 = 0.9989$	$y = -12.867x + 0.8782$ $R^2 = 0.9860$
$\tau \cdot 10^{-13} / (\text{s})$	$y = 146.62x + 10.333$ $R^2 = 0.9965$	$y = 113.53x + 8.7805$ $R^2 = 0.9991$	$y = 83.154x + 8.0397$ $R^2 = 0.9939$
S_n	$y = 2E+09x^4 - 5E+07x^3 +$ $417984x^2 - 732.02x + 1.2142$ $R^2 = 0.9996$	$y = -1E+09x^4 + 3E+07x^3 -$ $117812x^2 + 785.09x - 0.0182$ $R^2 = 0.9952$	$y = 1E+13x^5 - 3E+11x^4 + 2E+09x^3 -$ $1E+07x^2 + 17252x - 9.0944$ $R^2 = 1$

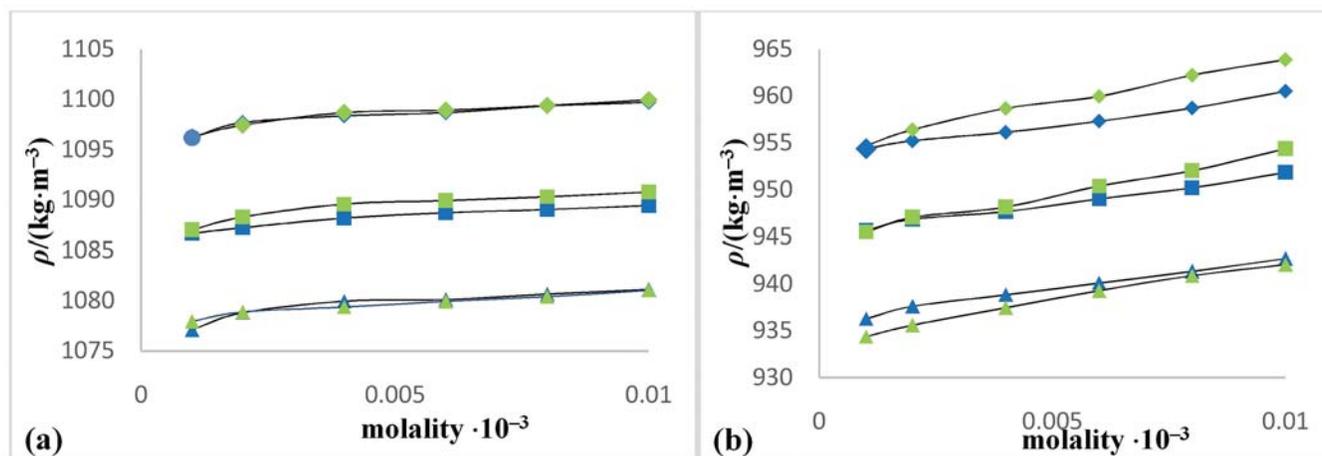


Fig. 2. The plots of Density (ρ) against molality (m) for BT₄ and BT₁₁ in (a) DMSO and (b) DMF at 298.15K [BT₄-(\blacklozenge), BT₁₁-(\blacklozenge)], 308.15K [BT₄-(\blacksquare), BT₁₁-(\blacksquare)] and 318.15K [BT₄-(\blacktriangle), BT₁₁-(\blacktriangle)].

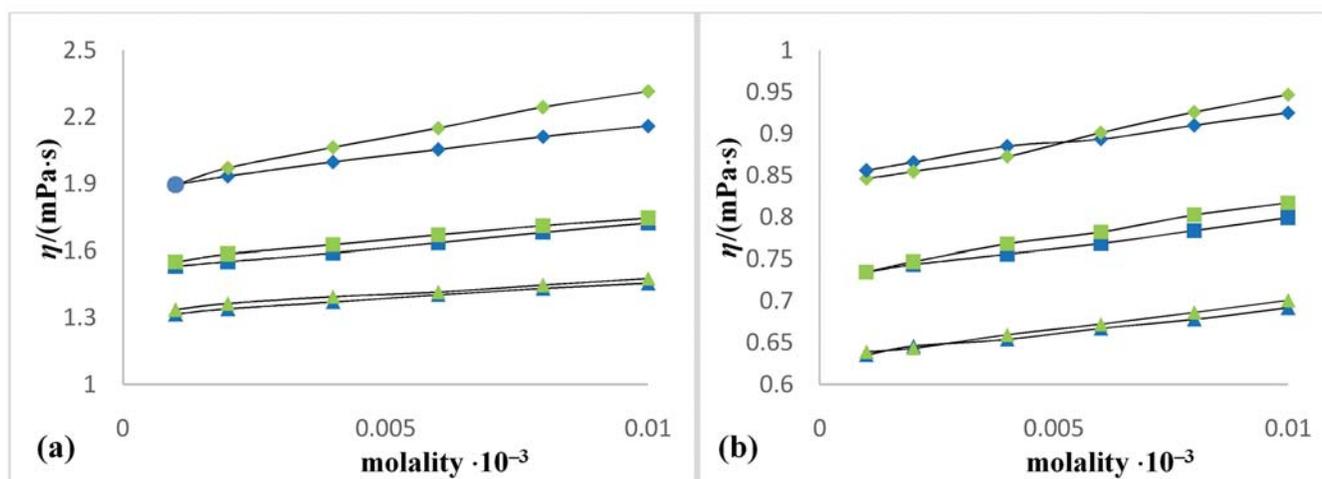


Fig. 3. The plots of viscosity (η) against molality (m) for BT₄ and BT₁₁ in (a) DMSO and (b) DMF at 298.15K [BT₄-(\blacklozenge), BT₁₁-(\blacklozenge)], 308.15K [BT₄-(\blacksquare), BT₁₁-(\blacksquare)] and 318.15K [BT₄-(\blacktriangle), BT₁₁-(\blacktriangle)].

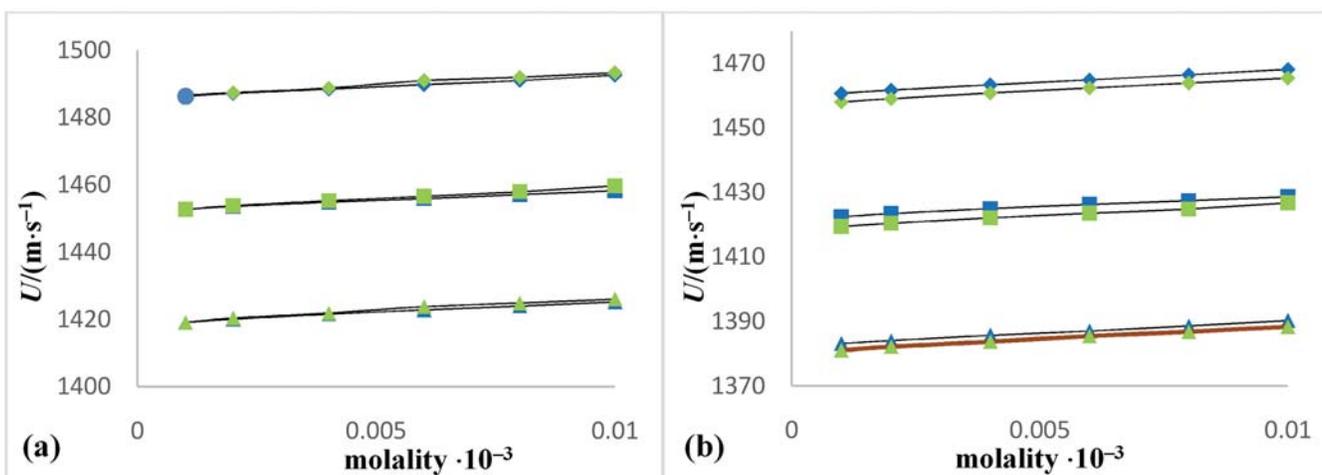


Fig. 4. The plots of ultrasonic sound velocity (U) against molality (m) for BT₄ and BT₁₁ in (a) DMSO and (b) DMF at 298.15K [BT₄-(\blacklozenge), BT₁₁-(\blacklozenge)], 308.15K [BT₄-(\blacksquare), BT₁₁-(\blacksquare)] and 318.15K [BT₄-(\blacktriangle), BT₁₁-(\blacktriangle)].

interaction energy, it causes the destruction of a previously formed structure. Thus, increasing of T favors the increase of kinetic energy and volume expansion and hence, a decrease in ρ and η was observed¹⁰.

Ultrasonic sound velocity (U) depends on intermolecular free path length (L_f) inversely. It is seen from Tables 4 that both ultrasonic sound velocity and acoustical impedance (Z) increased with C and decreased with T of solutes in DMSO and DMF systems. Due to solvent-solute interactions, the structural arrangement was significantly changed for both derivatives. The intermolecular free path length (L_f) was seen to decrease with C and increased with T for both derivatives suggesting the existence of solvent-solute interactions as summarized in Table 5. The distribution of U in the system gives information about the characteristic time of the relaxation process that causes the distribution. Detected viscous relaxation time (τ) was due to structural relaxation procedures. The viscous relaxation time (τ) of both the mixture was found decreased with T and increased with C in both of the binary systems.

