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Ultrasonic study on some monosaccharides in aqueous media at 298.15 K

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Abstract The ultrasonic velocity (U), density (ρ) and viscosity (η) at 298.15 K have been measured in the binary systems of monosaccharides (glucose, fructose and galactose) in aqueous medium. The acoustical parameters such as adiabatic compressibility (β), free length (L_f), free volume (V_f), internal pressure (π_i), acoustical impedance (Z), relative association (R_A), molar sound velocity (R) and molar sound compressibility (W) are calculated. The results are interpreted in terms of molecular interaction between the components of the mixtures.

1. Introduction

Ultrasonic velocity of a liquid is fundamentally related to binding forces between the atoms or molecules. Langemann and Dunbar (1945) have qualitatively deduced the degree of molecular association in liquids by the use of sound velocity data. In recent years, the measurements of ultrasonic velocity have been adequately employed in understanding the nature of molecular interaction in pure liquids, binary and ternary mixtures (Srinivasalu and Ramachandra Naidu, 1991; Varadarajalu and Lakshminarayanan Reddy, 1998; Aswar, 1998). However, the derived parameters from sound velocity such as adiabatic compressibility and free volume provide a better insight into the molecular environment of liquids (Hermers et al., 1988; Prakash et al., 1988; Blokhra and Nag, 1991). The inactivation of glucose-6-phosphate dehydrogenase in solution by low and high frequency ultrasound, and the observed antigenicity of alkali-treated protein, was compared with that of serum-blocking antigen prepared (Rachinskaya et al., 2004). The ultrasonic measurements of sucrose in binary solvent mixture of water and acetonitrile at various temperature from 28 °C to 45 °C and some measured parameters such as acoustical impedance, adiabatic compressibility, apparent molal compressibility, relative association, inter molecular free length, molar sound velocity, and Wada's number (Syal et al., 1998).

Most of the researchers in ultrasonic studies area were found to focus their view on organic systems and a detailed

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survey of the literature shows that the investigations on carbohydrate and enzyme systems are scarcely reported. The present work mainly deals about the aqueous mixture of monosaccharides (glucose, fructose and galactose), mostly of the same family and having many common properties with a sharp characteristic difference, which are the most significant biomolecules of life on the earth.

The carbohydrates, often termed as sugars, are the 'staff of life' for most organisms. They are widely distributed in both plant and animal tissues. They are indispensable for living organisms, serving as skeletal structures in plants and also in insects and crustaceans. They also occur as food reserves in the storage organs of plants and in the liver and muscles of animals. In addition, they are an important source of energy required for the various metabolic activities of the living organisms, the energy being derived as a result of their oxidation. They also serve to lubricate skeletal joints, to provide adhesion between cells and to confer biological specificity on the surface of animal cells. Plants are considerably richer in carbohydrates in comparison to animals. To accommodate a wide variety of compounds, the carbohydrates are nowadays broadly defined as polyhydroxy aldehydes or ketones and their derivatives or as substances that yield one of these compounds on hydrolysis.

2. Experimental

The monosaccharides (AnalaR Grade) supplied by S.D. Fine Chem. have been taken in the forms of solutions. Double distilled water is used throughout the work. Molar solutions of monosaccharides, from 0.01 m to 0.10 m, in steps of 0.01 m were prepared by weight. All the solutions are left for 2 h and complete solubility is found. The ultrasonic velocity in the prepared solutions was measured using ultrasonic interferometer (Mittal – 82), a variable path-fixed frequency type (2 MHz). The accuracy of sound velocity is ± 0.1 m s^{-1}. The density and viscosity of the mixture were measured using a specific gravity bottle (5 ml) and an Ostwald's viscometer (10 ml), respectively. The accuracy of density measurements is ± 0.0001 kg m^{-3} and that of viscosity measurement is ± 0.001 mN s m^{-2}.

Using the measured data, the acoustical parameters such as adiabatic compressibility (β), free length (L_f), free volume (V_f), internal pressure (π_i), acoustical impedance (Z), relative association (R), molar sound velocity (R) and molar sound compressibility (W) have been calculated from Nithiyanantham and Palaniappan (2008, 2009).

