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Dielectric relaxation study of aqueous sugar using time domain reflectometry

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Abstract : The complex permittivity of sucrose, lactose, maltose and glucose, in water mixtures for various concentrations and temperatures have been measured as a function of frequency between 10 MHz to 10 GHz using time domain reflectometry technique Dielectric relaxation parameters *i e* static dielectric constant and relaxation time were obtained from the complex permittivity spectra using the nonlinear least squares fit method The relaxation behavior of these mixtures are explained by the Debye model. The variation in static dielectric and relaxation time of sugar in water solution provides, information regarding complete association and hydrophilic character of the solute particle. Thermodynamic parameters were determined, from the values of relaxation time.

Keywords : Dielectric permittivity, time-domain reflectometry, carbohydrate-water mixtures

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Carbohydrate is one of the important class of biomolecule and it plays multiple roles in living nature. Carbohydrates serve as a main energy resource, used to maintain the structural and functional properties of cells. They are structural elements themselves and they participate specifically in many biochemical reactions. The solution properties of carbohydrates particularly in aqueous solutions are of considerable interest, for various aspects of basic research, because the interactions of these bio-molecules with unique hydrogen bond network of water is of wide interest currently in liquid-state physics [1-3]. Water plays a unique and important role in biological processes. Since biological process takes place in aqueous media at relatively low biomolecular concentrations, interactions between biomolecules and water are often confined to an interfacial region surrounding the biomolecule. It is to be expected that a fraction of water molecules find themselves in a local environment interacting strongly with the biomolecule. Therefore, the increasing interest in biophysical and biochemical research is presently being directed toward the novel subdiscipline termed "Glycobiology" [4].

Sugars (sucrose, lactose, maltose and glucose) are the most common carbohydrates and are widely distributed throughout the biological systems. Sucrose is a disaccharide and is obtained from the sugar cane juice and sugar beets. Glucose is one of the monomer of the sucrose and is found in all photosynthetic plants, sugar cane, fruits, seeds, flowers, honey *etc.* In disaccharides, two monosaccharide units are joined together by a α-1,4 glycosidic bond in which anomeric carbon of one unit and an –OH of the other interacts. Lactose is disaccharide soley of mammalian origin, found in the milk of mammals to the extent of 5 percent. Disaccharide, maltose derives its name from its presence from malt, juice from sprouted and other cereal grains. Glucose occurs in the juice of fruits and in honey and is a common hydrolytic product of polysaccharides.

Molecular relaxation properties of glucose in aqueous solutions were studied using Ultrasonic relaxation

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spectroscopy [5] in the frequency range 0.2–2000 MHz. Dielectric relaxation properties of aqueous solutions of monosaccharide were studied [6] as a function of frequency between 300 KHz and 40 GHz. The aqueous solutions of monosaccharide at thermal equilibrium were reported [7] using broad band accoustical spectrometry. Ultrasonic absorption spectra were studied [8] between 100 KHz and 2 GHz for the aqueous solutions of manosaccharide. Ultrasonic absorption spectra in the frequency range 1–500 MHz were reported [9] for solutions of carbohydrates, both with without added Ca^{2+} and Ba^{2+} ion. Calorimetric and dielectric relaxation measurement were studied on anhydrous glucose and in water mixtures over a frequency range of 1 Hz to 100 KHz [10]. Pissis *et al* [11] studied the dielectric behaviour of frozen aqueous solutions of glucose and was done by the depolarization thermocurrents (DTC) method. Tait *et al* [12] reported ac measurement on aqueous glucose solutions in the GHz frequency range. No dielectric relaxation study have been reported on aqueous solutions of maltose and lactose.

The objective of the paper is to report the dielectric properties of aqueous sugar solutions using time domain reflectometry technique in the frequency range 10 MHz to 10 GHz over the temperature range of 15°C to 45°C. The study of dielectric relaxation of sugar's *viz* lactose, maltose, sucrose and glucose in water mixtures at microwave frequency may reveal details about their interaction with water.

Sugars (sucrose, lactose, maltose, glucose) were obtained commercially and used without further purification. The mixtures of various compositions were prepared by dissolving sugars in deionised glass distilled water.