According to the equation, the value of ΔG^* was calculated using viscous relaxation time data at the different concentrations and temperatures. The variation of ΔG^* values with C and T for BT_4 and BT_{11} are summarized in Table 6. The least squares intrinsic values of ΔG^* for BT_4 and BT_{11} in DMSO and DMF systems were linearly increased with C and linearly decreased

with T . The values of ΔG^* were positive for BT_4 and BT_{11} in both solvent systems, signifying the occurrence of interaction between the molecules of the mixtures. The enthalpy of activation (ΔH^*) and entropy of activation (ΔS^*) measured in DMSO and DMF solutions are summarized in Table 4. The ΔH^* values were positive for BT_4 and BT_{11} in both the binary systems at all experimental temperatures. The positive values of ΔH^* indicates that the reaction was endothermic in nature¹¹. The values of ΔS^* were positive for BT_4 and BT_{11} in DMSO and DMF system, at all experimental temperatures. The positive magnitude of ΔS^* suggests that the process of complex formation was non-spontaneous. The values of ΔS^* and ΔH^* were positive for BT_4 and BT_{11} in DMSO and DMF solutions at all temperatures suggest that the transition state was related to the bond formation and the reaction was non-spontaneous. The analytical evidence suggests that the structure of compound BT_4 and BT_{11} was found more compact in its lattice.

The decreased of isentropic compressibility (kS) with C and increased with T in both the binary mixture might be due to the accumulation of solvent molecules around solute molecules supporting strong solvent-solute interactions and this occurrence can be attributed to the solvated molecules that were entirely compressed by the electrical forces. The linear fluctuations in Van der Waals constant (b) shown in Table 5, propose that complex or aggregate formation has taken place for

Table 4 – Variation of Gibbs free energy of activation, ΔG^* , enthalpy of activation, ΔH^* and entropy of activation, ΔS^* with concentration for BT_4 in different solvent systems at $T = (298.15, 308.15 \text{ and } 318.15 \text{ K})$.

System	$\Delta G^*/ (\text{J} \cdot \text{mol}^{-1})$			$\Delta H^* /$ ($\text{kJ} \cdot \text{mol}^{-1}$)	$\Delta S^*/$ ($\text{J} \cdot \text{K}^{-1} \text{mol}^{-1}$)
	T = 298.15 K	T = 308.15 K	T = 318.15 K		
DMSO+ BT_4	4652.88	4513.63	4374.38	8.8045	13.9247
	4693.86	4553.28	4412.71	8.8852	14.0577
	4763.25	4611.61	4459.98	9.2842	15.1635
	4829.56	4674.52	4519.47	9.4522	15.5044
	4896.81	4733.29	4569.76	9.7723	16.3524
	4951.76	4783.33	4614.90	9.9735	16.8429
DMF+ BT_4	3131.42	3065.03	2998.64	5.1109	6.6393
	3153.54	3083.46	3013.39	5.2428	7.0075
	3180.05	3105.84	3031.62	5.3928	7.4215
	3213.70	3136.54	3059.37	5.5144	7.7165
	3253.88	3172.14	3090.39	5.6911	8.1744
	3285.08	3196.98	3108.89	5.9116	8.8093

Table 5 – Variation of Gibbs free energy of activation, ΔG^* , enthalpy of activation, ΔH^* and entropy of activation, ΔS^* with concentration for BT₁₁ in different solvent systems at $T = (298.15, 308.15 \text{ and } 318.15 \text{ K})$.

System	$\Delta G^*/ (\text{J} \cdot \text{mol}^{-1})$			$\Delta H^* /$ (kJ . mol ⁻¹)	$\Delta S^*/$ (J . K ⁻¹ mol ⁻¹)
	T = 298.15 K	T = 308.15 K	T = 318.15 K		
DMSO+ BT ₁₁	4653.80	4536.51	4419.21	8.1510	11.7298
	4741.37	4600.13	4458.89	8.9525	14.1242
	4841.03	4673.43	4505.83	9.8380	16.7598
	4933.45	4733.67	4533.90	10.8897	19.9773
	5030.36	4805.56	4580.77	11.7327	22.4798
	5096.70	4859.35	4622.00	12.1734	23.7352
DMF+ BT ₁₁	3112.83	3060.55	3008.27	4.6716	5.2283
	3134.85	3080.40	3025.96	4.7581	5.4444
	3181.15	3123.60	3066.06	4.8968	5.7544
	3246.39	3182.22	3118.05	5.1597	6.4172
	3304.71	3235.39	3166.06	5.3717	6.9326
	3345.54	3275.72	3205.89	5.4274	6.9825

BT₄ and BT₁₁ in DMSO and DMF systems. The internal pressure (π) is subsequent forces of attraction and repulsion between the entities present in the solution. The results of isentropic compressibility and intermolecular free path length for BT₄ and BT₁₁ was decreased with C and increased with T . This was established from the results of internal pressure (π), which was found enlarged. The free volume (V_f) of a solute molecule at a specific temperature and pressure varies with the internal pressure of a liquid in which it was dissolved¹⁰. The increase of internal pressure (π) and decrease of free volume (V_f) indicates an increase of cohesive forces and vice versa in the solutions of BT₄ and BT₁₁ in both the binary systems.

The degree of interaction was also measured in relationships of solvation number (S_n). The significance of the S_n depends upon solvent-solute and solute-solute interactions. It is clear from outcomes that S_n values increased nonlinearly with C and decreased with T . The positive values of S_n indicate the structure forming tendency of BT₄ and BT₁₁ in both the binary systems. The difference in S_n with C and T values was also proposing the occurrence of strong dipole-dipole interactions.

Conclusion

The experiment has been carried out to see the influence of temperature and concentration on

Dihydropyrimidine substituted 1,2,4-triazole derivatives having the same molecular structure with a different functional group. From the experimental data, we have concluded that ρ , η and U increased with higher concentration and decreased with higher temperature. The compound BT₄ possesses -Br group, which is electron withdrawing group in nature and BT₁₁ possesses -OCH₃ group, which is electron donating group in nature and -Br group is high electronegative compared to -OCH₃ group, therefore, in some cases the behavior of both the derivatives in DMSO and DMF was found dissimilar. This was due to both groups were played an important role in molecular interactions. The derived parameters calculated from ρ , η and U data reveals that Dihydropyrimidine substituted 1,2,4-triazole derivatives acts as a structure former in the binary systems considered. While the positive value of ΔG , ΔH and ΔS signifying the occurrence of interaction between the molecules of the mixtures and the reaction was spontaneous at high temperature. Influential molecular interactions resulted in the structure forming as a judge on the foundation of positive values of solvation number.

Acknowledgments

Authors are thankful to Dr. Purvesh Dobariya, Associate Scientist, GVK Bio Sciences Pvt. Ltd, Bangalore for providing valuable guidelines for this topic.