3. Results and discussion

Table 1 lists the observed values for the three aqueous monosaccharides viz. glucose, fructose and galactose. In general, carbohydrates are the compounds in which the H:O ratio is maintained as 2:1 as shown in $C_n(H_2O)_n$ and are neutral polyhydroxy alcohols, containing active aldehyde or ketone groups (Nithiyanantham and Palaniappan, 2008). They are all reducing sugars or they liberate reducing sugars on hydrolysis.

All the monosaccharides taken up for the present study are hexoses having the same molecular formula. However, glucose and galactoses are aldohexoses, having an aldehyde group (–CHO) as the reducing group whereas fructose belongs to ketohexoses, having ketone as the functional reducing group.

The perusal of Table 1 clearly reveals that the measured parameters viz. sound velocity, density and viscosity increase with increase in molality of monosaccharides. As molality (m) increases, the number of molecules in the medium increases, making the medium denser, this leads to lesser compressibility and hence sound velocity increases. Further, the increase in the number of particles simply increases the frictional resistance between the layers of the medium and that tends to increase the coefficient of viscosity. Moreover, the existing particle–particle frictional resistance expects some interactions and this expectation is further supported by the non-linear increasing trend of the measured parameters.

A keen look at these tables suggests that the range of sound velocity and their variation with molality is almost the same for all the monosaccharides taken up for the present study. However, in general glucose shows a minimum sound velocity, whereas for fructose it is maximum. But considering the density variations, the maximum densities are recorded for galactose whereas glucose remains minimum. No such general trend is obtained from viscosity variation as it is highly fluctuating and seems to be particle sensitive.

All the three monosaccharides are existing in two different forms as (i) a straight chain compound and (ii) a cyclic compound. Further, the cyclic form may be of two types, furanose and pyranose, of which the pyranose form is more stable in solution (Nithiyanantham and Palaniappan, 2009). These furanose and pyranose are further classified as α- form and β-form depending on the position of group at carbon 1 atom. For glucose, all these forms viz. a straight chain aldehyde form and all types of cyclic forms are existing while the β-pyranose form predominates with mutarotation of +59.5°. This form is

Table 1 Measured values of ultrasonic velocity (U), density (ρ) and viscosity (η) of aqueous monosaccharides (glucose, fructose, galactose) at 298.15 K.

S. No.	Molality (m)	U (m s^{-1})			ρ (kg m^{-3})			$\eta \times 10^3$ (N s m^{-2})		
		Glucose	Fructose	Galactose	Glucose	Fructose	Galactose	Glucose	Fructose	Galactose
1	0.01	1502.8	1503.4	1503.3	998.2	1000.4	1001.0	0.9013	0.9159	0.9056
2	0.02	1504.6	1505.1	1504.9	998.8	1001.0	1001.6	0.9098	0.9242	0.9137
3	0.03	1506.5	1507.2	1506.7	999.6	1001.8	1002.3	0.9169	0.9340	0.9203
4	0.04	1508.7	1509.4	1508.5	1000.4	1002.6	1002.9	0.9282	0.9442	0.9272
5	0.05	1510.3	1511.3	1510.3	1001.0	1003.4	1003.5	0.9357	0.9494	0.9354
6	0.06	1512.1	1513.2	1512.0	1001.6	1004.1	1004.2	0.9534	0.9550	0.9422
7	0.07	1513.2	1514.9	1513.8	1002.0	1004.7	1004.8	0.9634	0.9632	0.9509
8	0.08	1515.0	1516.2	1515.5	1002.7	1005.2	1005.6	0.9728	0.9684	0.9592
9	0.09	1516.8	1518.1	1517.0	1003.3	1005.9	1006.2	0.9826	0.9772	0.9675
10	0.10	1518.2	1520.4	1518.6	1003.9	1006.7	1006.9	0.9874	0.9838	0.9761

a six-membered ring with less compactness. This may be attributed for the observed low sound velocity. In the case of galactose, the mutarotation is $+80.2°$ that indicates the conversion of α- to β-forms and vice versa via the straight chain. Aldo form is enormously higher than glucose, with a net pyranose structure. However, in the case of fructose, the rotation is levorotatory (92.4°) and the final structure is of furanose form (Nithiyanantham and Palaniappan, 2009), that too the active group is a keto group. All these structural variations are reflected in the observed trends of sound velocity.