The complex permittivity spectra was studied using time domain reflectometry (TDR) method. Detail of the TDR experiemental setup and procedure are described elsewhere [13,14]. A Tektronix 7854 sampling oscilloscope with 7S12 TDR unit was used. A fast rising step voltage pulse of 25 psec rise time generated by a tunnel diode was propagated through a coaxial line system. The sample cell was placed at the end of coaxial line in standard military application (SMA) coaxial cell of 3.5 mm outer diameter. All measurements were done under open load condition. The sampling oscilloscope monitored the change in the pulse after reflection from the sample placed in the cell. The reflected pulses $Rl(t)$ and $Rx(t)$ have been digitized with and without sample, respectively with 1024 sampling points, in time window of 5 ns. For the experiment, a SMA cell with 1.35-mm effective pinlength was used. The reflected pulses were added and subtracted in the oscilloscope memory and were transferred to PC/XT system for Fourier transformation as well as data processing [13,14]. The processing of the data was carried out to yield complex reflection coefficient spectra $\rho^*(\omega)$ over a frequency range from 10 MHz to 10 GHz [15]. The complex permittivity spectra $\epsilon^*(\omega)$ was obtained from reflection coefficient spectra $\rho^*(\omega)$ by applying the least squares fit method as described in our earlier publication [16,17].

The interaction between the solute and solvent molecules and also among themselves, play a vital role in determining the molecular structure of a solution. It is difficult to separate out the relaxation times of solute and solvent in the mixture from the dielectric spectra. The Debye function is generally used to obtain information about the average relaxation time in the mixture. The function has been fitted to the complex permittivity $[\varepsilon^*(\omega) = \varepsilon' - j\varepsilon'']$ spectra using nonlinear least squares fit method [16,17]. The static dielectric constant (ε_0) and relaxation time (τ) have been determined by fitting the complex permittivity spectra $\varepsilon^*(\omega)$ with the Debye equation

$$\varepsilon^*(\omega) = \varepsilon_\infty + (\varepsilon_0 - \varepsilon_\infty)/(1 + j\omega\tau) \qquad (1)$$

with ε_0, τ as fitting parameters in eq. (1). Since the permittivity spectra in the present study is the frequency range 10 MHz to 10 GHz, the ε_∞ in eq. (1) as determined from this study is just a fitting parameter. It was found to be a satisfactory procedure to keep the value of ε_∞ as a fixed parameter (3.5) for the determination of ε_0 and τ [16]. The resulting values of dielectric parameters for sugar-water mixtures at different temperatures are listed in Table 1. The static dielectric constant and relaxation time for pure water are in good agreement with earlier work [18,19]. These values are estimated by assuming 2% errors in the values of ε', ε'' from the goodness of fit of the data [17]. The variations of static dielectric constant and relaxation time with mole fraction of the sugar are shown in Figures 1(a,b). The values of static dielectric constant decreases as the concentration of maltose, sucrose, lactose and glucose in water increases. The decrease in static dielectric constant with increase in sugar in aqueous solution at high sugar, indicates formation of hydration sheaths so that water molecules are under the influence of sugar molecules, resulting in lowering the values of static dielectric constant. Thus, the decrease in the static dielectric constant indicates complete association of large amount of sugar in solution. The relaxation time increases in the presence of solute in water solution. The increase in relaxation time in water solutions may be due to the sugar molecules forming a structure around the water molecule so that it is difficult for water molecule to rotate. The change in the variations of relaxation time provides information regarding hydrophilic character of the solute particle. The trend in the relaxation time is typically same in lactose and maltose system while it shows comparatively sharp increase in case of glucose and sucrose. The dielectric constant as well as relaxation time decrease with increase in temperature.

Table 1. Temperature dependent dielectric relaxation parameters for sugar-water mixtures.