References

- 1 **Godhani D.R., Mulani V.B. and Mehta J.P.**, Cyclization and antimicrobial evolution of 1,3,4-oxadiazoles by carbohydrazide, *World Scientific News*, **124**(2) (2019) 304-311.
- 2 **Singh M., Gangwar M., Nath G. and Singh S.K.**, Synthesis, DNA cleavage and antimicrobial activity of 4-thiazolidinones-benzothiazole conjugates, *Indian J. Exp. Biol.*, **52**(11) (2014) 1062-1070.
- 3 **Shankar K.N., Adhikari J. and Noronha S.B.**, Computer-aided solvent selection and design for the efficient extraction of a pharmaceutical molecule, *Can. J. Chem. Eng.*, **97** (2019) 1605-1618.
- 4 **Godhani D.R., Patel Y.V. and Parsania P.H.**, Ultrasonic velocity and acoustical parameters of poly (4, 4'-diphenyl phthalidediphenyl methane-4, 4'-disulfonate)-DMF solutions at 30, 35 and 40°C, *J. Pure and Appl. Ultrason.*, **23** (2001) 58-62.
- 5 **Godhani D.R., Dobariya P.B., Sanghani A.M., Jogel A.A. and Mehta J.P.**, Effect of temperature and solvents on thermo-physical properties of 1,3,4-oxadiazole derivative at atmospheric pressure, *J. Mol. Liq.*, **180** (2013) 179-186.
- 6 **Godhani D.R., Dobariya P.B., Sanghani A.M. and Mehta J.P.**, Thermo-physical properties of 1,3,4-oxadiazole derivatives in pure solvents, *J. Chem. Thermodyn.*, **57** (2013) 436-444.
- 7 **Godhani D.R., Jogel A.A., Mulani V.B., Sanghani A.M., Kukadiya N.B. and Mehta J.P.**, Effect of temperature and solvent on molecular interactions of 1,2,4-triazole derivative, *Asian J. Research Chem.*, **11**(1) (2018) 32-36.
- 8 **Godhani D.R., Mulani V.B., Mehta J.P. and Kukadiya N.B.**, Synthesis and antimicrobial evolution of some new benzotriazole substituted 1,3,4-thiadiazoles, *World Scientific News.*, **100** (2018) 51-60.
- 9 **Godhani D.R., Mulani V.B. and Mehta J.P.**, Antimicrobial, cytotoxic and haemolytic activity of newly synthesized benzotriazole substituted 1,2,4-triazoles, *Chemistry & Biology Interface.*, **9**(3) (2019) 163-171.
- 10 **Riddick J.A., Bunger W.B. and Sakano T.**, Organic Solvents-Physical Properties and Methods of Purification, *Techniques of Chemistry*, New York, (1986).
- 11 **Vigoureux P.**, Ultrasonic, Chapman and Hall, London, 112 1952.
- 12 **Kinsler L.E. and Frey A.R.**, Fundamentals of Acoustics, Wiley Eastern Ltd., New Delhi, (1978).
- 13 **Shekaari H., Bezaatpour A. and Soltanpour A.**, Partial Molar Volumes of *N,N'*-1,2-Ethyl-bis (salicyladimine) Schiff Base (Salen) in Organic Solvents at $T = (283.15 \text{ to } 318.15) \text{ K}$, *J. Chem. Eng. Data.*, **55** (2010) 5927-5931.
- 14 **Gokavi G.S., Raju J.R., Aminabhavi T.M., Balundgi R.H. and Muddapur M.V.**, Viscosities and densities of binary liquid mixtures of dimethyl sulfoxide with chlorobenzene, pyridine and methyl ethyl ketone at 25, 35, 45 and 55°C, *J. Chem. Eng. Data.*, **31** (1986) 15-18.
- 15 **Keshapolla D. and Gardas R.L.**, Apparent molar volumes and isentropic compressions of benzylalkylammonium ionic liquid in dimethyl sulfoxide from 293.15 K to 328.15 K, *Fluid Phase Equilib.*, **383** (2014) 32-42.
- 16 **Papamatthaiakis D., Aroni F. and Havredaki V.**, Isentropic compressibilities of (amide + water) mixtures: A comparative study, *J. Chem. Thermodyn.*, **40** (2008) 107-118.
- 17 **Syal V.K., Patial B.S. and Chauhan S.**, Ultrasonic velocity, viscosity and density studies in binary mixtures of dimethyl formamide and ethylmethylketone at different temperatures, *Ind. J. Pure Appl. Phys.*, **37** (1999) 366-370.
- 18 **Pal A. and Kumar A.**, Excess molar volumes and kinematic viscosities for binary mixtures of dipropylene glycol monobutyl ether and dipropylene glycol tert butyl ether with 2-pyrrolidinone, N-methyl-2-pyrrolidinone, N,N-dimethyl formamide, and N,N dimethylacetamide at 298.15 K *J. Chem. Eng. Data.*, **50**(3) (2005) 856-862.
- 19 **Baluja S. and Shah A.**, Acoustical studies of some derivatives of 4-amino antipyrine in 1,4-dioxane and dimethyl formamide at 318.15 K, *Fluid Phase Equilib.*, **215** (2004) 55-59.

Acoustical parameters of binary mixtures of 1,4-dichlorobutane with some hydrocarbons at 308.15K, 313.15K and 318.15K

Kanchan Yadav and S.S. Yadava*

Department of Chemistry, D.D.U. Gorakhpur University, Gorakhpur-273009, India

*E-mail: ssyadava1@rediffmail.com

Several acoustical parameters viz. free length (L_f), Wada's constant (W), acoustical impedance (Z), available volume (V_a) and relaxation time (τ) have been evaluated from the experimental values of ultrasound velocities (u) and densities (ρ) of the binary mixtures of 1,4-dichlorobutane with several hydrocarbons viz. cyclohexane, benzene, methylbenzene, 1,2-dimethylbenzene, 1,4-dimethylbenzene and 1,3,5-trimethylbenzene over entire mole fractions range at three temperatures 308.15K, 313.15K, and 318.15K. L_f values decrease for all the systems as molefraction of 1,4-dichlorobutane increase in the binary mixtures at the three experimental temperatures. Wada constant increase for binary mixtures of 1,4-dichlorobutane with cyclohexane, benzene and methylbenzene as mole fractions of 1,4-dichlorobutane increase and for other three binary mixtures it decrease. Z and τ values increases with molefractions of 1,4-dichlorobutane for all binary mixture except τ values for the binary mixtures with cyclohexane which decrease. V_a values for binary mixtures of 1,4-dichlorobutane with benzene, methylbenzene and 1,2-dimethylbenzene increase and for other three experimental binary systems decrease as molefractions of 1,4-dichlorobenzene increase.

Keywords: Ultrasound velocity, binary mixtures, 1,4-dichlorobutane, aromatic hydrocarbons.

Introduction

Experimental values of ultrasound velocity and density for the selected binary mixtures have already been published¹. However, several acoustical parameters have not been analysed. It is therefore, thought worth while to evaluate several acoustical parameters viz. intermolecular free length (L_f), Wada's constant (W), acoustical impedance (Z), available volume (V_a), and relaxation time (τ) for the binary mixtures of 1,4-dichlorobutane with cyclohexane, benzene, methyl benzene, 1,2-dimethylbenzene, 1,4-dimethylbenzene and 1,3,5-trimethyl benzene at the experimental temperatures.

Materials and method

Chemicals used were cyclohexane (HPLC and chromatography grade, 98.7%), benzene (Merck, 99.7%), methylbenzene (BDH, 99.96%), 1,2-dimethyl benzene (Merck, $\geq 98\%$), 1,4-dimethylbenzene (Merck, $\geq 99\%$), 1,3,5-trimethylbenzene (Merck, $\geq 98\%$) and 1,4-dichlorobutane (Merck, $\geq 98\%$). These chemicals

were further purified by distilling over different size columns (viz. for cyclohexane and benzene 100 cm long column, for methylbenzene 60 cm long column and for remaining others 30 cm long column) and only middle fractions were collected and used for binary sample preparation. Purities of the distilled chemicals were checked by comparing experimental values of densities with the literature values. A good agreement was found between the two values as evident from table 1²⁻⁶.

Table 1 – Experimental and literature values of densities (ρ) at temperature 308.15 K.

Density (ρ) (k gm ⁻³)		
Compound	Experimental	Literature
Cyclohexane	765.9	762.5 [2], 764.40 [3]
Benzene	864.0	862.92 [4], 863.02 [5]
Methylbenzene	854.1	852.93[4], 850.90[6]
1,2-dimethylbenzene	871.4	869.40[2], 867.05[4]
1,4-dimethylbenzene	849.3	848.12[4], 847.50[6]
1,3,5-trimethylbenzene	853.3	853.63[4], 852.60[6]
1,4-dichlorobutane	1123.4	-----

Binary liquid mixtures of 1,4-dichlorobutane with hydrocarbon solvents were prepared gravimetrically covering whole mole fraction range. The component liquids were injected into glass sealed vials employing syringes with 24 number needles to avoid evaporation losses during sample preparation. All the masses were measured within $\pm 1 \times 10^{-8}$ kg by using single pan analytical balance.

Ultrasound velocities and densities were measured as described elsewhere¹.

Intermolecular free length, L_f which is the distance between the surface of molecules is evaluated by equation (1),

$$L_f = K/u \cdot \rho^{1/2} \quad (1)$$

Where 'K' is temperature dependent constant known as Jacobson's constant. The adiabatic compressibility $\beta_{ad} = u^{-2} \cdot \rho^{-1}$ is used to evaluate Wada's constant, W from the following equation,

$$W = M/\rho (\beta_{ad})^{-1/7} \quad (2)$$

Where M is the molar mass of the experimental liquid. It has been suggested by Kinsler *et al.*⁷ that acoustic impedance, Z given by Eq. (3),

$$Z = u \cdot \rho \quad (3)$$

is more significant parameters describing a medium and intermolecular interaction than ultrasound velocity and density individually. Available volume, V_a is the difference of molar volumes at experimental absolute temperature, V_T and absolute zero temperature, V_0 i.e., ($V_a = V_T - V_0$). On the basis of collision factor theory, Schaaff's equation for the evaluation of available volume is

$$V_a = V_T [1 - u/u_\infty] \quad (4)$$

Where u_∞ is the limiting value of ultrasound velocity when the entire volume of space in liquid is filled with molecules and is taken as $1,600 \text{ ms}^{-1}$. Experimental values of ultrasound velocity and density have been used to evaluate relaxation time, τ by Eq. (5).

$$\tau = (4/3)(\eta/\rho)(1/u^2) \quad (5)$$

Where η is viscosity of the liquid at experimental temperature.

