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# A dielectric analysis of liquid and glassy solid glucose/water solutions

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## Abstract

Dielectric relaxation data covering a temperature range from above room temperature to below the glass transition for 40% (w/w) and 75% (w/w) glucose/water solutions in the frequency range between 5 and 13 MHz are presented. These data are used to obtain correlation times for the dielectric relaxation in the viscous liquid and the glass and are compared with correlation times determined from deuterium nuclear spin relaxation times [*J. Chem. Phys.*, 110 (1999) 3472–3483]. The two sets of results have the same temperature dependence, but differ in magnitude by a factor of 3, implying that the relaxation is a small-step rotational diffusion. Both the structural relaxation ($\alpha$ process) and the slow $\beta$ process are present. In the 40% glucose/water sample, there is a dielectric relaxation attributable to the ice that forms at low temperature. It is shown that the reciprocal of the viscosity, the correlation time derived from the dielectric relaxation, and the dc conductivity have a similar dependence on temperature. © 2000 Elsevier Science Ltd. All rights reserved.

*Keywords:* Dielectric relaxation; Glucose; Water; Molecular motion; Glass; Correlation times

## 1. Introduction

Sugar/water solutions are simple binary glass-forming systems. In spite of their everyday use, there is a lack of understanding at the molecular level of both the structure and dynamics in both the viscous liquid and the glassy state. These systems are also of interest as simple model food systems [1]. The formation of crystalline ice in frozen foods can adversely affect their quality, stability and hence their shelf-life, whereas freezing foods as glasses can improve these factors [1–3]. Slade and Levine [4] authored a series of review articles demonstrating that carbohydrate/water mixtures and polymeric glasses [5] have very similar thermal and mechanical properties. Slade and Levine showed that many of the properties of materials commonly used in the food industry could be understood on the basis of the physics and chemistry of glasses without resorting to such concepts as ‘bound water’. Sugar/water systems can be used to study glass formation and how rapidly water is removed from a glass in the form of crystalline ice. Experimental data from studies of sugar/water glasses are also useful for comparison with recent molecular dynamics simulations [6].

In the frequency regime below $10^{10}$ Hz, supercooled liquids usually exhibit at least two relaxation processes. At high temperature, the molecular motion in the non-viscous liquid is

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primarily determined by collisions between molecules. When the liquid is supercooled, oscillatory motion of molecules encaged by neighbors becomes important as does the hopping from one local environment to another. Dielectric relaxation supports the notion of two processes: a structural relaxation ($\alpha$ process) and a faster relaxation ($\beta$ process) whose origin is not fully understood. The structural relaxation is arrested at the glass transition temperature, $T_g$, on decreasing the temperature. The $\beta$ process bifurcates from the $\alpha$ process at a temperature that is well above the glass transition and continues into the sub $T_g$ region. Ideally, it should be possible to identify the molecular motions associated with each of these processes and experimentally determine the correlation time describing the timescale of these processes.

There have been several studies [7–11] of dielectric relaxation in sugar/water mixtures. In a series of three papers, Sugget et al. [7] used dielectric relaxation together with NMR spectroscopy to study molecular reorientation in sugar/water solutions. In the third paper of the series, Suggett and co-workers concluded that the dielectric relaxation is dominated by the rotation of the hydroxyl group and the hydroxymethyl side groups in the sugar, whereas the NMR relaxation is sensitive to the isotropic rotation of the sugar ring. Chan et al. [9] observed the permittivity and dielectric loss in anhydrous glucose and 90, 85 and 70 wt.% glucose/water mixtures. The non-Arrhenius $\alpha$ process above $T_g$, was shown to follow a Vogel–Tamman–Fulcher (VTF) relation. A $\beta$ process with an Arrhenius temperature dependence and activation energy of about 60 kJ/mol was found which extended below $T_g$. The addition of water, which reduced $T_g$, enhanced the strength of the $\beta$ relaxation, but lowered its rate. Following the model put forward by Johari and Goldstein [12], the $\beta$ process was envisaged as molecular reorientation occurring in high-volume, high-entropy regions in an otherwise rigid glass matrix. The increase in the strength of the dielectric relaxation was attributed to an increased correlation of dipole moments in these localized regions. Plots of the maximum frequency of the relaxation processes versus temperature showed a bifurcation of the two processes at a temperature well above $T_g$.

Although in the limited range of frequencies studied, the $\alpha$ process appeared in some studies [10] to fit an Arrhenius relaxation, it has been shown more recently by Gangasharan and Murthy [13] that the $\alpha$ process observed in the sugar melts is better represented by the VTF equation. On the basis of their dielectric relaxation data, it was concluded that in the pure sugar melts, the $\beta$ process is due to segmental rotation in the linear chain conformation and is not due to intermolecular degrees of freedom. Noel et al. [11] carried out a comparative study of the dielectric relaxation behavior of glucose, maltose and their mixtures with water. They showed that adding water to the sugar melts reduces $T_g$, and as a result, shifts the $\alpha$ relaxation to lower temperatures. In both the glucose and maltose mixtures, the addition of water increased the strength of the $\beta$ relaxation, but more so in the maltose case. The $\beta$ relaxation was thought to arise from motions of the hydroxymethyl groups attached to the hexose rings and from reorientation of the water molecules.

In an important $^{13}$C NMR study using specifically labeled anhydrous glucose, van Duss-choten et al. [14] probed the timescale and geometry of the reorientation of the exocyclic $CH_2OH$ group in relation to the main glucose ring. From a comparison of the data from the label on the ring and the $CH_2OH$ group, they concluded that the glucose ring and the exocyclic $CH_2OH$ group mobility are strongly correlated and that the rotational freedom of the $CH_2OH$ group should not be used to explain the faster $\beta$ relaxation process.