It is further noticed that for any given molality of monosaccharides the sound velocity is always least for glucose and highest for fructose whereas the density values are maximum for galactose. It is interesting to note the specific variations of viscosity with molality. The observed viscosity values are minimum for glucose and maximum for fructose up to 0.03 m of monosaccharides, but in the range of 0.04–0.06 m, the least values are shifted to galactose and in the higher molarities, glucose records the maximum viscosity values. The coefficient of viscosity is a chief peculiar property that determines the inner nature of the medium. In one sense, the viscosity variations can offer much more information about the medium that is obtained from velocity or density variations. The variations of viscosity are the consequences of variations in the surface area of molecules, their relative velocity and their inner closeness, i.e., the structure of the molecule, and all these play a key role in deciding this parameter.

As regards glucose, at lower molality, the observed viscosity value suggests that the furanose form is predominating the other forms. As it is fivemembered, that too ring, it will occupy a relatively smaller area than the pyranose ring or the straight chain. The smaller area leads to small frictional forces and thus the coefficient of viscosity is low at lower molality. However, as molality increases, the number of molecules increases and so, the clustering of furanose becomes pyranose via the straight chain form that has maximum surface area and so the viscosity gradually shifts to the maximum limit with the molality of glucose. It should be remembered that the pyranose forms are more stable than the furanose form in solution (Nithiyanantham and Palaniappan, 2009). Considering the case of galactose, as there is no furanose form, the coefficient of viscosity initially shows an appreciable value but the deviations remain in a smaller range. But for fructose, irrespective of the geometry and mutarotation, the active group (ketone group) compresses the molecular size and so whatever be the

structure (straight chain or cyclic) the molecular area is comparatively less and hence the coefficient of viscosity shifts from maximum extreme to minimum side.

To explore the nature and type of existing interactions some thermoacoustical parameters have been determined and their extremes and trends are analyzed in the light of the existing structural variations. Compressibility is the ease with which a medium can be compressed. If the compressibility values are higher, it implies that the medium is loosely packed whereas the lower compressibility is an indication of maximum interaction. The gradual decrease in adiabatic compressibility with respect to increase in molality suggests that the medium becomes more and less compressible.

The calculated values of the chosen thermoacoustical properties are presented in Tables 2 and 3. Referring Table 2, it is observed that the trend shown by adiabatic compressibility (β) is a general decrease with increasing molality for all the monosaccharides. However, for glucose, the highest (β) values are recorded but the higher values are existing in fructose up to 0.02 m and then shifted to galactose in the rest of molalities.

The intermolecular free length (L_f) is again a predominant factor in determining the existing interactions among the components of the mixture. Analyzing the respective tables, it is noticed that the L_f reflects a similar trend as that of β. In each of these solutions, the medium is having only one solute, whose structure depends on the number of C atoms and the type of active group. Carbohydrates possess active groups, which are responsible for their chemical behavior and physical interaction. These groups are glycosidic –OH, alcoholic –OH and –CHO (or –CO). The property of galactose is that the equilibrium existing among the different forms is shifted in favor of the group which interacts. The aldehyde group observed in it and the alcohol group of five or four approaches each other to hemiacetals. This leads to compactness of various groups and hence a decrease in (β), is expected.

Moreover, glucose and fructose are interconvertible in weak alkaline solutions (Nithiyanantham and Palaniappan, 2009). Water being an amphiprotic solvent, can selectively act as a base and hence either the chain transformation or the active group transformation is suspected here.

As regards glucose solution the β and hence L_f values are initially maximum that conveys more available free space between the molecules than compared with other monosaccharides. It may be taken as a consequence of lesser molecular size. As glucose can exist in three different forms, lesser

Table 2 Calculated values adiabatic compressibility (β), intermolecular free length (L_f), free volume (V_f) and internal pressure (π_i) of aqueous monosaccharide (glucose, fructose, galactose) at 298.15 K.