Results and Discussion

The values of intermolecular free length, L_f calculated by Eq. (1) for all the binary mixtures at the three experimental temperatures 308.15 K, 313.15 K, 318.15 K as a function of molefractions of 1,4-dichlorobutane

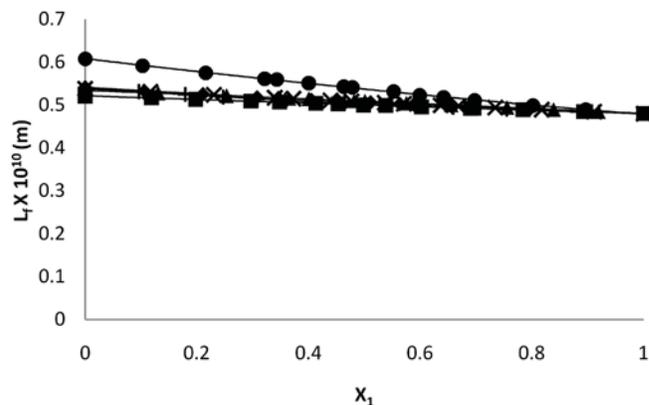


Fig. 1. Intermolecular free lengths (L_f) with molefractions (x_1) of 1,4-dichlorobutane for binary mixtures of 1,4-dichlorobutane + cyclohexane (●), + benzene (+), + methylbenzene (◆), + 1,2-dimethylbenzene (■), + 1,4-dimethylbenzene (x) and + 1,3,5-trimethylbenzene (▲) at temperature 308.15K.

(x_1) are shown at 308.15K only in Fig. 1 for the sake of brevity. It has been observed that L_f values decrease for all the studied systems with increase in molefractions of 1,4-dichlorobutane in the binary mixtures at the three experimental temperatures. The variations of Wada's constant, W evaluated by Eq. (2) at the three experimental temperatures with molefractions (x_1) of 1,4-dichlorobutane for all the systems studied at 308.15K only for the sake of brevity are shown in Fig.2. Figure 2 shows that values of W increase for binary mixtures prepared with cyclohexane, benzene, and methylbenzene at the temperature 308.15K while it decrease for binary mixtures of 1,4-dichlorobutane with 1,2-dimethylbenzene, 1,4-dimethylbenzene and 1,3,5-

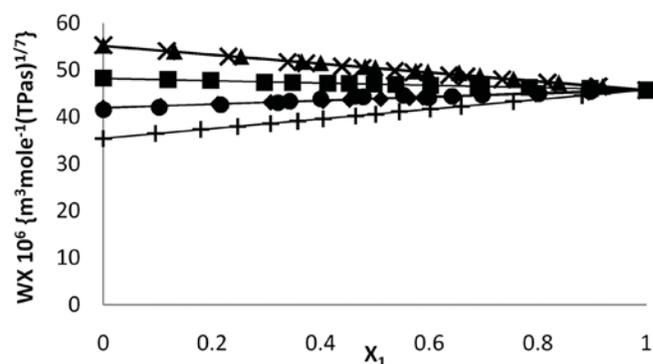


Fig. 2. Wada's constant (W) with molefractions (x_1) of 1,4-dichlorobutane for binary mixtures of 1,4-dichlorobutane + cyclohexane (●), + benzene (+), + methylbenzene (◆), + 1,2-dimethylbenzene (■), + 1,4-dimethylbenzene (x) and + 1,3,5-trimethylbenzene (▲) at temperature 308.15K.

trimethylbenzene. Further observation of the figures 2 reveals that W values are almost similar at any fixed composition for binary mixtures with cyclohexane and methylbenzene and for 1,4-dimethylbenzene and 1,3,5-trimethylbenzene. It is also evident that at every composition of the binary mixture, W values follow the trend benzene < cyclohexane \approx methylbenzene < 1,2-dimethylbenzene < 1,4-dimethylbenzene \approx 1,3,5-trimethylbenzene

Similar behavior of variations of W with x_1 is observed at 313.15K and 318.15K.

The values of acoustical impedance, Z at all the three experimental temperatures for the studied binary systems are shown in figure 3. Again for the sake of brevity the variations of Z with composition of mixtures at only one temperature 308.15 K are shown in Fig. 3. Figure 3 shows that Z values increase with molefractions of 1,4-dichlorobutane for binary mixture prepared with all the six hydrocarbon solvents. Z values are almost similar for binary mixtures prepared with all the aromatic hydrocarbons and are lesser than that for binary mixture with cyclohexane. The available volumes V_a are evaluated by equation 4, for all the binary mixtures studied at the three experimental temperatures 308.15 K, 313.15K and 318.15K. The variations of V_a with composition of mixtures are shown in Fig. 4 at only one temperature 308.15K. The values of V_a for binary mixtures of 1,4-dichlorobutane with benzene, methylbenzene and 1,2-dimethylbenzene increase with increase in molefractions of 1,4-dichlorobutane at all the three experimental temperatures. It has been found to decrease with molefractions of 1,4-dichlorobutane

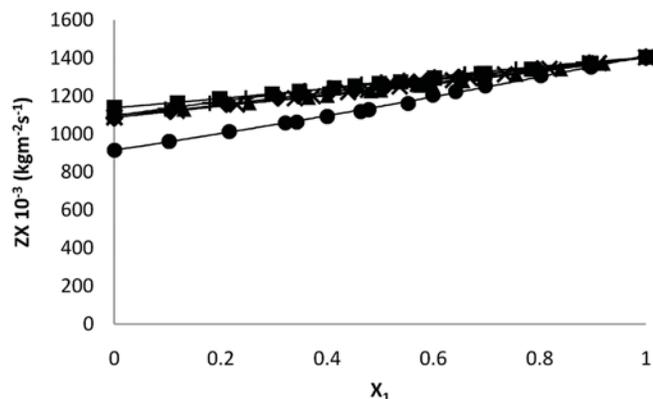


Fig. 3. Acoustical impedances (Z) with molefractions (x_1) of 1,4-dichlorobutane for binary mixtures of 1,4-dichlorobutane + cyclohexane (\bullet), + benzene (+), + methylbenzene (\blacklozenge), + 1,2- dimethylbenzene (\blacksquare), + 1,4-dimethylbenzene (\times) and + 1,3,5-trimethylbenzene (\blacktriangle) at temperature 308.15K.

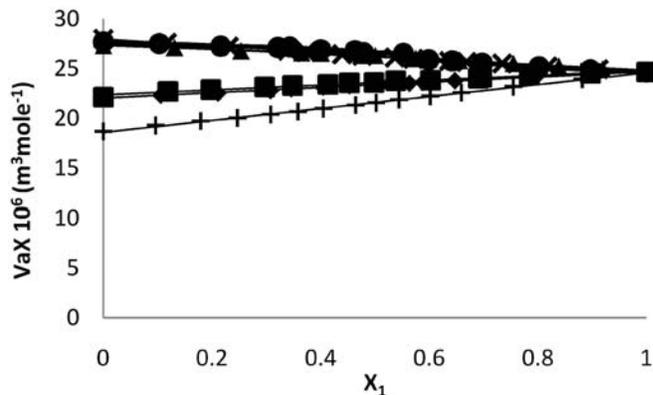


Fig. 4. Available volumes (V_a) with molefractions (x_1) of 1,4-dichlorobutane for binary mixtures of 1,4-dichlorobutane + cyclohexane (\bullet), + benzene (+), + methylbenzene (\blacklozenge), + 1,2- dimethylbenzene (\blacksquare), + 1,4-dimethylbenzene (\times) and + 1,3,5-trimethylbenzene (\blacktriangle) at temperature 308.15K.

for binary mixtures with cyclohexane, 1,4- dimethylbenzene and 1,3,5-trimethylbenzene. It has also been found that V_a values are almost similar at every composition for binary mixtures of 1,4-dichlorobutane + cyclohexane, + 1,4-dimethylbenzene and + 1,3,5-trimethylbenzene at the three experimental temperatures. V_a values for binary mixtures of 1,4-dichlorobutane with methylbenzene and with 1,2-dimethylbenzene are also almost similar at every composition and at all the experimental temperatures. Experimental values of ultrasound velocities, densities¹ and viscosities⁸ are used to evaluate relaxation time, τ with Eq. (5) at the three

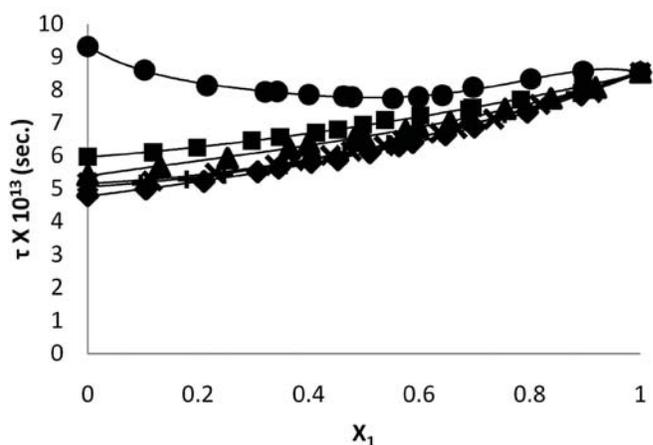


Fig. 5. Relaxation time (τ) with molefractions (x_1) of 1,4-dichlorobutane for binary mixtures of 1,4-dichlorobutane + cyclohexane (\bullet), + benzene (+), + methylbenzene (\blacklozenge), + 1,2- dimethylbenzene (\blacksquare), + 1,4-dimethylbenzene (\times) and + 1,3,5-trimethylbenzene (\blacktriangle) at temperature 308.15K.

experimental temperatures. Variations of τ values with composition of the mixture for all the binary mixtures with molefractions of 1,4-dichlorobutane are shown at 308.15 K only in Fig. 5. Figure 5 shows that τ values increase with increase in molefractions of 1,4-dichlorobutane in the mixture for all the systems studied. However for binary mixtures of 1,4-dichlorobutane + cyclohexane, τ values first decrease and at higher concentration of 1,4-dichlorobutane, it increases. τ values are larger at every composition of binary mixtures prepared with cyclohexane than that with all the aromatic hydrocarbon solvents.