Conc of sugar	15°C		25°C		35°C		45°C	
	ϵ_s	τ(ps)	ϵ_s	τ(ps)	ϵ_s	τ(ps)	ϵ_s	τ(ps)
				Sucrose				
0.0	83.9	10.7	78.4	8.3	75.9	6.5	71.3	5.5
0.2	82.6	17.6	77.5	15.3	73.8	14.4	70.6	13.1
0.4	78.2	19.8	74.8	17.6	71.5	16.1	68.7	14.2
0.6	76.3	21.3	73.1	18.9	69.4	17.2	66.9	15.5
0.8	75.2	26.3	72.0	22.5	68.2	19.6	65.0	18.0
1.0	74.7	30.4	71.6	25.8	67.8	23.1	64.7	21.1
				Lactose				
0.0	83.9	10.7	78.4	8.3	75.9	6.5	71.3	5.5
0.2	75.7	11.3	72.8	9.3	70.7	8.3	68.1	7.2
0.4	72.8	13.2	70.5	10.7	68.7	9.4	67.1	8.6
0.6	71.4	15.4	69.4	12.9	67.5	11.4	66.7	10.1
0.8	69.2	17.9	67.4	16.3	66.5	13.9	65.4	12.3
1.0	66.5	22.4	64.7	18.2	63.8	15.9	62.3	14.2
				Maltose				
0.0	83.9	10.7	78.4	8.3	75.9	6.5	71.3	5.5
0.2	78.9	11.2	75.8	10.0	72.2	8.7	68.1	7.6
0.4	75.2	13.1	73.6	11.1	70.1	9.8	67.0	8.6
0.6	73.4	14.2	72.0	12.8	69.4	10.6	66.2	9.5
0.8	72.5	16.8	70.0	14.2	67.5	12.4	65.8	11.6
1.0	71.2	19.7	69.1	18.4	66.5	16.3	65.0	14.7
				Glucose				
0.0	83.9	10.7	78.4	8.3	75.9	6.5	71.3	5.5
0.1	81.7	16.1	77.1	15.0	74.1	14.1	70.2	13.4
0.4	79.8	17.8	76.9	16.5	73.5	15.1	69.3	14.4
0.7	77.9	20.3	75.8	17.7	71.3	16.8	68.7	15.9
1.0	76.2	21.5	75.3	18.4	70.8	17.3	68.3	16.6
1.3	75.9	24.2	74.8	21.7	69.5	19.6	67.5	17.8
1.5	75.4	30.7	74.2	26.7	68.9	23.0	67.1	20.7

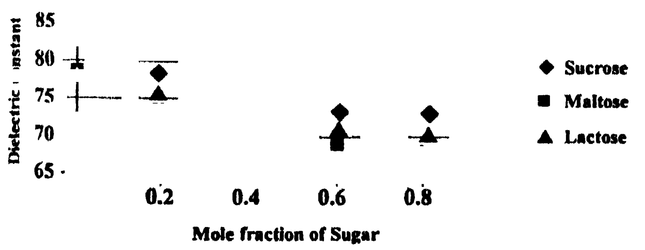

Figure 1(a). Variation of static dielectric constant with mole fraction of sugar in water.

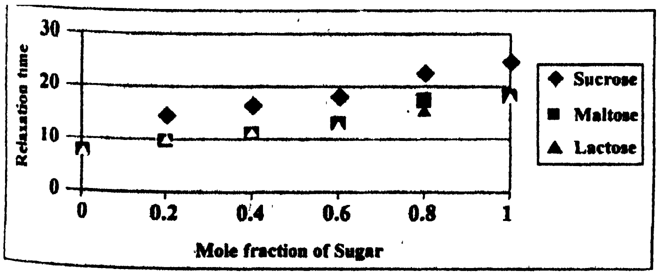

Figure 1(b). Variation of relaxation time with mole fraction of sugar in water

this is expected because of decrease of density with increase of temperature.

The thermodynamic parameters evaluated by using Eyring equation [20] for sugar-water system as follows :

$$\tau = (h/kT)\exp((\Delta H - T\Delta S)/RT), \qquad (2)$$

where ΔH and ΔS are enthalpy and entropy of activation in kJ/mol, respectively. τ is relaxation time, T is the temperature in degree kelvin and h is Planck's constant. The resulting values of activation energy as obtained by the least square fit method are reported in Table 2.

Table 2. Activation energies of sugar-water mixtures.

Conc. of sugar	ΔH KJ/mol		
	Sucrose	Lactose	Maltose
0.0	8.3	8.3	8.3
0.2	4.7	8.1	7.4
0.4	5.7	8.1	8.1
0.6	5.5	8.1	8.1
0.8	7.2	7.2	7.0
1.0	6.7	9.0	8.5

In this paper, a systematic study of dielectric relaxation parameters and thermodynamic properties of sugar-water mixtures has been carried out. The experimental dielectric relaxation data contains valuable information regarding solute-solvent interactions in the mixtures. The change in dielectric relaxation parameters in water solutions provides information regarding complete association and hydrophilic character of the solute particle.

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