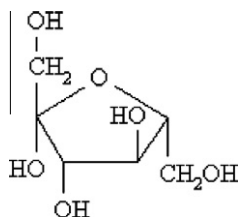| S. No.	Molality (m)	$\beta \times 10^{-10}$ (N^{-1} m^2)			$L_f \times 10^{-11}$ (M)			$V_f \times 10^8$ (m^3 mol^{-1})			$\pi_i \times 10^{-6}$ (N m^{-2})		
		Glucose	Fructose	Galactose	Glucose	Fructose	Galactose	Glucose	Fructose	Galactose	Glucose	Fructose	Galactose
1	0.01	4.4358	4.4224	4.4203	4.2024	4.1960	4.1950	1.8614	1.8182	1.8492	2716	2742	2727
2	0.02	4.4225	4.4098	4.4084	4.1961	4.1900	4.1893	1.8433	1.8012	1.8320	2723	2748	2733
3	0.03	4.4078	4.3939	4.3946	4.1891	4.1825	4.1828	1.8298	1.7811	1.8198	2728	2757	2733
4	0.04	4.3914	4.3776	4.3815	4.1813	4.1747	4.1766	1.8048	1.7602	1.8072	2739	2766	2743
5	0.05	4.3794	4.3635	4.3684	4.1756	4.1680	4.1703	1.7901	1.7531	1.7910	2745	2768	2749
6	0.06	4.3664	4.3495	4.3561	4.1694	4.1613	4.1644	1.7481	1.7454	1.7788	2765	2771	2754
7	0.07	4.3583	4.3371	4.3430	4.1655	4.1554	4.1582	1.7268	1.7300	1.7620	2774	2777	2761
8	0.08	4.3453	4.3272	4.3298	4.1593	4.1506	4.1518	1.7092	1.7226	1.7463	2781	2779	2767
9	0.09	4.3324	4.3135	4.3186	4.1531	4.1440	4.1465	1.6907	1.7066	1.7304	2790	2786	2774
10	0.10	4.3218	4.2972	4.3065	4.1480	4.1362	4.1407	1.6845	1.6974	1.7146	2792	2789	2780

Table 3 Calculated values acoustical impedance (Z), relative association (R_A), molar sound velocity (R) and molar sound compressibility (W) of aqueous monosaccharides (glucose, fructose, galactose) at 298.15 K.

S. No.	Molality (m)	$Z \times 10^6$ (kg m^{-2} s^{-1})			R_A			$R \times 10^{-3}$ (m$^{10/3}$ s$^{-1/3}$ mol^{-1})			W (m^3 mol^{-1} (N m^{-2})$^{1/7}$)		
		Glucose	Fructose	Galactose	Glucose	Fructose	Galactose	Glucose	Fructose	Galactose	Glucose	Fructose	Galactose
1	0.01	1.5001	1.5041	1.5049	1.0000	1.0019	1.0026	2.0688	2.0645	2.0632	3.9165	3.9095	3.9074
2	0.02	1.5028	1.5067	1.5074	1.0000	1.0022	1.0028	2.0718	2.0673	2.0660	3.9222	3.9150	3.9129
3	0.03	1.5059	1.5100	1.5102	1.0004	1.0025	1.0031	2.0743	2.0700	2.0687	3.9273	3.9202	3.9181
4	0.04	1.5094	1.5134	1.5130	1.0008	1.0028	1.0033	2.0770	2.0726	2.0716	3.9326	3.9255	3.9238
5	0.05	1.5119	1.5164	1.5157	1.0010	1.0031	1.0035	2.0798	2.0754	2.0745	3.9381	3.9308	3.9294
6	0.06	1.5146	1.5194	1.5183	1.0012	1.0034	1.0038	2.0828	2.0781	2.0774	3.9437	3.9361	3.9349
7	0.07	1.5163	1.5220	1.5210	1.0014	1.0036	1.0040	2.0858	2.0810	2.0803	3.9495	3.9417	3.9405
8	0.08	1.5190	1.5241	1.5240	1.0016	1.0038	1.0044	2.0887	2.0840	2.0827	3.9552	3.9473	3.9454
9	0.09	1.5218	1.5271	1.5264	1.0018	1.0041	1.0047	2.0916	2.0866	2.0854	3.9608	3.9526	3.9507
10	0.10	1.5241	1.5306	1.5291	1.0021	1.0044	1.0050	2.0944	2.0893	2.0880	3.9662	3.9579	3.9558