It has been suggested by Eyring *et al.*⁹ and others¹⁰ that variation of ultrasound velocity in mixture of liquids depends upon the increase or decrease of free length, L_f after mixing of the components. The ultrasound velocities increase if intermolecular free lengths decrease and vice versa. It is evident from figure 1 that in our case the binary mixture of 1,4-dichlorobutane with cyclohexane follows the above trend *i.e.* ultrasound velocity values increase and intermolecular free lengths decrease with molefractions of 1,4-dichlorobutane. However, binary mixtures of 1,4-dichlorobutane with other aromatic hydrocarbon solvents do not follow the reverse trend of u and L_f . In our case both decrease as molefractions of 1,4-dichlorobutane in the mixtures increase. However, it may be noticed that the slope of the straight line for L_f verses mole fractions (x_1) of 1,4-dichlorobutane has reduced to a considerable extent than that for the straight lines between u verses x_1 .

Conclusion

Intermolecular free lengths (L_f) decrease for all the binary mixture with molefractions (x_1) of 1,4-dichlorobutane at the three experimental temperatures. Acoustical impedences (Z) increase for all the binary mixtures with x_1 at the three experimental temperatures. Available volumes (V_a) increase with x_1 for the binary mixtures of 1,4-dichlorobutane + benzene, + methylbenzene and + 1,2-dimethylbenzene. For other mixture it decrease.

Acknowledgment

The authors are thankful to the U.G.C., New Delhi for financial assistance in the form of major research project no. 41-341/2012 (SR). The authors are also thankful to the Head, Department of Chemistry, D.D.U. Gorakhpur University, Gorakhpur for providing laboratory facilities.

The authors are also thankful to Prof. Sudha Yadav, Chemistry Department, D. D. U. Gorakhpur University for her valuable cooperation.

References

- 1 **Yadav K. and Yadava S. S.**, Ultrasonic studies on binary mixtures of 1,4-dichlorobutane with some hydrocarbons at 308.15K, 313.15K, 318.15K, *J. Pure App. Ultrason.* **40** (2018) 1-7.
- 2 **Narendra K., Narayanamurthy P. and Srinivasu C.H.**, Evaluation of excess thermodynamics parameters in a binary liquid mixtures (Cyclohexane + O-xylene) at different temperatures. *E. J. of chem.* **7**(3) (2010) 927-934.
- 3 **Rathnam M.V., Mankumare S. and Kumar M.S.S.**, Density, Viscosity and Speed of Sound of (Methyl benzoate + Cyclohexane), (Methyl benzoate + n-Hexane), (Methyl benzoate + Heptane), and (Methyl benzoate + Octane) at temp. of (303.15, 308.15, and 318.15)K, *J. Chem. Eng. Data* **55** (2010) 1354-1358.
- 4 **Nain A.K.**, Densities and Volumetric properties of binary mixtures of tetrahydrofuran with some Aromatic hydrocarbons at temperature from 278.15 to 318.15K, *J. Sol. Chem.* **35** (2006) 1417-1439.
- 5 **Yadava S.S., Singh Y. and Kushwaha N.**, Densities and Viscosities of alkylethanoates + Cyclohexane + Benzene + 1,4-dimethylbenzene and 1,3,5-trimethyl benzene at 308.15, *J. Actachim. Slov.*, **57** (2010) 707-715.
- 6 **Nayak J.N., Aralaguppi M.I. and Aminabhavi T.M.**, Density, Viscosity, Refractive Index and Speed of sound in the binary mixtures of Ethyl Chloroacetate with aromatic liquids at 298.15, 303.15, and 308.15K, *J. Chem. Eng. Data.*, **47**(4) (2002) (964).
- 7 **Kinsler L.E. and Frey A. R.**, Fundamentals of Acoustics, Wiley, New York (1962) 122.
- 8 **Yadav K. and Yadava S. S.**, Viscosities and excess molar volumes of binary liquid mixtures of 1,4-dichlorobutane and some hydrocarbon solvents at 308.15K, 313.15K, 318.15K, *Physics and Chemistry of Liquids*, **57**(3) (2018) 325-337.
- 9 **Eyring H. and Kincaid J.F.**, Free volumes and free angle ratios of molecules in liquids, *J. Chem. Phys.*, **6** (1938) 620.
- 10 **Ali A., Hyder S. and Nain A.K.**, Intermolecular interactions in ternary liquid mixture by ultrasonic velocity measurements, *Ind. J. Phys.*, **74B** (2000) 63-67.

Estimation of Debye temperature for binary liquid mixtures using ultrasonic techniques

A. Mathana Gopal¹, A. Moses Ezhil Raj² and J. Poongodi^{3*}

¹Department of Physics, Kamaraj College, Tuticorin-628003, India

²Department of Physics, Scott Christian College, Nagercoil-629003, India

³Affiliated to Manonmaniam Sundaranar University, Tirunelveli-627012, India

*E-mail: poongodinagaraj@gmail.com

Ultrasonic velocity measurements provide an important tool to study the liquid state. Such studies have been used to understand intra and intermolecular interactions in pure liquids, aqueous solutions and binary mixtures. Detailed information about the nature of molecular interactions can be obtained from various ultrasonic parameter computed. It has inspired many workers and extensive investigations have been carried out in both binary and ternary liquid mixtures using various methods. Acoustic methods have been applied to estimate the effective Debye temperature (θ_D) of liquid mixtures (binary). The Theoretical approach has been completely modified and a new approach has been suggested. Binary liquid mixtures have been undertaken for computing the values of effective Debye temperature using the present approach. There was a good agreement with the theoretical values compared with standard findings. The necessary experimental data needed have been taken from literature.

Keywords: Ultrasonic velocity, adiabatic compressibility, molar volume, Debye temperature.

Introduction

The properties of the liquids can be studied by measuring the ultrasonic velocity in liquids. The ultrasonic velocity in liquids is a dynamic description of the nature of the liquid and hence its measurements add valuable information regarding the specific interaction in a liquid. Several workers have reported sound velocity measurements in associated liquids and its mixtures and obtained the various thermo dynamical parameters like excess sound velocity, excess molar volume and excess free volume. One important difficulty in liquid state theories is that the knowledge of the microscopic intermolecular forces alone is not sufficient to interpret the bulk properties of liquids. Quite often, the predictions of various theoretical models are found to be at variance with the bulk properties. The literature on H-bonding of liquids by NMR and other spectroscopic techniques do provide useful information for the classification of the associated liquids, at the microscopic level.

The study of molecular interactions plays an important role in understanding the structure and properties of

liquids, solids and gases. The intermolecular forces that are responsible for the interactions can be classified into two types namely long range and short range forces. The long range forces are electrostatic, inductive and dispersion forces which mainly depend on the intermolecular separation. The short range forces, called chemical forces arising due to overlap of electron clouds, are often highly directional².

The elastic constants of a liquid are related to the internal and configuration energy. Thus the ease with which one can distort the fluid structure is determined by the energy holding the lattice together. In H-bonded liquids, it is reasonable to assume that this internal energy is directly related to the number of H bonds per unit volume. The ultrasonic wave propagation in such liquids results in the distortion of the equilibrium of the holes and sites in the liquid as well as the structural orientation of H-bonded species³⁻⁵.

Theory

The basic equation employed for the calculation of the effective Debye temperature (θ_D)⁶, can be written

$$\text{as: } \theta_D = \frac{h}{k} \left[\left(\frac{9N}{4\pi V} \right) / \left(\frac{1}{C_1^3} + \frac{2}{C_1^3} \right) \right]^{1/3} \quad (1)$$

Where all the symbols have their usual notations, without going into further details it can be shown that the resulting expression for computing⁷⁻⁸.