Recently [15], we have studied glucose/water solutions using deuterium NMR spectroscopy and have obtained correlation times for the molecular motions of both the glucose and the water molecules in the liquid and the glassy solid states. In comparison to NMR, what is particularly useful about dielectric relaxation is that a measurement of the frequency dependence can be directly fitted to a correlation function, thereby immediately providing the characteristic (or correlation) time for the process. Dielectric relaxation has been the principal source of information on the β process. In this paper, we present a dielectric relaxation study of 40% (w/w) and 75% (w/w) glucose/water solutions in the frequency range between 5 and 13 Hz. In this frequency window, the relaxation peaks are analyzed over a temperature range extending from above room temperature to below the glass transition. The primary interest of this work is to obtain correlation times for the dielectric relaxation in the viscous liquid and the glass and to relate these to correlation times determined from deuterium nuclear spin relaxation times previously obtained [15]. The objective is to compare correlation times originating from very different sources. In the case of dielectric relaxation, the correlation time is that of the fluctuating electric polarization of the sample, while for the deuterium nuclear spin relaxation, it is the time-dependent part of the electric field gradient at the deuteron site. Conductivities (dc) were measured as well, and these provide another avenue for studying the structural relaxation.

## 2. Experimental

Anhydrous dextrose (D-glucose) was purchased from Fisher Scientific Ltd. The water used to make the solutions was distilled, deionized and passed through a Millipore filter. This water was found to have a dc conductivity of approximately 4.0 μS/cm.

In order to measure the dielectric constant, two sample cells with a cylindrical geometry were used. In the first of these, no guard rings were included in the design. As this cell is for use at frequencies primarily below 4 MHz, fringe-field effects are small, and there is about a 4% error in the values of the complex dielectric constant. Open short compensation [16] was used to minimize the effects of residuals arising from the test leads and the test fixture. In these experiments, we are particularly interested in the peak frequency of the imaginary part of the dielectric constant. The assembly was totally encased in a copper cylinder that provided some electrical shielding, as well as ensuring the temperature was uniformly maintained. Using the pressure differential created by a carefully regulated vacuum pump, cool nitrogen gas was drawn through copper coils surrounding the cell. A heater wire wrapped between the copper coils provided a heat source. The heater, controlled by a Eurotherm digital controller, balanced the coolant and maintained the desired temperature. A copper–constantan thermocouple placed near the surface of the copper cell was used by the Eurotherm controller to set the temperature. This setup was used in the temperature range between 40 and − 100 °C. The temperature was regulated to within ± 0.5 °C over the course of a measurement, and the temperature of the sample was known to within ± 1 °C. The measurements on the 75% (w/w) glucose/water solution were performed using a high-precision cylindrical cell [17] having a Teflon coating a few micrometers thick separating a guard ring from the electrodes to minimize fringe-field effects. The temperature control for this cell was a slight modification of the arrangement described above. A heater was used, into which the cell fit tightly, and cold nitrogen gas was supplied from a pressurized dewar rather than by drawing the gas with a pump. This cell was far superior to the one initially constructed. The cell construction was such that the residuals were minimized to the point that the open/short compensation was no longer required. No new information would have been obtained by repeating the measurements of the 40% solution with this cell, however, because of the massive ice formation in this case.

At each temperature, the apparatus was allowed approximately 45 min to reach a stable temperature. A HP4192A impedance analyzer, controlled by a PC through an HP–IB interface connected to the BNC connectors of either cell in a four terminal pair (4TP) configuration, was used to measure the admittance, $Y$, as a function of frequency. The complex relative dielectric constant was determined from the admittance using the following relation:

$$\epsilon^*(\omega) = \frac{Y(\omega)}{i\omega C_0} = \epsilon'(\omega) - i\epsilon''(\omega) \qquad (1)$$

where $C_0$ is the empty cell capacitance.

For both cells used in this experiment, the effect of electrode polarization [18] has been neglected. Using typical values [18] for the polarization impedance, a calculation showed that there would only be a small discrepancy below 100 Hz due to the polarization. In liquids, there are frequently ions in solution that result in a dc conductivity. This conductivity manifests itself in the imaginary part of the relative dielectric constant [19]:

$$\bar{\epsilon} = \epsilon' - i\{\sigma_{\mathrm{dc}}/(\epsilon_0\omega) + \epsilon''\} \tag{2}$$

where $\bar{\epsilon}$ refers to the relative dielectric constant measured in an experiment.

Of the many phenomenological relations used in the description of dielectric relaxation Debye's model treats the ideal case of identical non-interacting dipoles. For this situation, all of the dipoles would relax with a single exponential relaxation time, $\tau$, and the resulting equation is:

$$\epsilon^* - \epsilon_\infty = \frac{\Delta\epsilon}{1 + (i\omega\tau)} \tag{3}$$

where $\epsilon_\infty$ is the relative permittivity at high frequency, and $\Delta\epsilon = \epsilon_s - \epsilon_\infty$ where $\epsilon_s$ is the static relative permittivity. Glasses and some other materials, however, display a markedly nonexponential relaxation. One of the attempts to model this behavior is the Havriliak–Negami (HN) modification [20] of the Debye equation:

$$\epsilon^* - \epsilon_\infty = \frac{\Delta\epsilon}{[1 + (i\omega\tau)^{1-\alpha_{\mathrm{HN}}}]^{\beta_{\mathrm{HN}}}} \tag{4}$$

which combines a symmetrical broadening ($\alpha_{\mathrm{HN}}$) and an asymmetrical broadening ($\beta_{\mathrm{HN}}$), where $\alpha_{\mathrm{HN}}$ and $\beta_{\mathrm{HN}}$ are constrained to lie between 0 and 1.

