



Studies of Thermo Acoustical Properties of Polyoxyethylene (10) oleyl ether in Presence of Polyvinylpyrrolidone at Different Temperatures by Ultrasound Measurement

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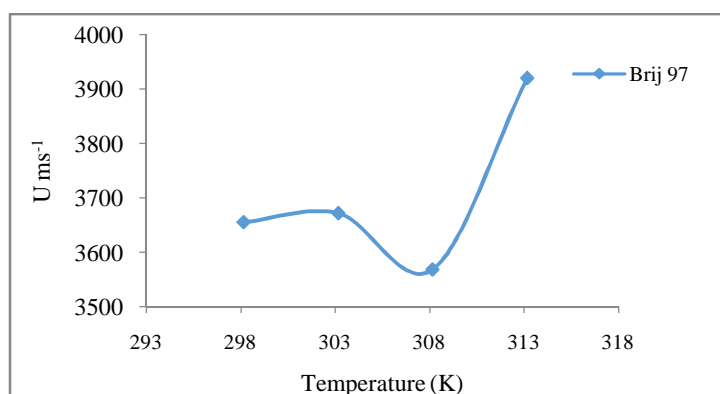
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ABSTRACT

Here we have investigated the effect of Polyvinylpyrrolidone (PVP) on thermo acoustical properties of Polyoxyethylene(10)oleyl ether (Brij-97) by ultrasound velocity measurement at different temperatures. In this paper, we examine the density (ρ), ultrasonic velocity (U), adiabatic compressibility (β_{ad}), molar volume (V_m), intermolecular free length (L_f), acoustic impedance (Z) and surface tension (γ) of aqueous solution of Brij-97 (0.029%) and in the presence of 0.005-0.05% w/v PVP at different temperatures (298.15, 303.15, 308.15 and 313.15K) and atmospheric pressure. The ultrasonic velocity, acoustic impedance and surface tension were increased up to 0.02% w/v PVP and decreases with further increase in concentration of PVP, while the adiabatic compressibility and intermolecular free length is found to be decreased with increase in the concentration PVP up to 0.02% w/v and then increases with further increase in concentration of PVP for all the temperatures studied

Graphical Abstract



Ultrasonic velocities of 0.029% w/v Brij-97 at different temperatures.

Keywords: Nonionic surfactant, Polymer, Acoustical properties, Ultrasound velocity.

INTRODUCTION

Surfactants are an important component of pharmaceutical and chemical industry. Their level of development has been regarded as an important indication of high-tech chemical technology industry all over the countries. Surfactant plays a major role in the formulation, paints, detergents, food polymer synthesis and formulations of drug and pesticides [1, 2]. Surfactant, a surface active agent is amphiphilic in nature, because surfactant molecule possesses both a hydrophilic and a hydrophobic portion. Many researchers have studied the various properties of aqueous solution of surfactant and with polymer in order to know the type of interaction within surfactant micelle and between added polymer [3-7]. When added into water, surfactant molecules orient at the surface so that their hydrophobic regions are isolated from the aqueous phase. Polyoxyethylene type nonionic surfactants become soluble in water by the hydration of the ether oxygen of the polyoxyethylene groups. As temperature increases the number of hydrogen bonds decreases between water molecule and surfactant hydrophilic group, which increases the micellar mass and thus CMC decreases. If the temperature continues to increase, the micelle becomes so large and the number of inter micellar interactions increase to such an extent that a sudden turbidity in the solution is observed, this temperature is called cloud point temperature. As temperature increases further this causes the solution to separate in to two phases, where one is surfactant-rich phase and the other is surfactant poor phase. Polymer interacts with surfactants in solution forming micellar complex having characteristic physicochemical features [8, 9].

