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Molecular mobility and the perceived sweetness of sucrose, fructose, and glucose solutions

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Abstract

The general relationship proposed in the literature between molecular mobility of water and the perceived sweetness was investigated. Sucrose, fructose, and glucose solutions, ranging in concentration from 0.15 mol/l to saturation were investigated. The rotational and translational mobilities of water and sugar molecules were monitored using a suite of nuclear magnetic resonance (NMR) techniques. The perceived sweetness was studied using the Time–Intensity (TI) method. Overall, it was found that the perceived sweetness of the sugar solutions, as quantified by the TI method, did not correlate with the measured NMR molecular mobility parameters.

Keywords: Sugars; Water; Mobility; Sweetness; Nuclear magnetic resonance (NMR) spectroscopy

1. Introduction

It is well recognized that relative sweetness varies widely among different sweeteners. An understanding of factors that affect the perceived sweetness is important in the development of new high-potency sweeteners. Mathlouthi and Seuvre (1988) explained the high sweetness intensity of 4,1′,6′-trichloro-4,1′,6′-trideoxygalactosucrose (TGS) in terms of the increased water mobility around the TGS molecule. The authors proposed that water molecules were repelled by the hydrophobic Cl substituents on the TGS molecule as indicated by a low intrinsic viscosity ([η]) and a high Huggins (k′) constant. This repulsion hinders the water molecules from competing with the TGS for the binding sites on the taste receptors (Mathlouthi, 1984). Mathlouthi and Seuvre (1988) proposed that the highly mobile water molecules could also disturb the iso-osmotic equilibrium between the saliva and blood serum on both sides of the receptor membrane. This results in a more active Na^+/K^+ transport across the membrane and an increase in perceived sweetness intensity.

Recently, Aroulmoji, Hutteau, Mathlouthi, and Rutledge (2001) investigated the role of water on the perceived taste of sucrose, caffeine, and sucrose–caffeine mixtures. The interaction between the sweet and bitter molecules with water was studied in terms of macroscopic (volumetric properties and surface properties) and molecular (NMR relaxation rates) methods. The authors suggested that these parameters (i.e., intrinsic viscosity, Huggins constant, apparent specific volume, hydration number, surface tension, contact angle, adhesion force, and NMR R_1 and R_2) can be used as a tool to elucidate the role of water in the perceived sweetness and bitterness.

Based on earlier reports by Mathlouthi and coworkers (Mathlouthi, 1984; Mathlouthi, Hutteau, & Angiboust, 1996; Mathlouthi & Seuvre, 1988), we hypothesized that, at equal molar concentrations, the higher the perceived sweetness of a sugar, the more mobile is the water associated with the sugar. The objectives of this study were to: (1) measure the rotational mobility of water molecules in three sugar solutions: sucrose, fructose, and glucose. The longitudinal relaxation rate (R_1), transverse relaxation rate (R_2), and

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effective transverse relaxation rate (R_2*) were measured using ^{1}H-decoupled ^{17}O NMR spectroscopy; (2) monitor the rotational mobility of sugar molecules in solution using ^{1}H NMR spectroscopy; (3) monitor the translational mobility of water molecules in the sugar solutions. Self-diffusion coefficient (D) of water was measured using a ^{1}H pulsed-field gradient spin-echo (PGSE) method; (4) monitor the translational mobility of the sugar molecules in solution by measuring the self-diffusion coefficient (D) of the non-exchangeable proton on the sugar molecule obtained by a ^{1}H NMR PGSE method; (5) measure the perceived sweetness of the sugar solutions using the time–intensity (TI) sensory evaluation technique; and (6) evaluate the relationship between the mobility of water and sugar molecules and the perceived sweetness of the sugar solutions.

2. Materials and methods

2.1. Materials

Reagent grade sucrose, fructose, and glucose were obtained from Sigma Chemicals (St. Louis, MO). The water used in this study was filtered drinking water obtained from Polar Water (Decatur, IL).