## 3. Results and discussion

*Analysis of the dielectric relaxation data.*— Fig. 1 shows some of the data for the imaginary part of the relative dielectric constant, $\epsilon''$, for the 75% glucose sample. At room temperature and above, there is an overwhelming contribution from the dc conductivity. The contribution to the observed $\epsilon''$ from the dc conductivity, $\sigma_{\mathrm{dc}}/\epsilon_0\omega$, is shown for the 6.1 °C data set in Fig. 1. At this temperature, there is evidence of a relaxation peak that had not yet moved into the frequency window available experimentally. The −27.8 °C data set in Fig. 1 shows that the dc conductivity has decreased by nearly two orders of magnitude, and a broad relaxation peak has moved well into the accessible frequency window. This set of data has been fit to a HN relaxation peak, as well as a dc conductivity. A further reduction in temperature decreased the dc conductivity, and at −43.4 °C a broad peak is visible, which resulted from the superposition of two relaxation peaks (the α and Johari-β relaxations), each of which was fit to a HN relaxation process. Also shown in Fig. 1 are the data for −43.4 °C, with the dc conductivity removed. The data (solid squares) clearly show that there are two relaxation processes at this temperature. The dc conductivity and the correlation times determined from this data set are given in the legend to the figure. At −63.2 °C the dc conductivity had dramatically decreased and only a single relaxation process can be seen within the frequency range of our apparatus at this temperature.

The correlation (relaxation) time, $\tau$, the HN parameters $\alpha_{\mathrm{HN}}$ and $\beta_{\mathrm{HN}}$ and the dc conductivity, $\sigma_{\mathrm{dc}}$, were determined at each temperature from the fitting procedure. It was found that for the data from both the 40 and 75% glucose samples the best fits to the relaxation peaks gave values of $\beta_{\mathrm{HN}} = 1$, indicating that the relaxation curves are symmetric. The value of $\alpha_{\mathrm{HN}}$ did vary depending upon the peak being fit and the specific values of $\alpha_{\mathrm{HN}}$ will be discussed below.

*Correlation times.*—The correlation times, $\tau$, derived from the fitting of the experimental data for $\epsilon''$ to the Havriliak–Negami relation given in Eq. (4), are shown for (a) the 40% glucose/water solution and (b) the 75% glucose/water solution in Fig. 2. When any experimental results for the glucose/water system are discussed, it is important to keep in mind the state diagram [1,15,21]. It is difficult to interpret the correlation times in the 40% solution due to the large amount of ice formation that occurred as the temperature was lowered. On the other hand, the data for the 75% solution are somewhat easier to interpret because upon cooling to the glass transition there was no evidence of any ice formation. In the 75% sample, $T_g$ is at −67 °C, while for the freeze-concentrated sample $T_g = -52$ °C. This nearly completely freeze-concentrated solution showed a single relaxation process at high temperature and a separation into an α and a Johari-β process at about −33 °C, which was well above the glass transition. At the beginning of the region where there was both an α and a β relaxation, the two peaks were not very well resolved and could only be fitted as a single broader-than-usual peak, indicating the overlapping of two relaxation processes. The results of two separate runs are shown in Fig. 2 for the 75% sample. A comparison of the two data sets is indicative of the error in the calculated correlation times arising from fitting the raw data with the HN function. There is a larger uncertainty in the fitting parameters in the region where there is overlap of the two dielectric relaxation peaks. In addition, the temperature dependence of the α process is not well represented in this figure, because the measurements were restricted to the observed times by the frequency limit of the dielectric relaxation apparatus. Had measurements at lower frequencies been available, a better picture of the arresting of the α process at $T_g$ would have been seen. At a concentration of 75% the parameter $\alpha_{HN}$ was about 0.6 for all the relaxation peaks. The degree to which $\alpha_{HN}$ increased from zero is a measure of the increase in the width of the

