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## On the pressure dependence of the viscosity of aqueous sugar solutions

Received: 13 August 2001  
Accepted: 5 February 2002  
Published online: 16 April 2002  
© Springer-Verlag 2002

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**Abstract** New experimental data for the pressure dependence of the viscosity of aqueous solutions of different sugars are presented.

Measurements were carried out with a gravity driven high pressure viscometer with a maximum pressure of 700 MPa. The influence of both concentration and temperature on the pressure dependence of the viscosity is considered. Next to the data, a viscosity model based on a suspension model is introduced and it is shown that it is able to predict the viscosity for sugar solutions in a

broad parameter range, including pressure. It is demonstrated that the relative viscosity for varying sugar mass fraction at constant pressure coincides with the pressure-dependent relative viscosity for a fixed sugar mass fraction, suggesting that there are no structural changes occurring under pressure. A brief interpretation of the viscosity model is given.

**Keywords** High pressure viscosity · Sucrose and glucose solutions · Viscosity model

### Introduction

High pressure treatment is a novel technology in food processing with various advantages. Chefteil (1992) gives an overview. At present, potential applications in science and engineering are discussed in various respects. For a better understanding of the processes occurring under pressure and the effects of pressure on cellular and molecular structures there is a need for data on thermophysical properties of food ingredients under pressure. So far, there is a lack of knowledge, because suitable in-situ measurement techniques are missing. An important thermophysical property is the viscosity, because it provides or supplements information on pressure-induced structural changes, on fluid dynamic and diffusion processes. There are some data in the field of petrochemistry – see, e.g. Kuss and Golly (1972); for lubricating oils see, e.g. Galvin et al. (1973) or Larsson and Jonsson (1997) and for polymers see Mackley and Spitteler (1996) or Briscoe et al. (1999). However there is a lack of data concerning the

pressure dependence of the viscosity of fluid food ingredients and aqueous solutions. Only few data are available, i.e. sodium chloride solutions (Horne and Johnson 1967), urea solutions (Sawamura et al. 1997) and water-ethanol mixtures (Matsuo 1994; Harlow 1967).

No data have been found in the literature by the authors for sugar solutions. The knowledge of these viscosities is important as they are the basis for real food systems, e.g. milk or fruit juices.

In addition to the lack of data, there is no theory to describe the pressure dependence of the viscosity of aqueous solutions. So far, only some theories exist for the pressure dependence of the viscosity of pure liquids, which molecules are spherical and have simple intermolecular interactions; see, e.g. Frisch et al. (1940) and Van Wijk and Seeder (1937). However, these theories are not able to predict the viscosity of unknown liquids in compressed state.

Therefore the present work provides new measurement data on the pressure dependence of the viscosity

of aqueous solutions of sucrose and glucose. Next to the measurement data, a model to describe the pressure dependence of the viscosity of the sugar solutions is presented. It is based on a suspension model that is valid for ambient pressure. It is shown that an extension of the model to describe viscosities at high pressure is possible.

## Material and methods

The glucose used in the experiments was D(+)-glucosemonohydrate with a molecular weight of 198.17 g/mol and was supplied by J.T. Baker, Holland. The sucrose solutions were prepared with commercial sugar as it has a very high purity. The solutions were prepared using demineralised water as solvent. The solutions were prepared by weight to achieve defined mass fractions  $w$ , i.e. the mass of sugar divided by the total mass of the solution. For D(+)-glucosemonohydrate the mass of sugar refers to the dry substance. The sugar solutions were prepared by heating them up to 50 °C and stirring with a magnetic stirrer for about 30 min.

The viscosity and density data for sucrose and glucose solutions at ambient pressure were taken from Mageean et al. (1991) in the temperature range of 20 °C  $\leq T \leq$  80 °C and in the mass fraction range of 0.005  $\leq w \leq$  0.80. The viscosity and density data for water at ambient pressure were taken from Sato (1989).