The variation of ultrasonic velocity with temperature indicates the formation of complex between polymer and surfactant molecules through hydrogen bonding [10, 11]. Surfactant molecules form micelles in aqueous solutions when the concentration exceeds a certain value, this concentration is called the critical micelle concentration (CMC). The ultrasound velocity data and other acoustical parameters give the important information about the polymer-surfactant interactions in aqueous solution. Water soluble polymers are interacting with surfactant to varying degrees depending on the nature and properties of the polymer and surfactant [12, 13]. As the temperature increases, there is breaking of the bonds between the associated molecules to form separate monomers, also increase of thermal energy weakens the inter molecular forces which results in to decrease of ultrasonic velocity [14, 15]. Intermolecular free length shows similar behavior as observed in adiabatic compressibility. The decreases in adiabatic compressibility leads the molecules to be closer resulting into a decrease of inter molecular free length. Intermolecular free length is a major factor in determining the variation of ultrasonic velocity of solutions. As ultrasonic velocity increases, inter molecular free length decreases, and vice versa. This indicates the inverse behavior in these two properties. The model for sound propagation explains the interdependence of intermolecular free length and the ultrasonic velocity [16]. The decrease in the values of adiabatic compressibility and intermolecular free length with increase in ultrasonic velocity shows that there is a significant interaction between the surfactant molecule and added polymer, due to which structural arrangement is considerably affected [17]. Acoustic impedance depends on the temperature and concentration of the solutions. The increase of acoustic impedance shows that there is strong interaction between the surfactant and polymer in aqueous solutions. The result obtained is confirmed by the decrease of intermolecular free length with concentration [18, 19].

MATERIALS AND METHODS

Experimental: The A.R. grade nonionic surfactant Polyoxyethylene-10-lyl ether (Brij-97) (Mole. Wt. 709 g mol⁻¹) was the products of Sigma Aldrich, USA (purity>99%) and uncharged polymer Polyvinylpyrrolidone (Mole. Wt. 4000 g mol⁻¹) (PVP) was the product of S. D. Fine Chemical Ltd. and are used as received. Doubly distilled deionized water with specific conductance 2-4 $\mu\text{s cm}^{-1}$ at 303.15 K is used for the preparation of solutions. 0.029% w/v aqueous solution of Brij-97 was prepared on using distilled water as solvent. The PVP solutions of 0.005 to 0.05 % w/v were prepared in 0.029% w/v Brij-97, which is critical micellar concentration (CMC) of Brij-97. The ultrasonic

velocities (U) of surfactant and polymer-surfactant mixtures were measured at 2 MHz using ultrasonic interferometer F-81 (Mittal Enterprises New Delhi). This instrument is connected to fully automatic microprocessor controlled software based instrument having provision to record readings on digital panel and in computer. The test solution in interferometric cell was maintained at required temperature by circulating thermostatic water with an accuracy of $\pm 0.1\text{K}$. The estimated accuracy of sound velocity was $\pm 0.2\%$. The reliability and accuracy of the measurements was checked by obtaining sound velocity data of water and carbon tetrachloride at 298.15K. The ultrasound velocity data were found to be reproducible with in $\pm 0.5\text{ ms}^{-1}$ [20].

Densities: Densities (ρ) of the solutions have been determined using bicapillary pycnometer (20 cm^3) as described earlier [21, 22]. The pycnometers are filled with experimental liquids and are kept in a water bath. The position of liquid levels in the two arms was noted which could read to 0.01mm. The accuracy of density measurement was $\pm 0.001\text{Kg/m}^3$. Three to four measurements were made with average deviation of 0.00005 g cm^{-3} . All the measurements were carried out such as to avoid various types of possible degradation of the polymer solution. The speed of sound as well as densities of aqueous solutions of studied surfactant and polymer-surfactant was simultaneously measured at the temperatures 298.15, 303.15, 308.15 and 313.15K.

RESULTS AND DISCUSSION

The various thermo acoustical parameters are obtained from measured values of ultrasonic velocity (U) and density (ρ) using the standard formulae,

- | | |
|----------------------------------|---|
| (i) Adiabatic compressibility | $\beta = 1/U^2 \rho$ |
| (ii) Inter molecular free length | $L_f = k \beta^{1/2}$ |
| (iii) Acoustic impedance | $Z = \rho U$ |
| (iv) Molar sound velocity | $R_M = (M/\rho) U^{1/3}$ |
| (v) Molar volume | $V_m = M/\rho$ |
| (vi) Surface tension | $\gamma = (U^{3/2}) (6.3 \times 10^{-4}) \times \rho$ |

where U is ultrasound velocity, ρ is density, β is Adiabatic compressibility, L_f Inter molecular free length, k is Jacobson's temperature dependent constant $[(93.875 + 0.375T)10^{-8}]$, Specific acoustic impedance (Z), Molar sound velocity (R_m), Molar volume (V_m) Surface tension (γ) and M is the effective molecular weight which can calculated using the relation,

$$M = X_1 M_1 + X_2 M_2$$

In above relation M_1 and M_2 are the molecular weights, X_1 and X_2 are the mole fractions of component-1 additive and component-2 surfactant solution as solvent [5, 19].