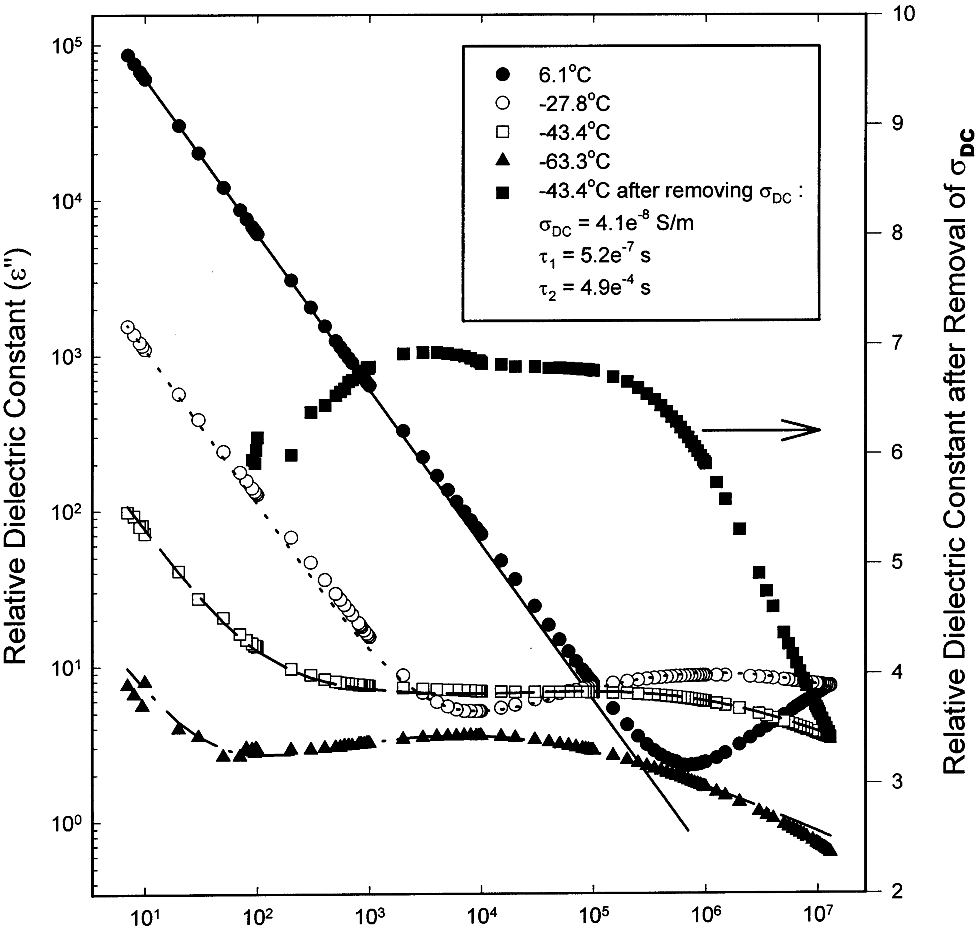
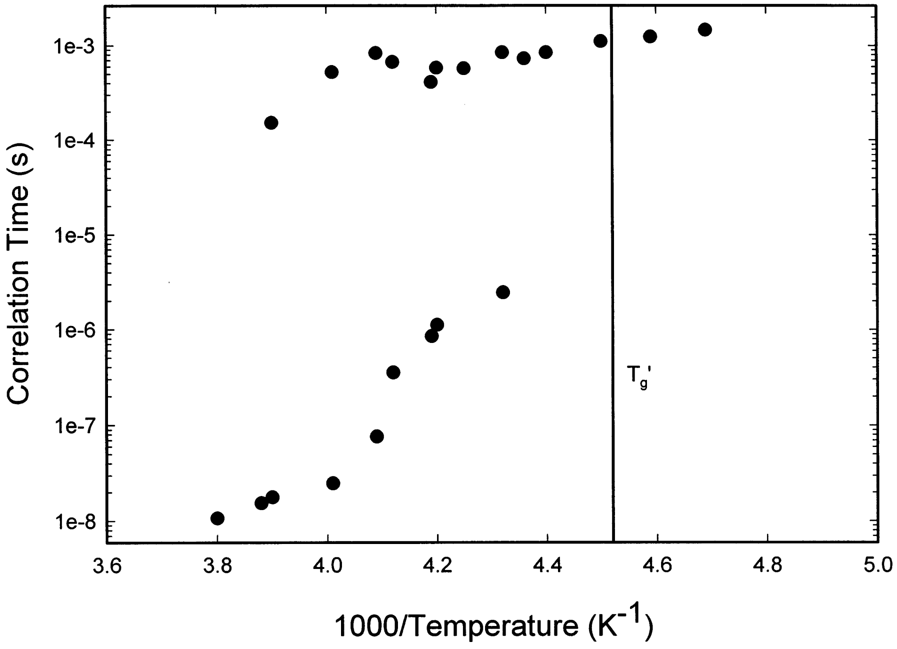


Fig. 1. Representative $\epsilon''$ data for the 75% glucose sample. The symbols are the experimental data, while the solid and dashed curves are fits to the data, taking into account the dc conductivity and relaxation processes. The contribution due to the dc conductivity is given by $\sigma_{dc}/\epsilon_0$, as indicated in Eq. (2). At 6.1 °C $\sigma_{dc} = 3.36 \times 10^{-5}$ S/m. The relaxation processes were modeled using the Haviliak–Negami equation given in Eq. (4). At −27.8 °C, $\sigma_{dc} = 6.13 \times 10^{-4}$ S/m and $\tau_\alpha = 1.3 \times 10^{-7}$ s. At −43.4 °C $\sigma_{dc} = 4.1 \times 10^{-8}$ S/m, $\tau_\alpha = 4.9 \times 10^{-4}$ s and $\tau_\beta = 5.2 \times 10^{-7}$ s. At −63.3 °C, $\sigma_{dc} = 3.36$ S/m and $\tau_\beta = 2.7 \times 10^{-5}$ s. At −43.4 °C, the data are redisplayed with the dc conductivity removed.