The viscosity measurements were carried out with a rolling ball method as described by Hoeppler (1933), but with variable inclination angle  $\alpha$ . The viscosity was determined according to the equation

$$\eta = Kt(\rho_s - \rho_f) \quad (1)$$

Here  $\eta$  is the shear viscosity,  $K$  a pressure and temperature dependent calibration factor, which has been determined experimentally,  $\rho_s$  the density of the solid sphere and  $\rho_f$  the fluid density. The rolling time  $t$  is measured by an inductive method; i.e. a small voltage peak is produced when the steel sphere passes one of two pickup-coils mounted on the outside of the high pressure tube.

The pressure is generated by compressing the fluid to be measured with a manual piston pump. The tube is kept at constant temperature by a water jacket connected to a thermostat and is connected to a pressure transducer (Wika GmbH, Klingenberg, Germany). The sphere is brought to its initial position by rotating the viscometer. The set-up of the measurement technique as well as the pressure dependent calibration procedure and validation of the method is described in more detail by Först et al. (2000). The pressure dependence of the viscosity was determined relative to the viscosity at ambient pressure.

For both the determination of the viscosity with the rolling ball method and the application of the viscosity model, the pressure dependence of the density must be known. For the solvent water the pressure dependence of the density is known from Saul and Wagner (1989). No data for the pressure dependence of the density exist for sugar solutions. Therefore, the pressure dependence of the density has been determined experimentally. For the density measurement a direct compression method was used. The manual piston pump, that has a defined volume and length of stroke, was fixed to a tube with a volume of 2 ml connected to a pressure transducer. The volume of the manual piston pump, that was also used for the viscosity experiments, is 4 ml. The length of stroke was read on a scale on the piston pump and the pressure dependent volume reduction  $\Delta V$  was measured allowing a determination of the density under pressure.

## Pressure dependence of the viscosity of sugar solutions

In the following section some experimental results for the pressure dependence of the viscosity of sugar solutions are described briefly in order to demonstrate the influences of temperature and concentration. A more detailed interpretation of the results is given later where the viscosity model is presented. The first set of experiments was carried out with sucrose solutions. The measurement range refers to mass fractions between 0.01 and 0.60 and a temperatures between 5 °C and 60 °C. The pressure dependence of the viscosity of sucrose solution for a constant temperature  $T=20$  °C and with the mass fraction  $w$  as parameter is graphically shown in Fig. 1.

Figure 1 shows a strong increase of the pressure dependence of the viscosity with growing mass fraction  $w$ . For  $w=0.6$  the viscosity increases about 6.75-fold going from ambient pressure to 700 MPa; for water the increase is only 1.37-fold in the same pressure range. In particular, for the higher mass fractions the data can be approximated by an exponential function with the exponent being concentration dependent. The exponential pressure dependence is already known for pure liquids, see e.g. Kuss (1965). For the low mass fraction ( $w=0.01$ ) the measurement values in Fig. 1 slightly deviate from the exponential function and the viscosity initially decreases with increasing pressure and increases at higher pressures. The viscosity minimum is caused by the anomalous behaviour of the solvent water that is dominating the system for low concentrations. The anomalous viscosity behaviour for water has been reported in an earlier publication; see Först et al. (2000). The

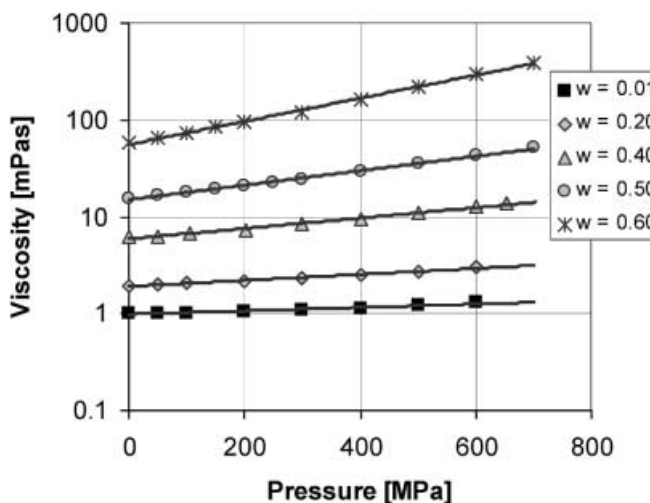


Fig. 1. The pressure-dependence of the viscosity of sucrose solution for  $T=20$  °C and different mass fractions. Exponential functions (lines) are fitted to the measurement data (symbols)