molecular size can be attributed only for five-membered structures, i.e., furanose form (Fig. 1). Further this is the predominating cyclic form of glucose (Nithiyanantham and Palaniappan, 2009). However, as molality of glucose increases, the β (and L_f) values are in a decreasing trend indicating that the medium is getting denser. This is possible if the structure of furanose glucose were changed to any one of the other two forms. As the straight chain form is not a predominating one, the only possibility is becoming a pyranose chain. Thus, the decreasing trend of β and L_f with increasing molality may be attributed to the fact that glucose molecules are getting changed from furanose form to pyranose form (Fig. 2).

The values of β and L_f for fructose and galactose are not as much for glucose. As regards galactose, it may be in straight chain form or in pyranose form. On comparing the β (and L_f) values observed for galactose and fructose with that of glucose, for any given molality, it can be suggested that the starting structure may not be a cyclic form as for glucose. However, the deviations of β (and L_f) from 1 m to other are almost same as for glucose. Hence, the form conversion may come into picture. But in galactose, the originally available straight chain form becomes a cyclic pyranose form with an increase in molality (Nithiyanantham and Palaniappan, 2009). In the case of fructose, β and L_f are higher than those observed for galactose till 0.02 m, which suggests that at lower molality, fructose may exist in chain form. But the values and trend in other molalities of fructose reveal nothing about the structure but it is true that the ketone group may have a key role to be explored.

The average available volume between the molecules of the mixture is referred to as the free volume (V_f) whereas the resultant force per unit area between the components as the internal pressure (π_i). The perusal of Table 2 reveals that the V_f values are maximum for glucose up to 0.03 m and least for fructose up to 0.06 m and maximum for galactose in the remaining
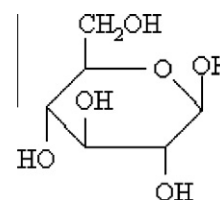
Figure 2 Structure of furanose (six-membered ring).

molalities whereas the least values are found for glucose in the molality range 0.07–0.10 m. Internal pressure is in general expected to show a reverse trend to that of V_f, some deviations are found in the reverse trend, i.e., the reverse trend as expected appears in glucose up to 0.03 m and for galactose from 0.06 m onwards. However, at 0.04 m and 0.05 m the reverse trend is not noticed and in these molalities among the three monosaccharides V_f is maximum for galactose, whereas for the same two molarities π_i is minimum for glucose and not for galactose. As regards fructose, the reverse trend is almost totally satisfied in these two parameters over all the molality ranges.

As already cited, the structural variations and the rearrangements within the molecule along with the active group participation may be attributed for these changes. The pyranose form is the only possible isomer in galactose whereas it is not so for glucose (Chakrabarti et al., 1972). Further, glucose exhibits all the three forms and it is needless to say that all the three forms are existing at all molalities but at different proportions and hence the extent of this geometry leads to these irregular variations of V_f and π_i in glucose and galactose whereas no such controversy appears in fructose.

Acoustic impedance (Z) is the impedance offered to the sound wave by the components of the mixture whereas the relative association (R_A) is the measure of extent of association of the components in the medium. The increasing trend in these parameters suggests the strengthening of interaction among the components. The interaction may be solute–solute or solute–solvent or solvent–solvent type. To ascertain the exact type, the trend of these two parameters may also be used. It is peculiar to note that these two parameters depend on density. Increase in density of the mixture clearly reveals that more mass is accumulated in the components. This reveals the significant

Figure 1 Structure of furanose (five-membered ring).

characteristics of the medium, i.e., though the density and velocity apparently seem to be independent of each other, in actual case, it is not so. For a single particle of a component in the mixture, the sound velocity depends on the size and mass of the particle. Size is important in offering cohesion effects whereas mass is important regarding the inertial effects. If the propagated sound energy is of enough strength, then only it can lift that mass and transfer the energy to the next particle. If the nearby particles are close, then the transfer speed will be more.