(a) 40% (w/w) Glucose/Water



(b) 75% (w/w) Glucose/Water

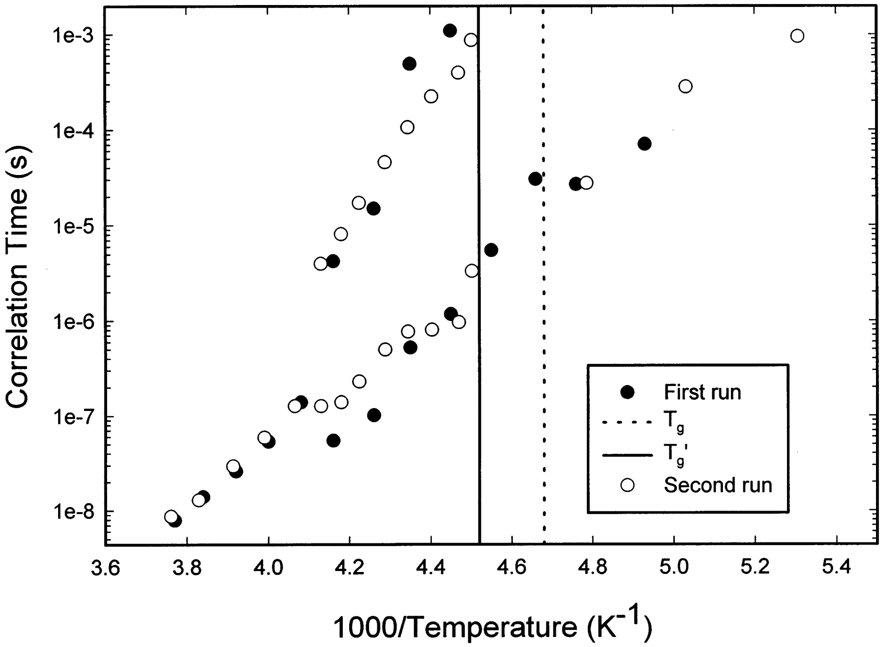


Fig. 2. Correlation times derived from fitting the $\epsilon''$ data to the Havriliak–Negami equation. For the 40% mixture shown in (a), the two peaks in the dielectric relaxation arise from the α process in the viscous liquid and the crystalline ice. Two sets of data are shown for the 75% mixture in (b). There is a bifurcation into α and β processes well above $T_g$.

relaxation from that of a pure Debye process. From the Kohlrausch–Williams–Watt [22] parameter, $\beta_{KWW} = 1 - \alpha_{HN} \approx 0.4$, it can be seen that there was a broad spectral width in these glucose/water solutions.

Although it is difficult to interpret the results from the 40% solution, a few general observations can be made. In the viscous liquid, the correlation times for 40% glucose/water were very similar to the times measured in the 75% solution. The values of the correlation time in the 40% solution, however, display a steeper slope probably resulting from the changing environment due to the freeze concentration of the solution. Another relaxation peak appeared at the ice formation temperature, approximately −23 °C, and the correlation times lengthened at a faster rate with decreasing temperature. The corresponding correlation times are shown in the upper part of Fig. 2(a). At the start, this peak was very weak, but as more ice formed in the system, the signal from this process dominated the observed relaxation data. The temperature dependence is well represented by an Arrhenius relation. The parameter $\alpha_{HN}$ was about 0 for this process indicating that it can be described by the Debye equations. The similarity of this data with these for pure $H_2O$ ice [23] leads to the conclusion that this relaxation peak and the associated correlation times in the upper part of Fig. 2(a) do not represent a β process in the viscous liquid, but rather a separate process resulting from the ice in this system. It was expected that the viscous liquid signal in the 40% solution would separate into an α and a Johari-β process as in the 75% solution; however, this was not observed presumably because the massive amount of ice formation produced a signal that obscured these details.

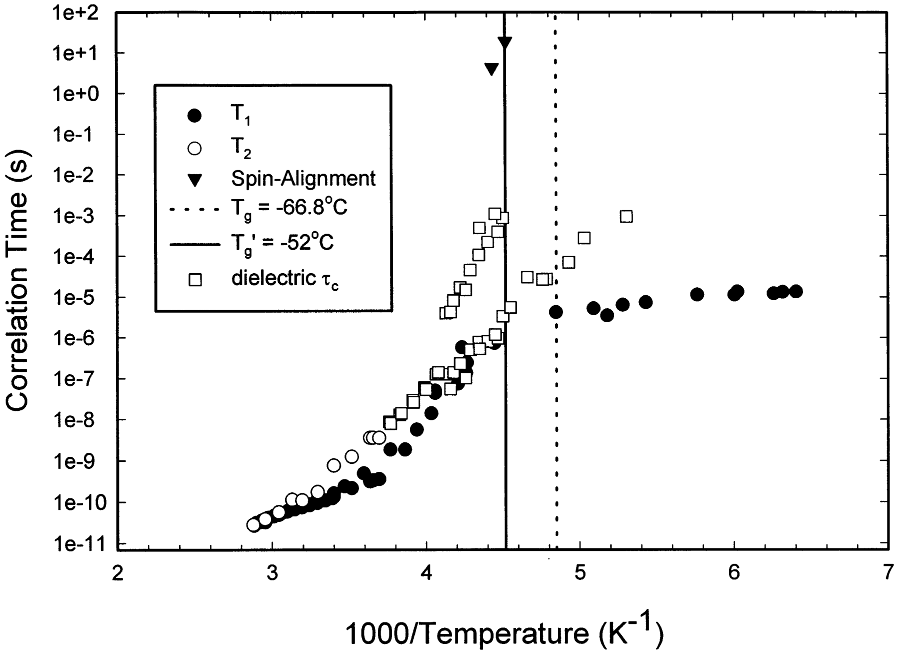
It is of interest now to compare the correlation times determined in this experiment with those previously determined by deuterium NMR spectroscopy [15]. In Fig. 3, the dielectric correlation times for the 75% solution are superimposed over the NMR correlation times for the motions of (a) the $D_2O$ molecules and (b) the glucose molecules in the 75% solution. The NMR measurements led to a correlation time for a second-rank tensor interaction that is the nuclear electric quadrupole interaction, whereas the dielectric measurements led to a correlation time for a first-rank tensor interaction [24]. The ratio of the dielectric correlation times to the NMR correlation times should therefore equal three in the case of small-step rotational diffusion. For random rotational jump models, the correlation times are independent of rank. Thus, for comparison, the dielectric correlation times have been reduced by a factor of 3 in Fig. 3. The dielectric correlation times agree well with the NMR correlation times for the motion of the water molecules above $T_g$. Hence, it is clear from the figure that the dielectric measurements have measured the correlation time of the motion of the water molecules and not the glucose molecules. The dielectric measurements were dominated by the water because of the much larger dipole moment of the water molecule. The dielectric relaxation and the nuclear spin relaxation times show evidence for both the α and β processes.