measurement data for sucrose solution are listed in Table 1. For comparison, the data for water are also included.

Next to the concentration, the temperature exerts a strong influence on the pressure dependence of the viscosity. With increasing pressure, the temperature dependence of the viscosity increases. For atmospheric pressure and going from 60 °C to 5 °C the viscosity increases 8.7-fold, whereas at 600 MPa the increase is 13.4-fold in the same temperature range. The Arrhenius function holds for the description of the temperature dependence of the viscosity in the whole pressure range considered here with the activation energy being pressure dependent. The results for the viscosity with the temperature as parameter are listed in Table 2.

Both the influence of concentration and of temperature combined show that the pressure dependence of the viscosity is strongest for low temperature and high mass fractions of sugar. These results are interpreted in more detail in the section below, where the viscosity model is introduced.

In addition to sucrose solution, the behaviour of pressurised glucose solutions was investigated for

different temperatures and  $w=0.20$ . The results are shown in Table 3. Comparing Table 1 and Table 3 for  $w=0.20$  and  $T=20$  °C it follows that the pressure dependence of the viscosity of glucose is slightly weaker than for sucrose solutions. This is due to the different shapes of sucrose and glucose molecules as discussed in the next section.

### Viscosity model for sugar solutions

In the following section a model is presented that allows the prediction of the pressure dependence of the viscosity in a broad parameter range if the pressure dependence of the viscosity of the solvent and of the density of the solution are known. At first, sucrose solution is considered. The viscosity model is based on a suspension model that Vand (1948a, 1948b) has derived from hydrodynamic considerations. According to this model the viscosity of sucrose solution at ambient pressure can be described with the equation

$$\frac{x}{\ln(\eta_r)} = q_0 + q_1x + q_2x^2 + \dots \text{with } x = \rho w, \quad (2)$$

**Table 1.** Measurement data for sucrose solution (in mPas) for  $T=20$  °C in a mass fraction range between  $0.01 \leq w \leq 0.60$ . The data for water (second column) are from Först et al. (2000)

Pressure [MPa]	Solvent viscosity [mPas]	w = 0.01	w = 0.20	w = 0.40	w = 0.50	w = 0.60
0.1	1.00	1.03	1.95	6.17	15.54	58.08
100	0.99	1.01	2.04	6.65 <sup>a</sup>	17.94	73.86
200	1.01	1.04	2.15	7.37	21.30	93.93
300	1.06	1.08	2.32	8.31	24.78	121.02
400	1.12	1.15	2.51	9.45	29.77	162.09
500	1.20	1.21	2.75	10.84	35.17	219.93
600	1.28	1.29	2.98	12.66	42.55	300.80
700	1.37	–	–	–	51.93	391.83

**Table 2.** Measurement data for the viscosity of sucrose solution (in mPas) for  $w=0.50$  in a temperature range between  $5$  °C  $\leq T \leq 60$  °C. The data for water in the left column for each temperature are from Först et al. (2000), if not specified otherwise

Pressure [MPa]	T = 5 °C <sup>a</sup>	T = 20 °C	T = 40 °C	T = 60 °C <sup>b</sup>
0.1	1.53	32.95	1.00	15.54
100	1.44	35.86	0.99	17.94
200	1.46	42.35	1.01	21.30
300	1.54	51.40	1.06	24.78
400	1.67	64.80	1.12	29.77
500	1.83	81.13	1.20	35.17
600	2.03	103.58	1.28	42.55
700	–	132.06	1.37	51.93