Ultrasonic velocity of Brij-97: In the present investigation, the concentration of Brij-97 is taken to be 0.029 % w/v which is CMC of the surfactant. The ultrasonic velocity measurement was made at temperatures 298.15, 303.15, 308.15 and 313.15K. The thermo acoustical parameters of Brij-97(0.029 % w/v) at various temperatures are shown in table 1. The ultrasonic velocity and surface tension of the pure surfactant solution increases with temperature and decreases at 303.8K to obtain minima and then increases appreciably at 313.15 K (Figure 1).

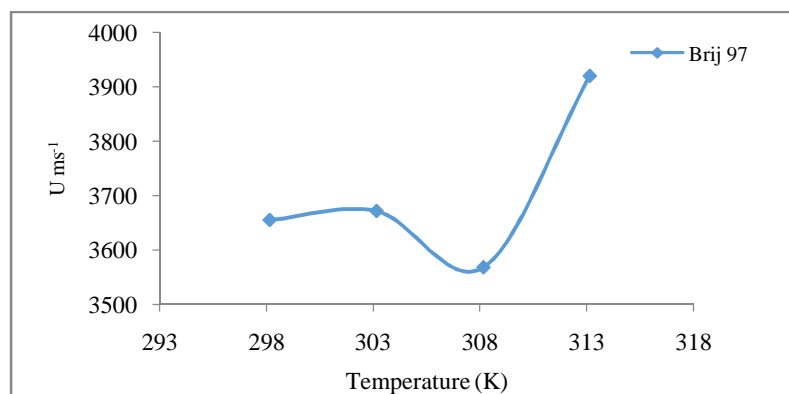


Figure 1. Ultrasonic velocities of 0.029% w/v Brij-97 at different temperatures.

For pure surfactant system ultrasound velocity initially decreases as temperature increases, attains minima and on further increases in temperature, the ultrasound velocity suddenly increases, this due the fact that as the temperature increases available thermal energy facilitates the breakings of the bonds between the associated molecules in to their monomer and weakens the molecular forces which tends to decrease the ultrasound velocity.

Table 1. Ultrasound velocity and other acoustic parameters of Brij-97 (0.029 %) at different temperatures

Temp K	Density (ρ) kg m^{-3}	Ultrasonic Velocity (U) ms^{-1}	Adiabatic Compressibility (β_{ad}) $\times 10^{-10} \text{ kg}^{-1} \text{ ms}^2$	Intermolecular Free length (L_f) \AA	Acoustic Impedance (Z) $\times 10^6 \text{ kg m}^{-2} \text{ s}^{-1}$	Molar Volume (V_m) $\times 10^{-3} \text{ L.mol}^{-1}$	Molar Sound Velocity (R_M) $\times 10^{-4} \text{ mmol}^{-1} (\text{N/m}^{1/2})^{-1/3}$	Surface Tension (γ) $\times 10^4 \text{ Nm}^{-1}$
298.15	1012.1390	3656.00	0.7392	0.1690	3.7004	126.81855	1953.67	14.096
303.15	1011.8060	3672.00	0.7330	0.1699	3.7154	126.86029	1957.16	14.184
308.15	1011.4020	3568.00	0.7767	0.1766	3.6087	126.91096	1939.28	13.580
313.15	1010.4810	3920.00	0.6440	0.1623	3.9611	127.02663	2002.89	15.624

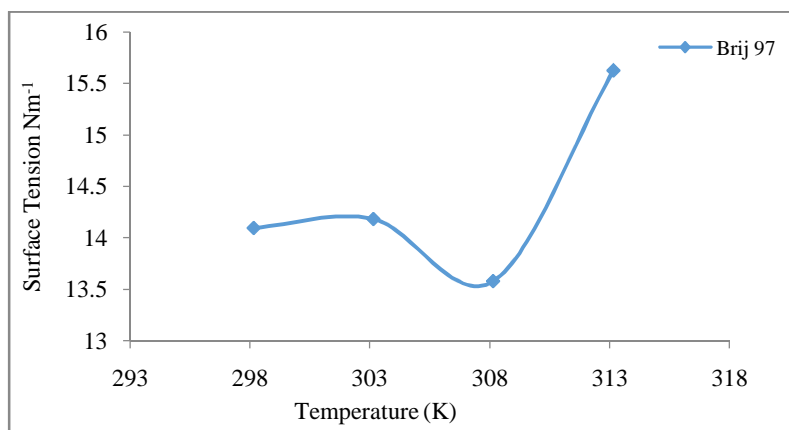


Figure 3. Surface tension of 0.029% w/v Brij-97 at different temperatures.