In Fig. 3(a), the correlation times from the two measurements show some discrepancy in describing the α process near $T_g$. The correlation times derived from spin-alignment measurements were much longer than those from dielectric relaxation. The cause could be that the average relaxation times derived from the spin-alignment data are an arithmetic mean [15], as compared with the geometric mean of the distribution obtained from the other NMR relaxation results. Below $T_g$ there was a very real difference in the magnitude and temperature dependence of the correlation times determined for the β process from NMR and dielectric relaxation. The source of this difference needs further investigation. As alluded to in Section 1, the origin of the β process in glucose/water glasses is not yet clear. In a recent paper Blochowicz et al. [25] have studied glass-formers that do not exhibit a β process. These glass-formers also exhibit a kink in the NMR $T_1$ data near $T_g$ as seen in Fig. 3. It was concluded by Blochowicz et al. [25] that although the NMR $T_1$ data below $T_g$ would be affected by the β process, the data could not be attributed solely to that source. A study is in progress using glucose that is deuterated at various sites in order to determine first the origin of the β process in the glucose/water system, and also to determine how the β process influences the NMR $T_1$ data.

The ratio of the correlation times from dielectric relaxation and NMR being equal to 3 is an interesting result. In the case of some simple molecular glass-formers such as 2-terphenyl, toluene and glycerol, the correlation times derived from dielectric relaxation and the Kerr effect (rank 2) reported by Williams [26] were independent rank and it was argued that this result was evidence for molecular reorientation taking place via large-angle jumps. However, this is contrary to the evidence for small-angle processes derived from deuterium NMR stimulated-echo experiments [27].

The data presented in this paper are in agreement with the measurements of Chan et al. [9] and Noel et al. [11] as shown in Fig. 4.

(a) $D_2O$



(b) Glucose

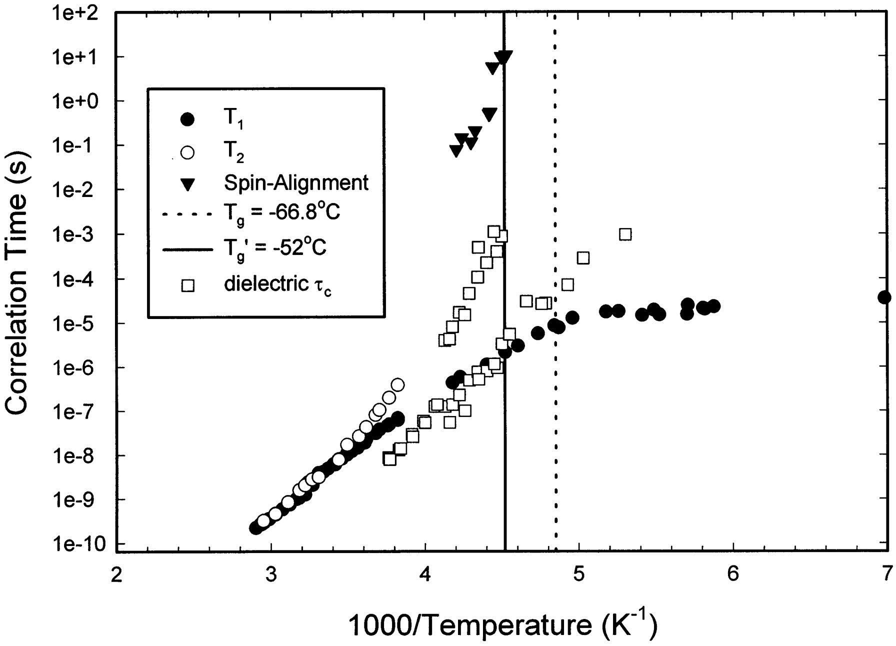


Fig. 3. Comparison of dielectric spectroscopy correlation times with NMR correlation times for the molecular motions of (a) the deuterated water, and (b) the glucose, determined in 75% (w/w) glucose/water solution.