Table 1 – Values of adiabatic compressibility (β_{ad}), molar volume (V) and Debye temperature (θ_D) for pure liquids at 303K

Liquids	ρ kkgm ⁻³	γ	$\beta_{ad} \times 10^{-10}$ N ⁻¹ m ²	V μm^3	θ_D (K)
Butyl Acetate	0.869	1.280	8.656	133.30	60.96
Ethanol	0.779	1.179	10.570	59.10	73.44
Butanol	0.778	1.151	9.487	95.28	65.28
3 Methyl -1-butanol	0.804	1.156	8.637	109.59	64.32
2 Pentanol	0.801	1.154	8.598	110.10	64.80

Table 2 – Values of adiabatic compressibility (β_{ad}), molar volume (V) and Debye temperature (θ_D) for liquid mixtures at 303K

X_2	ρ kkgm ⁻³	γ	$\beta_{ad} \times 10^{-10}$ N ⁻¹ m ²	V μm^3	θ_D (K) Eqn. 2	θ_D (K) Eqn. 3	θ_D (K) Eqn. 4
System I: n - Butyl Acetate + Ethanol							
0.000	0.779	1.179	10.570	59.10	73.44	73.44	73.44
0.103	0.795	1.195	10.210	66.94	71.52	72.15	71.08
0.301	0.824	1.216	9.675	81.37	68.16	69.68	67.61
0.500	0.840	1.236	9.356	96.36	65.28	67.20	65.07
0.699	0.851	1.253	9.186	111.43	62.88	64.72	63.14
0.895	0.859	1.268	9.053	126.32	60.96	62.27	60.64
1.000	0.869	1.280	8.656	133.30	60.96	60.96	60.96
System II: n - Butyl Acetate + Butanol							
0.000	0.778	1.151	9.487	95.28	65.28	65.28	65.25
0.092	0.806	1.162	9.187	96.76	65.28	64.88	64.86
0.301	0.822	1.185	9.167	105.55	63.36	63.98	63.92
0.509	0.836	1.209	9.156	114.22	61.92	63.08	63.01
0.680	0.850	1.231	9.053	120.78	61.44	62.34	62.28
0.901	0.863	1.262	8.948	129.70	60.48	61.39	61.36
1.000	0.869	1.280	8.656	133.60	60.96	60.96	60.96
System III: n - Butyl Acetate + 3 Methyl - 1 - butanol							
0.000	0.804	1.156	8.637	109.59	64.32	64.32	64.32
0.097	0.814	1.167	8.603	111.59	64.32	63.99	63.97
0.296	0.826	1.187	8.754	116.75	62.88	63.32	63.27
0.497	0.841	1.211	8.761	121.39	61.92	62.95	62.58
0.716	0.853	1.243	8.682	126.90	61.44	61.91	61.86
0.899	0.866	1.269	8.641	130.94	61.44	61.30	61.28
1.000	0.870	1.280	8.646	133.60	60.96	60.96	60.96
System IV: n - Butyl Acetate + 2 Pentanol							
0.000	0.801	1.154	8.598	110.10	64.80	64.80	64.80
0.094	0.805	1.161	8.787	112.81	63.36	64.44	64.45
0.296	0.819	1.183	8.844	117.76	62.40	63.66	63.70
0.494	0.833	1.206	8.906	122.42	61.44	62.90	62.95
0.693	0.846	1.234	8.845	127.10	60.96	62.14	62.18
0.898	0.859	1.262	8.895	137.83	60.96	61.35	61.37
1.000	0.869	1.280	8.656	133.59	60.96	60.96	60.96

$$\theta_D = \frac{h}{k} \left[\frac{(9N / 4\pi V)}{(\rho\beta_a)^{3/2} \left(\frac{1}{1+\gamma} \right)^{3/2} + 2 \left(\frac{4}{3\gamma} \right)^{3/2}} \right]^{1/3} \quad (2)$$

$$\theta_D = \sum X_i \theta_{Di} \quad (3)$$

$$\theta_D = \frac{\sum_{x1} C_{pi} \theta_{D1}}{\sum_{x1} C_{pi}} \quad (4)$$

Where C_{pi} is the molar specific heat capacity at constant pressure of the i th component. Using these methods we have calculated the Debye temperature of mixture and compared with experimental findings.

Results and Discussion

The values of Debye Temperature (θ_D) for four binary liquid systems n - Butyl Acetate + Ethanol, n-Butyl Acetate + Butanol, n - Butyl Acetate + 3 Methyl - 1 - butanol and n - Butyl Acetate + 2 Pentanol have been computed with a help of the above procedures. The required data needed for the computations are taken from literature⁹. Equations 2, 3 and 4 have been used to obtain the value of Debye Temperature (θ_D) of the liquid mixtures. θ_D computed by the two methods 3 and 4 are compared with the standard values obtained with the help of the equation 2. Here Lorentz - Bertholet combination rule gives very fine result when compared to other results. The ideal mixing relation using equation 3 shows large deviation for these systems. This may be due to the strong H-bonding in the liquid mixtures. In all the systems under our investigation the value of

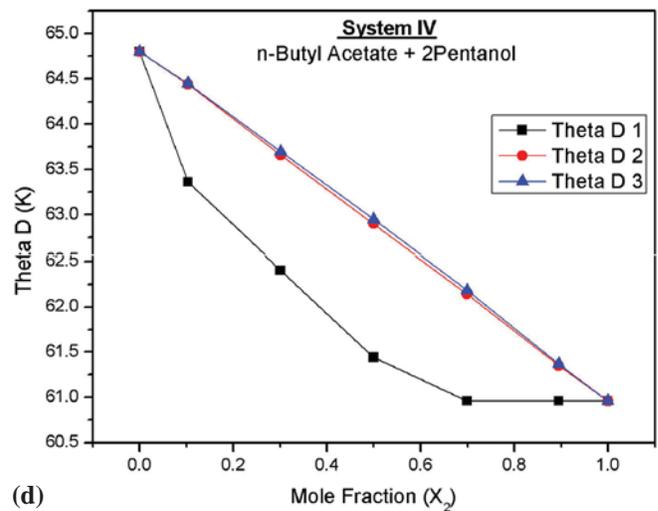
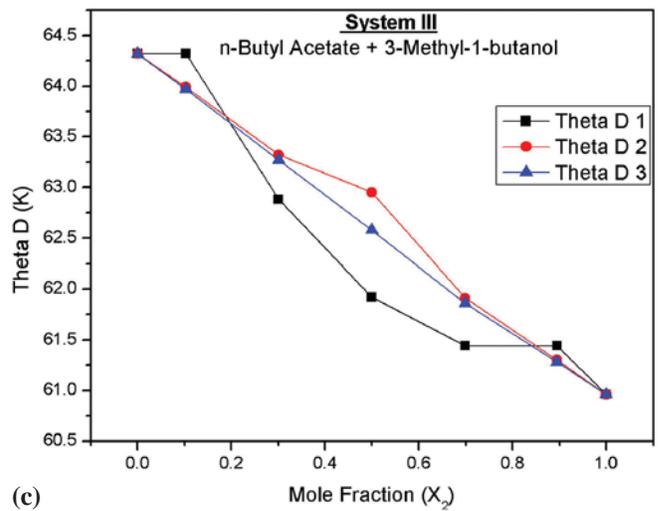
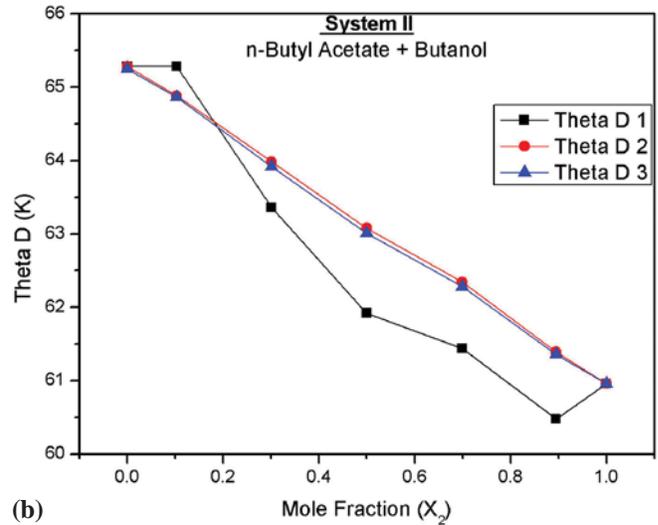
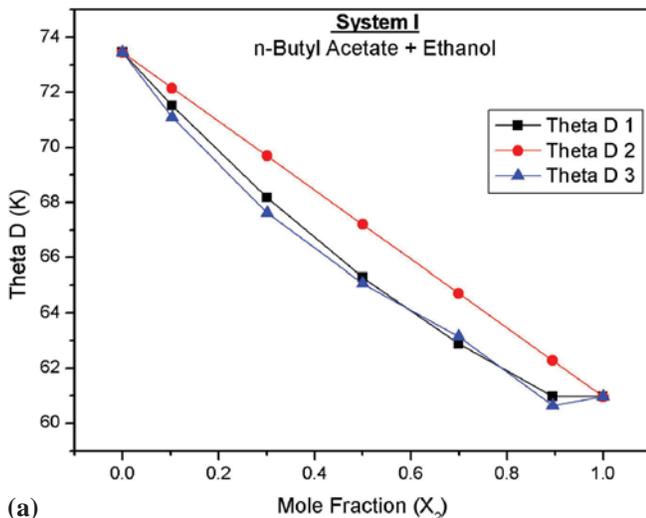


Fig. 1(a-d): Variations of Debye Temperature (θ_D) for n-Butyl Acetate with Ethanol (a), Methanol (b), 3-Methyl-1-butanol (c), 2-Pentanol (d) at 303K

Debye temperature decreases with increases of mole fraction. Here an attempt has been made to study the effective Debye temperature of Pure liquids and also the multi component liquid mixtures. The variation of θ_D with mole fraction is shown in figure 1. It is concluded that on the basis of quasi crystalline structure and also with experimental evidences it is possible to estimate the Debye temperature in liquids. Previously, these calculations were made only for liquids.