Ultrasonic velocity of Brij-97-PVP systems: The effect of PVP concentration on ultrasonic velocity and other acoustical parameters of Brij-97 at various temperatures is shown in table 2 and 3.

The ultrasonic velocity measurement was made at different temperatures 298.15, 303.15, 308.15 and 313.15K. The ultrasonic velocity and density of 0.029% Brij-97 at CMC value in the presence of PVP having different concentrations and temperatures have been reported in table 2 and 3. The various acoustic parameters such as adiabatic compressibility (β), inter molecular free length (L_f),

Table 2. Ultrasonic velocity and other acoustic parameters for 0.029 % Brij-97 + PVP at 298.15 and 303.15K

Conc. of PVP % (w/v)	Density (ρ) kg m^{-3}	Ultrasonic Velocity (U) ms^{-1}	Adiabatic Compressibility (β_{ad}) $\times 10^{-10}$ $\text{kg}^{-1} \text{ms}^2$	Intermolecular Free length (L_f) A°	Acoustic Impedance (Z) $\times 10^6$ $\text{kg m}^{-2}\text{s}^{-1}$	Molar Volume (V_m) $\times 10^{-3}$ L.mol^{-1}	Molar Sound Velocity (R_M) $\times 10^{-4}$ $\text{mmol}^{-1} (\text{N/m}^{1/2})^{-1/3}$	Surface Tension (γ) $\times 10^4$ Nm^{-1}
298.15K								
0.005	1005.1090	2832.00	1.2405	0.2190	2.8465	127.70555	1806.79	9.543
0.01	1016.5040	3384.00	0.8591	0.1822	3.4398	156.46569	2349.07	12.607
0.02	1017.1080	5200.00	0.3636	0.1186	5.2890	186.54656	3231.88	24.028
0.03	1071.3400	3792.00	0.6491	0.1584	4.0625	234.29618	3655.16	15.761
0.04	1020.0360	3440.00	0.8285	0.1790	3.5089	306.35978	4624.71	12.966
0.05	1021.0570	3104.00	1.0165	0.9182	3.1694	366.16761	5341.38	11.124
303.15 K								
0.005	1004.9840	3056.00	1.0655	0.2049	3.0712	127.72144	1853.45	10.696
0.01	1015.7630	3488.00	0.8092	0.1786	3.5430	156.57983	2374.62	13.182
0.02	1015.9960	5764.00	0.2963	0.1080	5.8562	186.75074	3348.40	28.010
0.03	1071.1550	3704.00	0.6805	0.1637	3.9676	234.43666	3627.29	15.212
0.04	1019.6610	3520.00	0.7915	0.1766	3.5892	306.47245	4662.00	13.416
0.05	1020.6850	3280.00	0.9107	0.1894	3.3478	366.30106	5442.47	12.079

Table 3. Ultrasonic velocity and other acoustic parameters for 0.029 % Brij-97 + PVP at 308.15 and 313.15K

Conc. of PVP % (w/v)	Density (ρ) kg m^{-3}	Ultrasonic Velocity (U) ms^{-1}	Adiabatic Compressibility (β_{ad}) $\times 10^{-10}$ $\text{kg}^{-1} \text{ms}^2$	Intermolecular Free length (L_f) A°	Acoustic Impedance (Z) $\times 10^6$ $\text{kg m}^{-2}\text{s}^{-1}$	Molar Volume (V_m) $\times 10^{-3}$ L.mol^{-1}	Molar Sound Velocity (R_M) $\times 10^{-4}$ $\text{mmol}^{-1} (\text{N/m}^{1/2})^{-1/3}$	Surface Tension (γ) $\times 10^4$ Nm^{-1}
308.15 K								
0.005	1004.2530	4040.00	0.6101	0.1565	4.0572	127.81441	2035.67	16.246
0.01	1014.8390	4480.00	0.4910	0.1404	4.5465	156.72240	2583.59	19.171
0.02	1014.7020	5928.00	0.2804	0.1061	6.0152	186.98889	3384.17	29.177
0.03	1016.4140	4520.00	0.4816	0.1390	4.5942	247.06271	4084.95	19.459
0.04	1019.9410	5072.00	0.3811	0.1237	5.1731	306.38831	5264.19	23.210
0.05	1019.2950	4080.00	0.5894	0.1538	4.1587	366.80058	5861.16	16.735
313.15 K								
0.005	1003.1530	3448.00	0.8385	0.1852	3.4589	127.95456	1933.06	12.796
0.01	1013.9160	3928.00	0.6392	0.1617	3.9827	156.86507	2475.05	15.725
0.02	1013.9650	5104.00	0.3786	0.1244	5.1753	187.12480	3221.82	23.293
0.03	1015.8590	3912.00	0.6432	0.1622	3.9740	247.19769	3895.03	15.659
0.04	1019.0130	3712.00	0.7122	0.1707	3.7826	306.66733	4748.29	14.519
0.05	1018.5490	3376.00	0.8614	0.1877	3.4386	367.06923	5506.58	12.587