**Table 3.** Measurement data for the viscosity of glucose solution (in mPas) for  $w=0.20$  in a temperature range between  $5$  °C  $\leq T \leq 40$  °C. The data for water in the left column for each temperature are from Först et al. (2000), if not specified otherwise

Pressure [MPa]	T = 5 °C <sup>a</sup>	T = 20 °C	T = 40 °C
0.1	1.53	3.11	1.00
50	1.46	3.08	0.99
100	1.44	3.09	0.99
200	1.46	3.23	1.01
400	1.67	3.77	1.12
600	2.03	4.63	1.28

where  $x$  is the sugar mass concentration that can be calculated from the mass fraction  $w$  and the density  $\rho$  of the solution. The relative viscosity  $\eta_r$  of the solution is defined as

$$\eta_r = \frac{\eta(T_1, p_1)}{\eta_s(T_1, p_1)} \quad (3)$$

where  $\eta$  is the viscosity of the solution and  $\eta_s$  of the solvent in the same state. The coefficients  $q_i$  of the equation are related to physical properties of the suspended particles and their interactions. The coefficient  $q_0$  provides information on hydration and shape of the molecules and refers to the dilute solution. The coefficients  $q_1$  and  $q_2$  give information on intermolecular interactions and on the change of flow field due to the presence of the particles. Therefore they are related to the behaviour of the more concentrated solution. For  $q_0$  and  $q_1$  Vand (1948b) gives the relations

$$q_0 = \frac{D_s}{h_0 k_1} \text{ and } q_1 = \frac{1}{k_1} \left( \frac{3(h_0 - 1)}{h_0(h_0 + 2)} - \frac{r_2(k_2 - k_1)}{k_1} - Q \right), \quad (4)$$

where  $D_s$  is the density of the non-hydrated molecules in solution,  $h_0$  is a dimensionless factor describing the volume increase of the molecules due to hydration in diluted state and  $k_1$  and  $k_2$  are shape factors related to a single particle and a particle in collision, respectively.  $Q$  is a constant describing the hydrodynamic interactions and  $r_2$  is a time constant describing the time fraction a particle spends in collision with another particle. The shape factors for spheres are  $k_1 = 2.5$  and  $k_2 = 3.175$ .

The model of Vand (1948b) is not able to predict viscosities, but can be used to give insights into hydration effects and particle-particle interactions.

Here, the model was used to describe the concentration dependence of the viscosity in the temperature range 20–80 °C. Furthermore, it was checked whether it can be extended to high pressures. The coefficients  $q_i$  were determined by approximating the viscosity data at ambient pressure using Eq. (2). It has been found out that linear functions are sufficient to describe the data in the form  $x/\ln(\eta_r)$  vs  $x$  and therefore the second power term in Eq. (2) can be neglected.

Data analysis shows that the variable  $q_0$  is temperature dependent, whereas  $q_1$  can be considered as temperature independent. A further investigation of the temperature dependence of  $q_0$  shows that it can be described with an Arrhenius function:

$$q_0 = q_0^* e^{-(E_a/RT)}. \quad (5)$$

where  $E_a$  is an energy of activation and  $R$  is the gas constant. Inserting Eq. (5) into Eq. (2) and neglecting the second power term of Eq. (2) yields the non-dimensional Eq. (6):

$$\frac{x^*}{\ln(\eta_r)} = 1 + q_1 x^* \text{ with } x^* = \frac{x}{q_0^* e^{-(E_a/RT)}}. \quad (6)$$

Separation of the variables yields

$$\ln(\eta_r) = \frac{x^*}{1 + q_1 x^*}. \quad (7)$$

Equation (2) is now reduced to a very simple form and it is possible to make a comparison between Eq. (6) and the measurement data at ambient pressure. This comparison is presented graphically in Fig. 2.