References

- 1 **Eyring H.** and **Kincaid J.F.**, Free volume and free angle ratios of molecules in liquids, *J. Chem, Phys*, **6** (1938) 620-629.
- 2 **Pandey J.D.**, Ultrasonic propagation parameters and the effective Debye temperature in liquid mixtures, *Acoustic Letters*, **3** (1970) 90-94.
- 3 **Altar W.**, A study of the liquid state, *J. Chem. Phys.* **5** (1937) 577-586.
- 4 **Eyring H.** and **Bresler.**, Viscosity, Plasticity and Diffusion as examples of Absolute reaction rates, *J. Chem, Phys.*, **4** (1936) 283-291.
- 5 **Pandey J.D.**, Effective Debye temperature of and specific heat ratio in liquid methane and pentane, *Ind. J. Chem*, **14A** (1976) 607-608.
- 6 **Satendra S., Aruna S.** and **Shailendra B.**, Computation of Effective Debye Temperature (θ_D) of binary liquid mixtures, *Int. J. Res. Pharm Chem.*, **7**(3) (2017) 302-305.
- 7 **Pandey J.D., Singh A.K.** and **Day R.**, Effects of isotropy on thermo acoustical properties, *Ind. J. Pure Appl. Ultrason*, **26** (2006) 100-104.
- 8 **Pandey J.D., Sanguri V., Mishra R.K.** and **Singh A.K.**, Acoustic method for the estimation of effective debye temperature of binary and ternary liquid mixtures, *J. Pure Appl. Ultrason*, **26** (2004) 18-29.
- 9 **Unnamalai K.**, A Study of Physico Acoustical Parameters in Complexes of alcohols in n-Butyl Acetate, M.Phil. Thesis. Alagappa University, July 1992.

Study of highly viscous polyacryle measurements of viscosity and ultrasonic velocity

S.V. Khangar^{1,*}, O.P. Chimankar², D.V. Nandanwar³,
J.N. Ramteke³ and Swapnil A Kale³

¹Department of Physics, Mahatma Fule Arts, Commerce & Sitaramji Chaudhari Science College, Warud-444906, India

²Department of Physics, RTM Nagpur University, Nagpur-440033, india

³Department of Physics, Shri Mathuradas Mohota College of Science, Nagpur-440009, india

*E-mail: sugandhakhangar2000@gmail.com

In order to perform the polymerization process, it is necessary to measure viscosity. In case of highly viscous fluid, viscosity starts to be dependent of the vibration and rotation frequency of the sensing element. The ultrasonic technique provides an effective and reliable tool to investigate properties of polymer solutions in the light of phase separation studies. The propagation of ultrasonic waves and the measurement of their viscosity in solution form an important tool for the evaluation of various acoustical and thermo-dynamical parameters which give an insight into the nature of miscibility/compatibility and molecular interactions in polymer solution. The phenomenon polymer-solvent miscibility may arises due to any specific molecular interactions such as hydrogen bonding, dipole- dipole interactions and charge transfer complexes for homogeneous polymer- solvent mixture. Miscibility is an important phenomenon in polymer solution to achieve mechanical integrity, better adhesion and better processing. In present research work we have chosen polyacrylamide solution as a highly viscous fluid.

Keywords: Viscosity, miscibility, polyacrylamide solution, pulse echo technique.

Introduction

Propagation of ultrasonic waves in polymer solution forms the basis of qualitative characterization. Acoustical studies in polymer solutions and in solid polymers have been the subject of research in recent years¹. Ultrasonic is the universally accepted nondestructive technique to study the physical & chemical properties of the liquids, liquid mixtures, electrolytic solutions and polymeric solutions². The different acoustical parameters interpret the nature and strength of molecular interaction that exist in the system. Being fast, nondestructive and versatile, ultrasonic technique is useful for testing structure-property relationship of macromolecules. Recently, studies in organic polymer have attracted the attention of large number of workers³⁻⁵.

Exhaustive literature survey was carried out and it was found that not much work has been reported in the field of ultrasonic studies in polyacrylamide polymers.

This has motivated us to carry out the ultrasonic measurements in such polymers.

In present research work we have chosen polyacrylamide solution as a highly viscous fluid. Polyacrylamide is a carbon- carbon chain water-soluble polymer and is used as a thickening agent and a flocculent. Polyacrylamide (PAA) solutions of different concentrations have been prepared with water and their miscibility investigated with ultrasonic pulse echo technique. Thermo acoustic parameters ultrasonic velocity (u), density (ρ), viscosity (η) for polyacrylamide solutions is measured at different concentrations (Wt %) *i.e.* 0.05 Wt % 0.3 Wt %. The measurements were carried out at 288 K to 308 K and at frequencies 2 MHz and 5 MHz. The results are reported. Variation in viscosity (η) is found to be linear. These linear variation confirms the miscibility of polyacrylamide in water solution at that concentration range.

Experimental

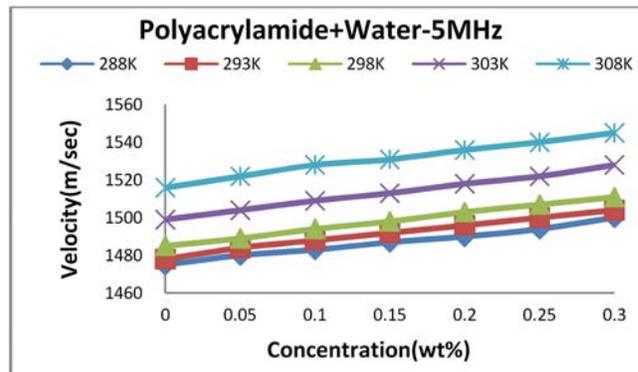
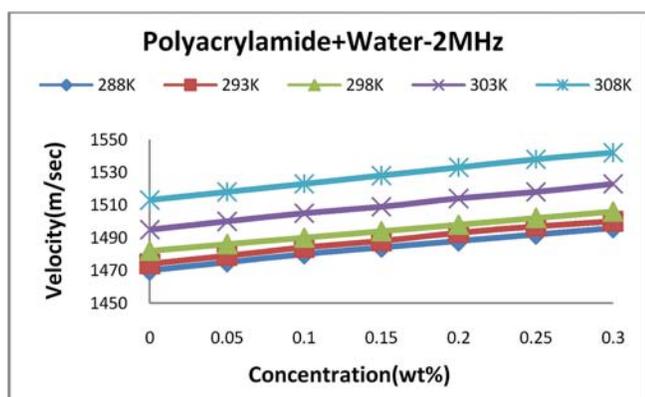
AR grade Polyacrylamide is used in present study. Ultrasonic velocity, density and viscosity values are estimated in the aqueous polyacrylamide solution. The solution was prepared by adding a known weight of polyacrylamide to a fixed volume of distilled water and then stirred until clear solution was obtained. The concentration range chosen in the solution are 0.05, 0.1, 0.15, 0.2, 0.25, & 0.3 wt.%. Velocity measurements are carried out using ultrasonic pulse echo technique by using MHF-400 High frequency pulser - receiver at frequencies of 2 MHz & 5 MHz and at temperature range 288 K - 308 K with an accuracy of ± 1 m/sec. Density has been measured by pycnometer method. The viscosity of liquid was measured by Oswald's viscometer. Temperature is maintained at a constant range by Plasto Crafts Thermostat and other related parameters calculated by standard formulae.

Formulae

1. Density $\rho = \frac{M_1}{M_w} \rho_w$. Where M_1 - mass of experimental liquid, M_w - mass of water & ρ_w - density of water
2. Viscosity $\eta = \frac{\rho_1 t_1}{\rho_w t_w} \eta_w$

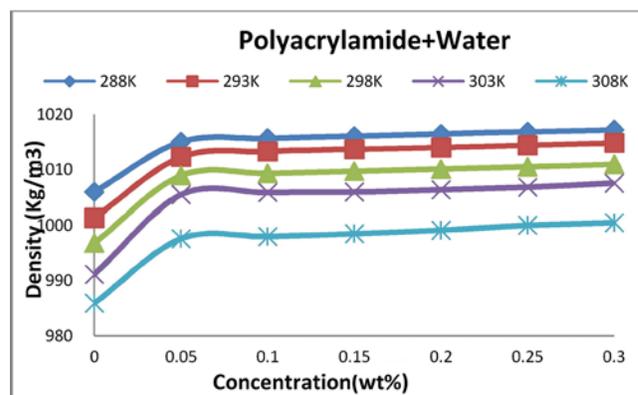
Results and Discussion

The ultrasonic velocity (u) increases linearly with the concentration of PAA in water. It is maximum at higher concentration (0.3 wt. %). This linear relationship between velocity and concentration of solution suggest its miscible behavior⁷. In PAA solutions, hydrogen bonds are formed between the solute and solvent molecules. When PAA is dissolved in water, the ultrasonic velocity



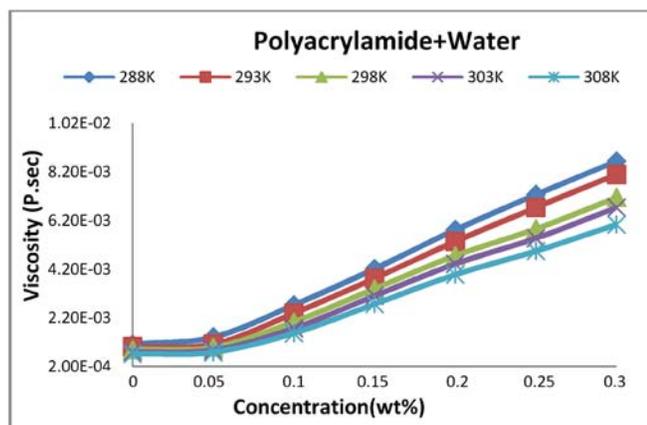
increases, because at this stage polyacrylamide must be well dispersed hence at this stage compressibility is minimum, confirms the association of PAA molecule with water.