For NMR and sensory experiments, six concentrations of sucrose solution were prepared: 0.15, 0.30, 0.60, 0.90, 1.50 mol/l and saturated solution (2.60 mol/l at 25 °C).

For NMR experiments, eight concentrations of fructose solution were prepared: 0.15, 0.30, 0.60, 0.90, 1.50, 2.50, 3.50 mol/l and saturated solution (6.19 mol/l at 25 °C). However, due to the limitation in the number of samples that can be effectively evaluated by the panellists during each tasting session, only six concentrations of fructose solution were evaluated: 0.15, 0.30, 0.60, 1.50, 3.50, and 6.19 mol/l.

For NMR experiments, seven concentrations of glucose solution were prepared: 0.15, 0.30, 0.60, 0.90, 1.50, 2.00 mol/l and saturated solution (3.40 mol/l at 25 °C). Due to the limitation in the number of samples that can be effectively evaluated by the panellists during each tasting session, only six concentrations of glucose solution were evaluated: 0.15, 0.30, 0.60, 1.50, 2.00, and 3.40 mol/l.

2.2. ^{1}H-decoupled ^{17}O NMR

A UI-500WB NMR spectrometer (Varian Associates, Palo Alto, CA), operating at 67.8 MHz was used for the measurements of the rotational mobility of the water molecules, R_1, R_2, and R_2*. A multinuclear 10-mm probe was used. Samples were placed in a 10-mm NMR tube. The experiments were carried out at 25±1 °C. Two determinations of two separate replicates were done for each sample.

R_1 values were measured using the Inversion-Recovery (IR, 5T$_1$-180°-τ-90°) method (Vold, Waugh, Klein, & Phelps, 1968). The 90° pulse width was 11.75 μs and the τ delay values ranged from 0.00075 to 0.192 s.

R_2 values were measured using the Carr–Purceil–Meiboom–Gill (CPMG, 90°-(τ-180°-2τ-180°-τ)$_n$) method (Carr & Purcell, 1954; Meiboom & Gill, 1958). The 90° pulse width was 11.75 μs. The total time for transverse relaxation (i.e. 4n×τ) ranged from 0.0008 to 0.0096 s.

R_2* values were measured using a single pulse experiment. The 90° pulse width was 11.75 μs. The recycling time was 200 ms. The R_2* value was calculated from the line width at half-height ($\Delta\upsilon_{1/2}$) by the following formula (Dwek, 1973):

$$R_2{}^* = \pi\Delta\upsilon_{1/2} \quad (1)$$

2.3. ^{1}H NMR

A UI-500WB NMR spectrometer (Varian Associates, Palo Alto, CA), operating at 499.9 MHz was used for the measurements of R_1 and D values. A multinuclear 5-mm probe was used. Samples were placed in a 5-mm Ultem® plug (magnetic susceptibility, χ, of −0.71) which was fit into a 5-mm NMR tube. The experiments were carried out at 25±1 °C. Two determinations of two separate replicates were done for each sample.

2.3.1. Rotational mobility of sugar molecules

R_1 values were measured using the IR method (Void et al., 1968). The 90° pulse width was 6.625 μs and the τ delay values ranged from 0.10 to 0.45 s.

2.3.2. Translational mobility of water molecules

The D values for water in the sugar solutions were measured using the pulsed-field gradient spin-echo (PGSE) method (Tanner & Stejskal, 1968). The 90° pulse width was 6.625 μs. A set of 16 gradient amplitudes (G) with a duration (δ) of 2.0 ms were used in this study. The time interval (Δ) was 21.41 ms.

2.3.3. Translational mobility of sugar molecules

D value of the sugar molecules was measured using the PGSE method (Tanner & Stejskal, 1968). The 90° pulse width was 6.625 μs. A set of 16 G with δ of 2.5 ms were used in this study. The Δ was 32.91 ms.

2.4. Time–Intensity (TI) sweetness measurements

The sensory panel consisted of 13 University of Illinois students. Each sugar was evaluated separately. The sensory experiment was divided into two phases: training and sample tasting.