It is shown in Fig. 2 that the measured values can be described very well with Eq. (6). Therefore Eq. (6) can be used to predict the viscosity of sucrose solutions in a broad parameter range of temperature and concentration, but only for ambient pressure. In a next step it is checked whether Eqs. (6) and (7) can be extended to predict also the viscosity at high pressure. To study this, the measured pressure dependent viscosity values (cf. Tables 1 and 2) have to be transformed into a relative viscosity analogous to the ambient pressure data using Eq. (3). Furthermore, the pressure dependent concentration  $x$  has to be determined, which is calculated from the mass fraction times the pressure dependent density  $\rho$  of the solution using Eq. (2). If Eq. (7) is used and compared to the measurement data obtained at high pressures for sucrose solutions it is shown in Fig. 3 that the model is also valid for high pressures up to at least 700 MPa.

The maximum relative deviation between the model equation and the measurement points is 10%. This may be due to uncertainties in the density. Another reason could be slight pressure-induced structural changes in the sugar solution.

The same procedure that is explained here for sucrose solutions has been carried out for glucose solutions and

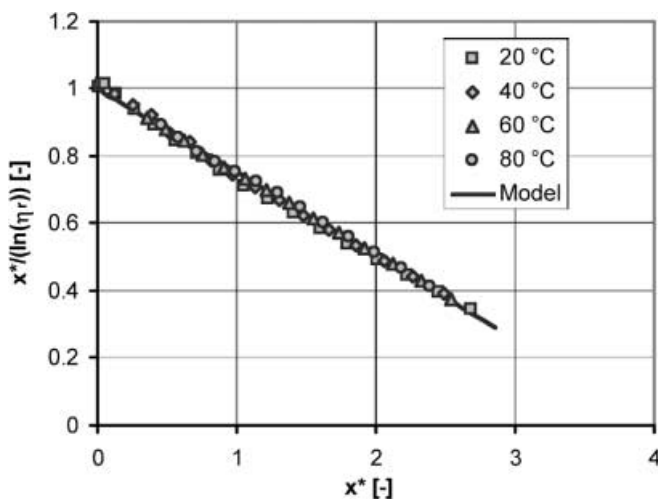


Fig. 2. Comparison between the ambient pressure measurement data for sucrose solutions (taken from Mageean et al. 1991), plotted as  $x^*/\ln(\eta_r)$  vs  $x^*$  (symbols), and Eq. (6) (line)

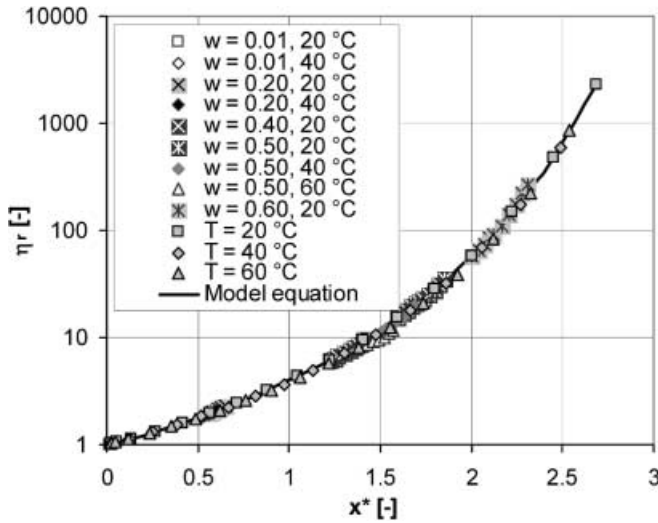


Fig. 3. Comparison between the high pressure measurement data, plotted as  $\ln(\eta_r)$  vs  $x^*$  (symbols) and Eq. (7) (line)

similar results were found. The only difference between glucose and sucrose is related to the numerical values of the coefficients in Eq. (4). This difference is related to the fact that glucose is a monosaccharide whereas sucrose is a disaccharide.