It is observed that density increases with increase in concentration of polyacrylamide (PAA) in water, increase in density decreases the volume indicating association in component molecules. This increase in density is due to the fact that the number of polymer chain added to the solution increases with increase in concentration of polymer solution. Polymers are characterized by large molecular weight compared to the solvents. This also contributes to increase in the density of the solution⁸.



It is observed that viscosity linearly increases with increase in concentration (wt. %) of polyacrylamide (PAA) in water. According to Kauzman and Eyring⁹, the viscosity of a mixture strongly depends on the entropy of mixture, which is related with the liquid structure as well as molecular interactions between the components of the mixtures. Thus the viscosity depends upon the molecular interaction as well as on the shape and size of the molecules.

Such water soluble polyacrylamide used to encourage the flocculation of particles in water & such flocculate polyacrylamide is used for waste water treatment industry³. Polyacrylamide + water at particular (0.05%-0.3%) concentration range & temperature range can be used in all solid -liquid separation process in general and in municipal' waste water treatment.



Conclusion

- As water soluble polyacrylamide used to increase the viscosity of water or to encourage the flocculation of particles in water & such flocculate polyacrylamide is used for waste water treatment industry. Polyacrylamide + water at particular concentration & temperature can be used in all solid -liquid separation process in general and in municipal' waste water treatment.
- PAA flocculate react with waste water to form insoluble hydroxide which on precipitating, link together to form larger floc. The floc can be filtered or removed more easily.
- The viscous solution of polyacrylamide can be used for the oil production in oil industry.
- The linear variation of ultrasonic velocity with concentration gives the evidence to enhance the compatibility among the molecules presence in the PAA solution. .
- The hydration state and its dynamic behavior are also explained in the light of formation of inter or intra-molecular hydrogen bonds between adjacent OH groups.
- The linearity in the viscosity indicates there is a

single phase formation. It predicts strong polymer-solvent interaction and strong association in the PAA solution.

- Ultrasonic velocity determine the elasticity of medium.

References

- Khangar S.V., Chimankar O.P. and Bhandakkar V.D.,** Molecular interaction in binary mixtures, *Glob. J. Res. Analy.*, **3** (2014) 73-76.
- Khangar S.V. and Chimankar O.P.,** Adiabatic compressibility and salvation of drug reducing polymers in aqueous solutions, *Int. J. Phys. Sci.*, **4** (2014) 1351-1357.
- Khangar S.V. Chimankar O.P., Shriwas R.S. and Patil S.,** Ultrasonic investigation of dipole- dipole interaction in binary solution of cellulose acetate + water, *Int. J. Sci. Res.* pp. 331-334, <https://www.ijsr.net/conf/ISU-2015/ISU-075.pdf>
- Mather M.L., Collings A.F., Bajenov N., Whittaker A.K. and Baldock C.,** Ultrasonic absorption in polymer gel dosimeters, *Ultrasonics*, **41**(2003) 551-559.
- Majumdar S., Holay, S.H. and Singh, R.P.,** Adiabatic compressibility and salvation of drug reducing polymers in aqueous solutions, *Eur. Poly. J.*, **16** (1980) 1201-1204.
- Schmelzer C.E.H., Zwirbla W., Rosenfeld E. and Linde B.B.J.,** Acoustic investigations of pseudo stable structures in aqueous solutions of polyethylene glycols. *J. Mol. Struct.*, **47** (2004) 699-702.
- Bell W. and Pethrick R.A.,** Adiabatic compressibility of polymer solutions, *Eur. Poly. J.*, **8** (1972) 927-930.
- Falguni D.K. and Parsania P.H.,** Ultrasonic velocity studies and allied parameters of poly (4-4'-cyclohexylidene- R-R'-diphenylene diphenylmethane-4-4' disulfonate) solutions at 30, *Eur. Polym. J.*, **36** (2000) 519-521.
- Ramanathan K. and Ravichandran S.,** Ultrasonic study of mixed salt solutions of ammonium and ammonium chloride solution, *Ind. J. Pure Appl. Ultrason.*, **26** (2004) 12-16.
- Choudary N.V. , Ramamurthy M., Sastry G.S. and Naidu P.R.,** Ultrasonic studies in binary liquid mixtures with 1, 2,2 tetrachloride ethanease common component, *Ind. J. Pure Appl. Phys.*, **22** (1984) 409-415.

Author Index

Agarwal, Rekha	44	Nandanwar, D.V.	94
Arora, G.K.	25, 26	Pandey, Dharmendra Kumar	1, 57
Banerjee, A.	74	Pawar, Mrunal	74
Bhandakkar, V.D.	14	Pawar, N. R.	14, 74
Bhat, V.R.	14	Poongodi, J.	24, 90
Chavhan, R.D.	74	Ramteke, J.N.	94
Chimankar, O.P.	14, 74, 94	Sarangi, A.	51
Dhawan, P.K.	69	Saxena, Indu	36
Dey, Ranjan	19	Sharma, Arun Kumar	63
Dhoble, S.J.	74	Sharma, Rashmi	63
Dorik, R.G.	27	Singh, Arvind Kumar	19
Gangwal, Amita	63	Singh, Devraj	1, 19, 44
Godhani, Dinesh R.	79	Singh, Dhananjay	1
Jadhav, K.M.	27	Singh, Gaurav	9
Kale, Swapnil A.	94	Singh, Shakti Pratap	9, 69
Kandpal, Charu	19	Singh, Vinod Kumar	19
Khan, Aftab	1	Tripathi, Sudhanshu	44
Khangar, S.V.	94	Tripathy, Amita	55
Kumar, Vijay	36	Verma, Alok Kumar	9, 69
Mathana Gopal, A.	90	Yadav, Chadreshvar Prasad	1
Mehta, Jignasu P.	79	Yadav, Kanchan	86
Moses Ezhil Raj, A.	90	Yadav, Navneet	9
Mishra, S.P.	51	Yadav, Raja Ram	9, 69
Mulani, Vishal B.	79	Yadava, S.S.	86
Nalle, Pallavi B.	27	Yadawa, P.K.	69
Nath, G.	51		

Journal of Pure and Applied Ultrasonics

(INDEXED IN: Indian Citation Index, Google Scholar, i-Scholar, UGC List)

INFORMATION FOR AUTHORS

1. Type of Contribution

JOURNAL OF PURE AND APPLIED ULTRASONICS welcomes contributions on all aspects of ultrasonics including ultrasonic studies in medical ultrasonics, NDT, underwater, transducers, materials & devices and any other related topic. Contributions should fall into one of the following classes.

Paper - These should be on original research work contributing to scientific developments. They should be written with a wide readership in mind and should emphasize the significance of the work.

Reviews and Articles - Includes critical reviews and survey articles.

Research and Technical notes - These should be short descriptions of new techniques, applications, instruments and components.

Letters to the editor - Letters will be published on points arising out of published articles and papers and on questions of opinion.

Miscellaneous - Miscellaneous contributions such as studies, interpretive and tutorial articles, conference reports and news items are also accepted. Recommended contribution lengths are: Papers 2000-4000 words. Reviews and Surveys 2000-5000 words; Conference Reports 500-1500 words; News Items, Research and Technical Notes up to 1000 words.

2. Manuscripts

Manuscripts should be typed on one side of the paper in double spacing with 25 mm margin on all sides of A4 size paper. A soft copy of the manuscript in MS

WORD for text and MS EXCEL for illustrations and a PDF file thereof may be sent by e-mail or CD/DVD. Colour images should be formatted as JPEG files. Figures submitted in colour would be published in colour. Colour should be avoided unless it is required in order to convey a message or serve a purpose in the image.

Title - Titles should be short and indicate the nature of the contribution.

Abstract - An abstract of 100-200 words should be provided on the title page of paper and review article. This should indicate the full scope of the contribution and include the principal conclusions.

Mathematics - Mathematical expressions should be arranged to occupy the minimum number of lines consistent with clarity e.g., $(x^2+y^2)/(x-y)^{1/2}$.

Illustration - The line illustrations along with captions should be clearly drawn with black Indian ink. Figures in Excel are preferred.

References - References should be referred to in the text by number only. The reference number should be given as superscript. The corresponding reference shall contain the following information in order; names and initials of author (s)(bold), title of the work, journal or book title (italic), volume number (bold), year of publication in brackets, page number, e.g., **Kumar S.** and **Furuhashi H.**, Anisotropic divergence controlled ultrasonic transmitter array for three dimensional range imaging, *J. Pure Appl. Ultrason.*, **38** (2016) 49-57.

Units and Abbreviations - Authors should use SI units wherever possible.