For the training phase, the panellists were first trained to rate sweetness intensity on a 15-line scale anchored from "none" to "extreme." One cm of the 15-cm line scale represented one sweetness intensity unit. The reference solutions used in the training were filtered water, 0.15 moles/l glucose solution, 0.15 mol/l sucrose solution, 0.30 mol/l sucrose solution, 0.60 mol/l sucrose solution, 0.90 mol/l sucrose solution, 1.50 mol/l sucrose solution, 2.60 mol/l sucrose solution, and 6.19 mol/ fructose solution. These reference solutions were chosen to cover the sweetness intensity range from 0 to 15 cm. Samples (15 ml) were served at room temperature (25±2 °C) in odour-free plastic cups. Filtered water (Polar Water, Decatur, IL) and unsalted-top crackers (Meijer, Grand Rapid, MI) were used to clear the palate. The panellists were instructed to pour the entire sample into their mouths and hold the sample until they were prompted to expectorate the sample at 12 s. The panellists immediately recorded sweetness intensity on

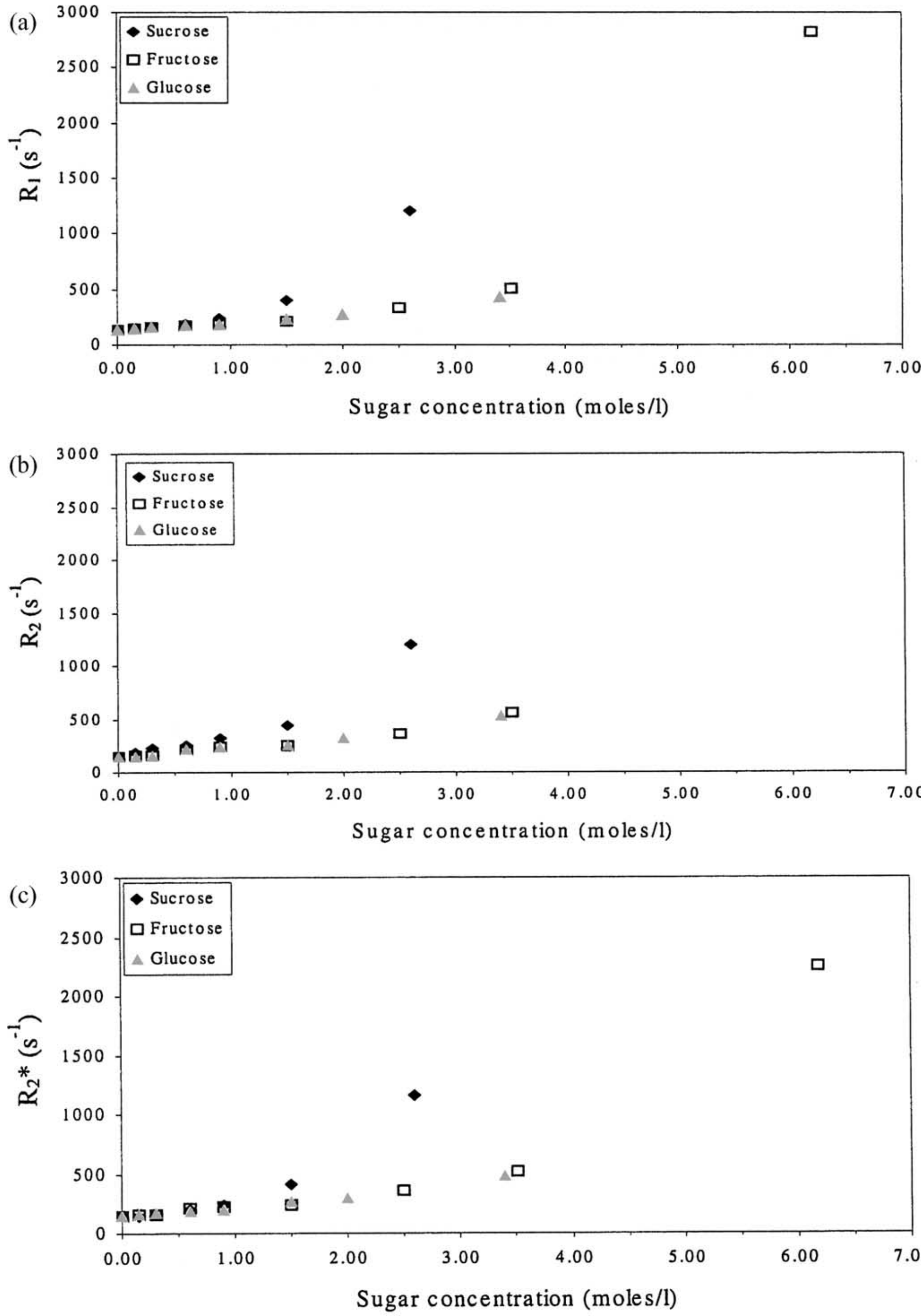