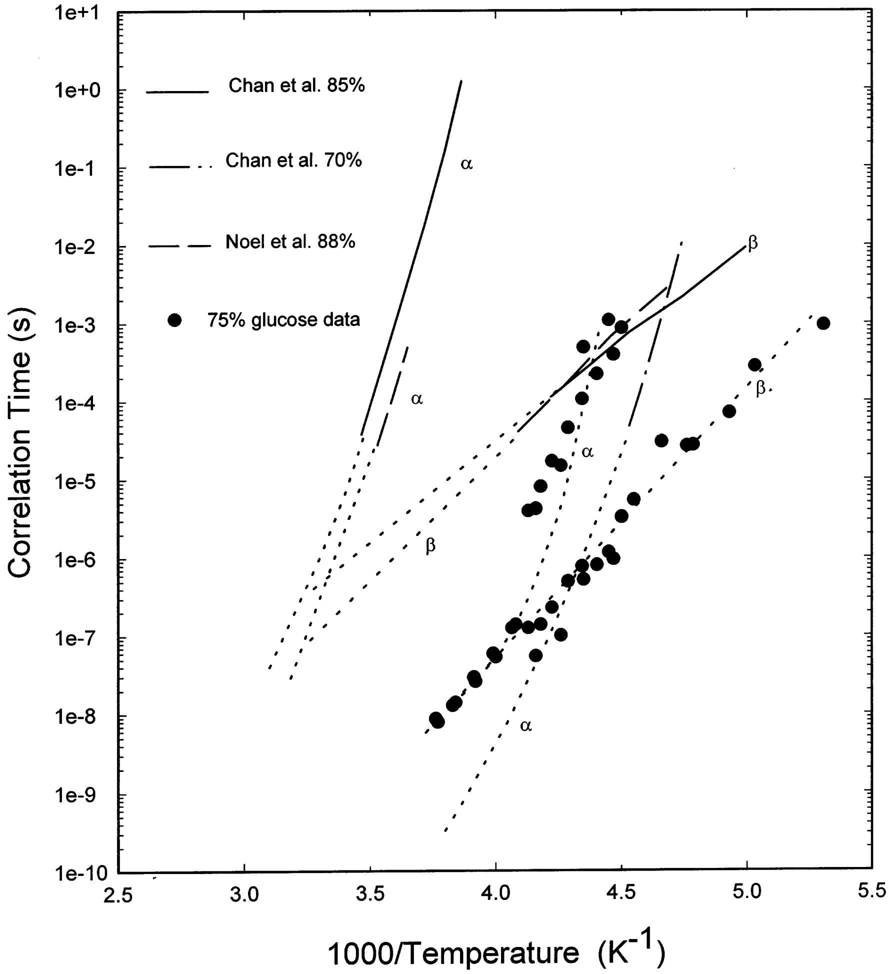


Fig. 4. A comparison of correlation times from this work to those of Chan et al. [9] and Noel et al. [11]. Extrapolations of the original data from Chan et al. and Noel et al. are indicated with the dashed curves. The dashed curves through the data points from this report indicate the bifurcation of the α and β processes.

The frequency dependence of the dielectric relaxation data reported previous to this paper was limited to the region below 100 kHz. As a result, the coalescence of the α and β processes into a single process at high temperatures was not observed. The collected data, however, give a reasonably coherent picture of the evolution of the correlation times for both the α and β processes as functions of temperature and water content.

*dc Conductivity*.—The-measured dc conductivities in 40% (w/w) and 75% (w/w) glucose/water solutions are shown in Fig. 5. It is difficult to identify the source of the dc conductivity. However, we are satisfied that the probable source is trace amounts of $Na^+$, $K^+$ and $Cl^-$ ions. The differences in conductivity for these two solutions can be understood if the state diagram is taken into account. The 75% solution produced very little ice, if any at all, upon cooling and hence did not undergo significant freeze concentration, forming a glass near − 67 °C. The 40% solution, on the other hand, produced a large amount of ice thus rapidly freeze concentrating the remaining solution, which formed a glass nearer to $T'_g$ ( − 52 °C), the glass transition temperature for a maximally freeze-concentrated solution [1]. The conductivity for the 40% solution showed a change in slope at the point (indicated by the arrow in Fig. 5) where ice began to form, and showed markedly different temperature dependencies on either side of this division. The conductivity for the 75% solution case, however, varied continuously over the entire temperature range, indicating the absence of ice formation. Notice as well, that at the point at which the dc conductivities became so small that they were beyond the range of the measuring instrument, the concentrated solutions were near to their respective glass transitions.

Stickel et al. [28] showed that the reciprocal of the viscosity, the conductivity due to ionic impurities and the peak frequency from the dielectric relaxation in several simple molecular glasses including salol, propylene carbonate and 1-propanol had the same temperature dependence. Fig. 6 shows $\sigma_{dc}$ for the 40 and 75% glucose solutions as a function of reciprocal temperature superimposed on the reciprocal of the measured correlation times derived from the peak dielectric relaxation frequencies and on the reciprocal of the viscosity taken from the paper by Kerr et al. [29]. In Fig. 6, the dashed curves represent the reciprocal of the viscosity, which is a direct measure of the structural relaxation of the supercooled liquid. The scales have been adjusted to bring the data from the different measurements roughly into coincidence in the center of the graph. It is interesting to compare the temperature dependence of the structural relaxation with that for the dc conductivity and for the reciprocal of the correlation times derived from the dielectric relaxation. For the 75% solution $\sigma_{dc}$ and the $\alpha$ process from dielectric relaxation followed the structural relaxation as expected. The $\beta$ process followed a much different path. For the 40% solution, the $\alpha$ process from dielectric relaxation again followed the structural relaxation as expected; however, $\sigma_{dc}$ follows the structural relaxation only until ice began to form in the sample. At the temperature where a dielectric peak was observed from the ice in the solution, the temperature dependence of the $\alpha$ process deviated from the structural relaxation.