If the addition of solute increases the density (mass appreciably) of the mixture the medium is made more inertial and this leads to enhancement of the acoustic impedance. But in this case nothing can be revealed about the interactions. However, in most cases, the size will develop that increases the cohesion effects and so the components tend to move fast. This increase in particle movement is restricted by the surrounding particle that increases the net acoustic impedance. This shows the existence of specific interactions among the components, in particular solute–solute or solute–solvent. To have a much more deeper insight into this process, relative association may be used as a supporting parameter.

If the increase in velocity is due to size enlargement of the component in the mixture, cohesion will increase and thus it may lead to solvent–solvent interaction and in this case relative association will decrease with increasing molality. On the other hand, if the increase in velocity is due to mass accumulation, it may lead to either solute–solute interaction or solute–solvent interaction that depends on the magnitude of velocity variation. Larger variation is characterized by solute–solute, whereas a small variation indicates solute–solvent interaction. Such things can be best studied by relative association. If relative association increases but negligibly, after mixing the solute, then there will be poor association and this belongs to solute–solvent type. But if the variation of relative association is appreciable then it is to be taken as solute–solute type.

The observed trend of acoustic impedance and the relative association for each of the monosaccharides with molality are in general increasing, indicating two facts that the specific interactions are not solvent–solvent type and also the chances for complexation are enhanced with increasing molality.

Observing Table 3 and comparing the values of acoustic impedance (Z) and relative association (R_A), it is observed that acoustic impedance values are least for glucose over the entire molality range whereas maximum acoustic impedance is noticed for galactose up to 0.03 m and thereafter fructose shows maximum Z values. Further up to 0.03 m, acoustic impedance values of galactose and fructose closely resemble each other and hence it may be taken that fructose shows appreciable higher Z values at all molalities than galactose. Considering the relative association values, maximum values are recorded for galactose and minimum for glucose over all the molalities taken up. This maximum relative association for fructose may be due to the availability of ketone group (=CO), where free sites are more easily available than in aldehyde group (–CHO). Further, these vacant sites are also responsible for the observed increasing trend of acoustic impedance values. Hence it is clear that among the three monosaccharides taken up, fructose exhibits maximum interactions than glucose because of its keto group and the type is solute–solvent rather than the other two types. However, the recorded maximum R_A values for galactose reveal that galactose exhibits maximum interaction with the components, than glu-

cose and fructose. Further, the larger variation in the relative association values clearly suggests that both solute–solvent and solute–solute interactions predominantly exist in galactose solution. The relatively higher values of relative association signify that unlike interactions are relatively strong compared to like interactions. The former tends to increase while the latter causes a decrease in relative association and hence it leads a support to the solute–solvent interactions than the solute–solute interactions. Amalendu Pal (2001) observed similar trends in some binary mixtures.

The observed values of molar sound velocity (R) and molar compressibility (W) in all the monosaccharides are of increasing trend with molality, indicating that the magnitude of interaction is enhanced. The increasing trend of molar compressibility or molar sound velocity with increasing molality indicates the availability of more number of components in a given region, thus leading to a tight packing of the medium and thereby increasing the interactions. On comparing these values for the monosaccharides taken up, glucose records maximum values whereas galactose shows minimum values. On comparing the three monosaccharides, it is interesting to note that the sound velocity is minimum for glucose, whereas R is maximum for the same glucose. Similar controversy is appearing for galactose. This is true for W also. Such controversies clearly show that density plays the key role that characterizes the medium and sound propagation. Further it is needless to say that as molality increases, more mass is accumulated in the components of the mixture rather than size enlargement, as conceived from the trend of relative association. This clearly confirms the predictions obtained from relative association and acoustic impedance that the specific interactions are of solute–solute and solute–solvent types and the relative abundance of the solvent molecule compared to solute particles leads to the predominate presence of solute–solvent type.

4. Conclusions

(1) Geometry plays a key role in deciding the strength and type of interaction.
(2) Solute–solvent interaction predominates over the other type of interactions.
(3) Galactose is found to have the maximum interaction of all the three monosaccharides taken up for this study.

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