In the following section a brief interpretation of the model is given in order to understand the rheological behaviour of the solution under pressure. Figure 3 shows that Eq. (7), that has been derived from Eq. (2), also holds for the prediction of the high pressure viscosities in the whole pressure range examined in this work. The pressure dependence of the viscosity can therefore be described by its concentration dependence. As  $x = \rho w$  (see Eq. 2), a change in the concentration  $x$  can be achieved either by raising the pressure and hence the density  $\rho$  (holding the mass fraction  $w$  constant) or the mass fraction  $w$  (holding the pressure  $p$  constant). As shown in Fig. 3, a change in  $\eta_r$  is achieved by changing one or both variables; e.g. for a sucrose solution of  $T = 20^\circ\text{C}$  and  $w = 0.50$  a pressure jump from 0.1 to 700 MPa is equivalent to a change from  $w = 0.50$  to  $w = 0.56$  at ambient pressure. The combination of both arguments means that the coefficients  $q_0$  and  $q_1$ , which can be easily determined from ambient pressure viscosity and density data, are independent of pressure. Taking into consideration Eq. (4) it is concluded that there is no direct influence of pressure on shape, hydration and intermolecular interactions. Pressure exerts only an indirect influence by the increase in concentration  $x$ . Therefore, the model is capable of predicting unknown viscosities of sugar solutions under pressure if the density of the solution and the pressure dependence of the solvent viscosity are known. Furthermore, the model can be used to give insights into pressure dependent hydration. The pressure dependent hydration charac-

Table 4. Numerical values of the coefficients of Eq. (2) and the energy of activation of Eq. (5) for sucrose and glucose.  $h_0$  is calculated from Eq. (2)

	Sucrose	Glucose
$q_0^*$ [g/cm <sup>3</sup> ]	1.12	1.23
$E_a$ [kJ/mol]	2.61	2.76
$q_1$ [-]	-0.248	-0.238
$h_0$ [-]	1.62	1.62
$k_I$ [-]	2.57	2.50

teristics are not a subject of this work and are described in Först (2001).

The hydration factor at infinite dilution can be derived from  $q_0$  in Eq. (4). Assuming that the density  $D_s$  has only a weak temperature dependence and the shape factor  $k_I$  is temperature independent, it follows from Eqs. (4) and (5) that the activation energy  $E_a$  is mainly related to  $h_0$  and describes hydration changes with temperature. The numerical values of the coefficients  $q_0^*$  and  $q_1$  as well as  $E_a$  and  $h_0$  are shown in Table 4 for both glucose and sucrose solution. The shape factors  $k_I$  are also given. For sucrose the shape factor is given by Vand (1948b). The shape factor for glucose has been derived from the extrapolation of the reduced viscosity to  $x = 0$ .

## Conclusions

Measurement data for the pressure-dependence of the viscosity of sucrose solutions and the influence of temperature and concentration are presented. The data were obtained using a rolling ball viscometer with a maximum pressure of 700 MPa. Additionally, some data for glucose solutions were presented to point out the difference between the two sugars. These data allow a better understanding of the processes occurring at high pressures, e.g. thermofluidynamic processes.

In addition to the measurement data a viscosity model is introduced. The application of the model to sugar solutions shows that it can be used as a predictive model if the pressure dependence of the solvent viscosity and of the density of the solution are known. A pressure increase for constant mass fraction is equivalent to an increase in mass fraction  $w$  at constant pressure if referred to the same concentration  $x$ . The numerical values for the coefficients of the model equation can easily be determined with the ambient pressure data. These results suggest that there are no direct pressure induced changes in hydration, shape and intermolecular interactions occurring in the system. The results are in agreement with the exponential increase of the absolute viscosity with pressure which has also been observed for pure liquids that undergo no structural changes.

**Acknowledgments** The authors wish to thank the Deutschen Forschungsgemeinschaft (DFG) for support of the joint project Effect of high pressure on molecular and cellular systems in foods.

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