Fig. 1. ^{1}H-decoupled ^{17}O NMR (a) R_1, (b) R_2, and (c) R_2* of water in sucrose, fructose, and glucose solutions vs. sugar concentration (mol/l). Due to an exceptionally weak ^{17}O signal, the R_2 value for the saturated fructose solution was not obtained.

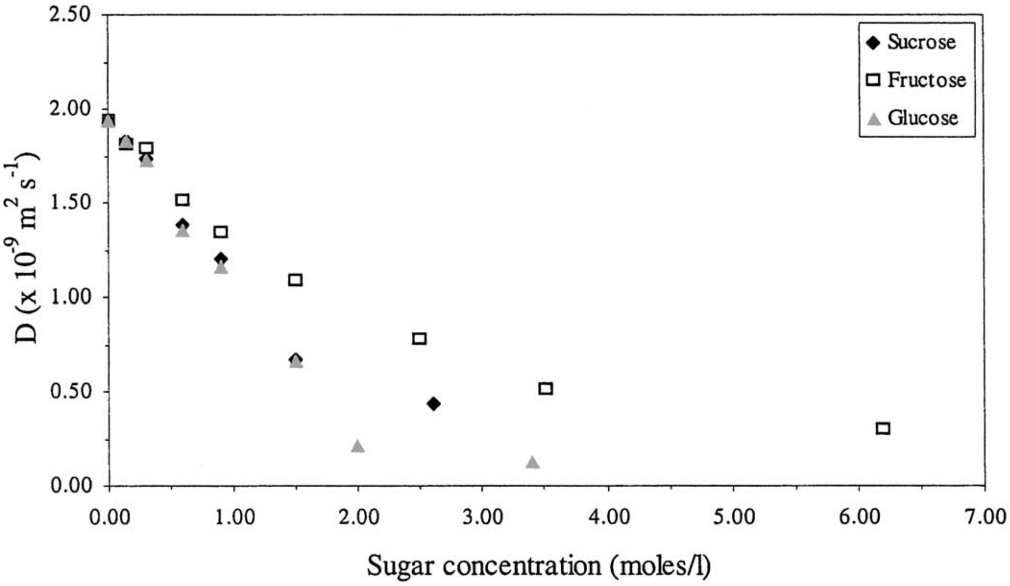

Fig. 2. Self-diffusion coefficient (*D*) of water in sucrose, fructose, and glucose solutions vs. sugar concentration (mol/l).

the 15-cm line scale. After rinsing their mouths, the panellists were instructed to wait 3 min, or until the taste sensations subsided, before evaluating the next sample. The panellists then participated in training sessions where they evaluated sweetness intensity of unknown solutions relative to the reference solutions. Lastly, the panellists were trained for the TI measurement procedure. The sweetness intensity of a sample was recorded as a function of time. The panellists were instructed to take the sample at 0 s and expectorate the sample at 12 s. The perceived sweetness intensity was recorded on each 15-cm line scale at 7, 12 (immediately after expectoration), 20, 30, 45, 60, 90, 120, 150 and 180 s. Panellists' performance was monitored and additional training was provided to improve accuracy.