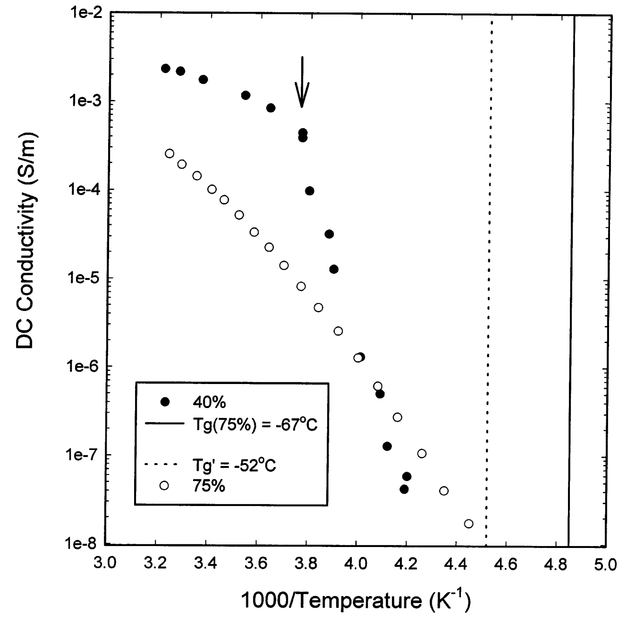


Fig. 5. The temperature dependence of the dc conductivities in the 40 and 75% glucose/water solutions.

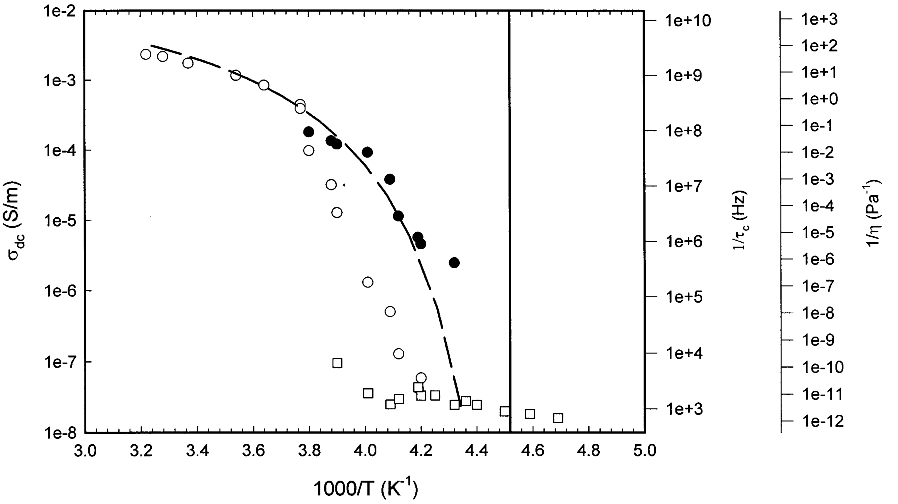
## 4. Conclusions

In the 75% glucose/water solution the correlation times for the motion of the $D_2O$ molecules, as determined from NMR and dielectric relaxation, agreed very well, thus verifying that the dielectric response was dominated by the large dipole moment of the water molecule. Both the structural relaxation and a secondary $\beta$ process were observed. The NMR and dielectric relaxation data showed the same temperature dependence, but differed in magnitude by a factor of 3, suggesting that the relaxation occurred as a result of small step rotational diffusion.

In the 40% solution, the formation of ice and resulting freeze concentration were reflected in the temperature dependence of the $\alpha$ process and the dc conductivity. With the formation of crystalline ice, a new secondary relaxation peak well described by the Debye equations was observed. This peak was attributed to the ice that formed in these solutions.

In the future, work will be required to determine the origin of the $\beta$ process and the reasons for the differences observed by dielectric relaxation and nuclear spin relaxation below $T_g$. Because the $\beta$ process is relativity slow on an NMR timescale, it may be possible to investigate the molecular reorientation involved in this process using two-dimensional NMR exchange techniques.

(a) 40%



(b) 75%

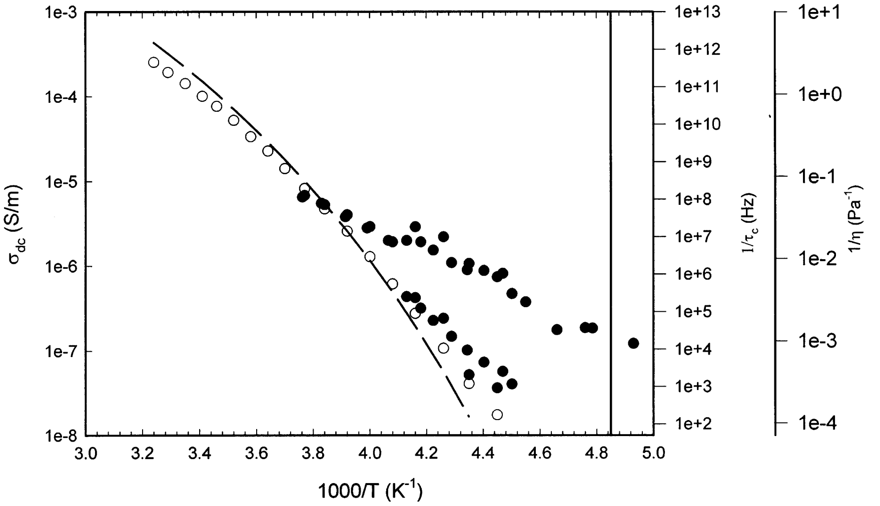


Fig. 6. A comparison of the data from dielectric relaxation, dc conductivity and viscosity in the region around the glass transition for both the 40 and 75% glucose/water mixtures. The dashed curves represent the reciprocal of the viscosity taken from the results of Kerr et al. [29]. The open circles represent the dc conductivity and the closed circles are the reciprocal of the correlation times derived from the dielectric relaxation measurements. The open squares represent the reciprocal of the correlation times derived from the dielectric relaxation in the crystalline ice.

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