The ultrasonic velocity measurement was made at different temperatures 298.15, 303.15, 308.15 and 313.15K. The ultrasonic velocity and density of Brij-97 at CMC value (0.029%) value in the presence of PVP having different concentrations and temperatures have been reported in table 2 and 3. The various acoustic parameters such as adiabatic compressibility (β), inter molecular free length (L_f), specific acoustic impedance (Z), molar sound velocity (R_m), molar volume (V_m) and surface tension (γ) are presented in table 2 and 3. The sound speed increases with increase in temperature where as density decreases this may be due the temperature induced conformational changes in polymer-surfactant. From the above table 2 and 3 it is clear that for a given concentration of nonionic surfactant (0.029%) and additive PVP, the ultrasonic velocity, acoustic impedance, molar volume, molar sound velocity and surface tension increases with increase in temperature, whereas adiabatic compressibility (β) and intermolecular free length (L_f) decreases with increase in temperature. This is due to decreases in density with increases in temperature of Brij-97-PVP solution [24, 25]. The plot of ultrasound velocity vs. concentration of PVP at various temperatures is shown in (Figure 4), which shows that the maximum interaction exists at 308.15K. Initially the ultrasound velocity increases as temperature increases this is due to when the temperature increases from 303.15K to 308.15K the

ultrasound velocity increases and attains maxima which indicates the strong interaction between the surfactant and polymer.

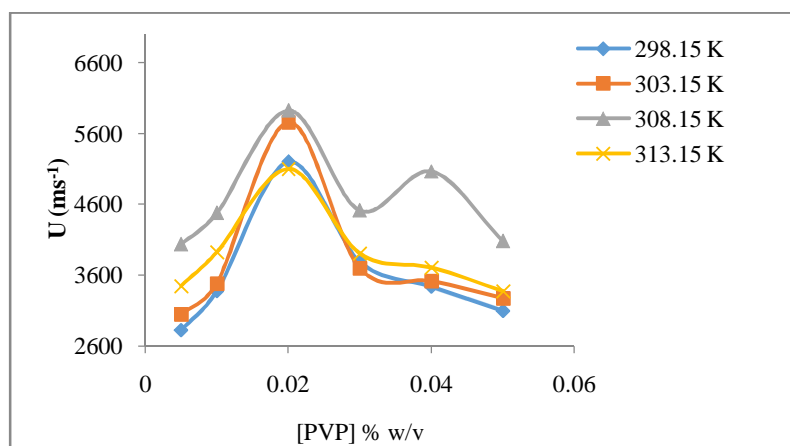


Figure 4. Ultrasonic velocities 0.029% w/v Brij-97 in presence of PVP at various temperatures and concentrations.

The ultrasonic velocity, acoustic impedance and surface tension were increases up to 0.02% w/v PVP and then decreases with further increase in concentration of PVP, while the adiabatic compressibility and intermolecular free length were decreased with increase in the concentration PVP up to 0.02% w/v (Figure 5) and then increases with further increase in concentration of PVP for all the temperatures studied. The decrease in adiabatic compressibility with increase of concentration of PVP indicates the formation of a large number of closely bound systems this may be due to a more rigid liquid structure associated with hydrogen bonding of PVP with surfactant. Such reduction in compressibility has been found in the solution due to solvent molecules [26].

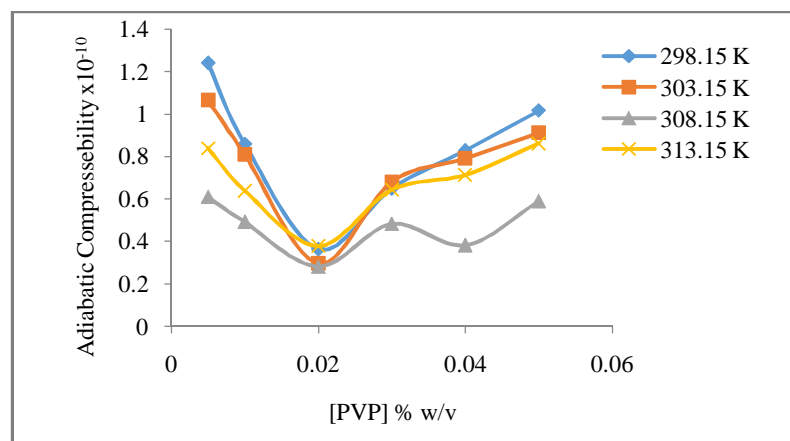


Figure 5. Adiabatic compressibility of 0.029% w/v Brij-97 in presence of PVP at various temperatures and concentrations.

The molar volume and molar sound velocity were increases with concentration of PVP for all the temperatures studied. It has been observed that as concentration of PVP increases surface tension increase up to 0.02% and then after it decreases (Figure 6).

Also as temperature increases surface tension increases up to 308.15 K and then decreases up to 313.15K. The variation of ultrasonic velocity with temperature indicates the formation of complex and strong interaction between polymer and surfactant. Also it is noted that with increase in temperature,

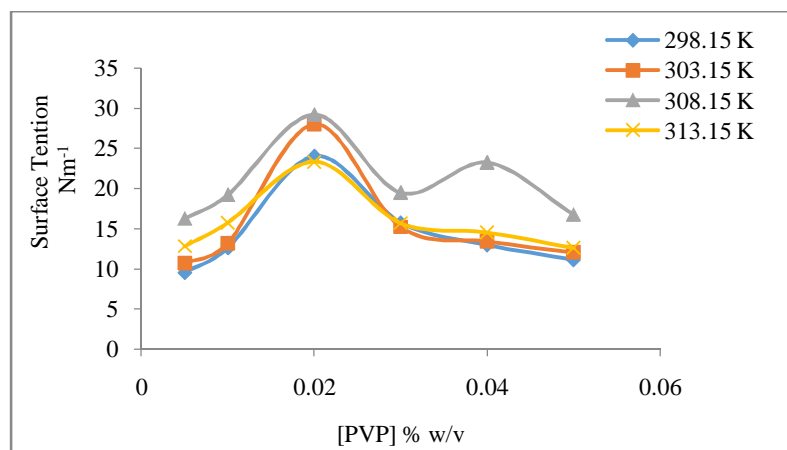


Figure 6. Surface tension of 0.029% w/v Brij-97 in presence of PVP at various temperatures and concentrations.

the ultrasonic velocity decreases, because available thermal energy breaks the bonds between the associated molecules to form their monomers. Increase of temperature weakens the intermolecular forces which decreases the ultrasound velocity. Intermolecular free length (L_f) shows almost similar behavior as shown by adiabatic compressibility (β). Intermolecular free length is a major factor in determining the variation of ultrasonic velocity in solutions. When ultrasonic velocity increases, intermolecular free length decreases or vice versa. The decreased compressibility leads the molecules to a closer packing and thus decreases the intermolecular free length (L_f). The model for sound propagation explains the dependence of intermolecular free length on ultrasonic velocity [16, 19]. The decrease in value of adiabatic compressibility and intermolecular free length with increase in ultrasonic velocity indicates that there is a significant interaction between the surfactant and polymer molecule due to which structural arrangement is greatly affected [17].

APPLICATION

Ultrasonic techniques are being employed to gain some insight into the molecular behavior of solutions and liquid mixtures. Studies on sound velocities of surfactants solutions and surfactant-polymer mixed solutions are of great use in characterizing the structure and properties of solutions. Compressibility is an important thermodynamic parameter to know the behavior of a solute in a solvent.

CONCLUSION

The ultrasonic velocity data and acoustical parameters, adiabatic compressibility (β), inter molecular free length (L_f), specific acoustic impedance (Z), molar sound velocity (R_m), molar volume (V_m) and surface tension (γ) give important information to understand the polymer-surfactant interactions in aqueous solutions. The trends in acoustical parameters of Brij-97-PVP system suggest the strong interaction between surfactant and polymer. This may be due to the aggregation of surfactant micelles on the polymer chain. Further the variation in acoustical parameters of surfactant alone might be due to the formation of micelle rods in the solution.

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