For sample tasting, the panellists tasted the samples in individual booths under fluorescent light and at room temperature (25±2 °C). The CompuSense Paper® computer program (CompuSense, Ontario, Canada) was used to generate the TI score sheet, randomize the sample presentation order, and compile the data. Randomized block design was used for the experiment. The panellists evaluated each sample three times. Six samples were evaluated in each session. The data were analyzed using analysis of variance (ANOVA) using the SAS program (SAS Institute, Cary, NC). A Least Significance Difference test (95% confidence level) was used to determine whether the sweetness intensity at a particular time point differed among the samples.

The average sweetness intensity scores for each sample were used to plot a TI curve (sweetness intensity vs. time). Nine parameters, related to the onset, perceived sweetness and aftertaste, were obtained from the TI curves. These parameters are: maximum sweetness intensity (I_{max}), time to reach the maximum sweetness intensity (T_{max}), total sweetness duration (T_{tot}), rate of adsorption (M_{ads}), rate of desorption (M_{des}), area under the curve from onset to expectoration (AUC_{stim}), area under the curve after expectoration (AUC_{aft}), total area under the TI curve (AUC_{tot}), and aftertaste (AT). The definition of these TI parameters can be found in Ketelsen, Keay, and Wiet (1993).

3. Results and discussion

3.1. Rotational mobility of water molecules

The rotational mobility of water molecules was monitored by ^{1}H-decoupled ^{17}O NMR. The ^{1}H-decoupled ^{17}O NMR R_1, R_2, and R_2* of water as a function of solution concentration is shown in Fig. 1. At a low concentration range ($\leqslant$0.90 mol/l), the R_1, R_2 and R_2* values gradually increased (decreasing water mobility) with increasing sugar concentration. Padua and Schmidt (1992) studied the rotational mobility of water in sucrose, fructose, and glucose solutions using ^{1}H NMR R_1. They reported that the rotational mobilities in the three sugar solutions were similar up to a concentration of 0.7 g sugar/g water (1.42 mol/l for sucrose solution, and 2.70 mol/l for fructose and glucose solutions). Padua and Schmidt (1992) explained the linear increase in relaxation rate with increasing solution concentration at the low concentration range in terms of the two-state model with fast exchange (Zimmerman & Brittin, 1957).

At concentrations higher than 0.90 mol/l, the relationship between the R_1, R_2 and R_2* vs. solution concentration deviated from linearity. Derbyshire (1982) proposed that this departure from linearity was due to the changes in the hydration of the solute molecule and the changes in the relaxation rate of the bound water. This departure from linearity was also reported by other researchers for a variety of sugar solutions (Lai, 1990; Lai & Schmidt, 1990; Padua & Schmidt, 1992; Richardson, Baianu, & Steinberg, 1987). In this study it was found that, at the same molar concentration, the sucrose solution exhibited higher relaxation rates (lower water mobility) than the fructose and glucose solutions. Mora-Gutierrez and Baianu (1989) suggested that the heavier sucrose molecule tumbles more slowly than the fructose and glucose molecules and this could increase the correlation time of the bound water associated with the sucrose molecule.

Mahawanich (2000) reported that there was a linear correlation between log R_1, R_2, and R_2* values of water and log viscosity. The correlations between log R_1, R_2, and R_2* values and log viscosity were similar between the fructose and glucose solutions, but different from that of the sucrose solutions. Padua (1989) also observed a similar trend between log ^{1}H NMR R_1 values of water and log viscosity of sucrose, fructose, and glucose solutions. Padua and Schmidt (1992) suggested that the increased in viscosity is due to the aggregation of the sugar molecules into clusters of slower tumbling motion, causing a decrease in mobility of the water bound to the sugar molecules.

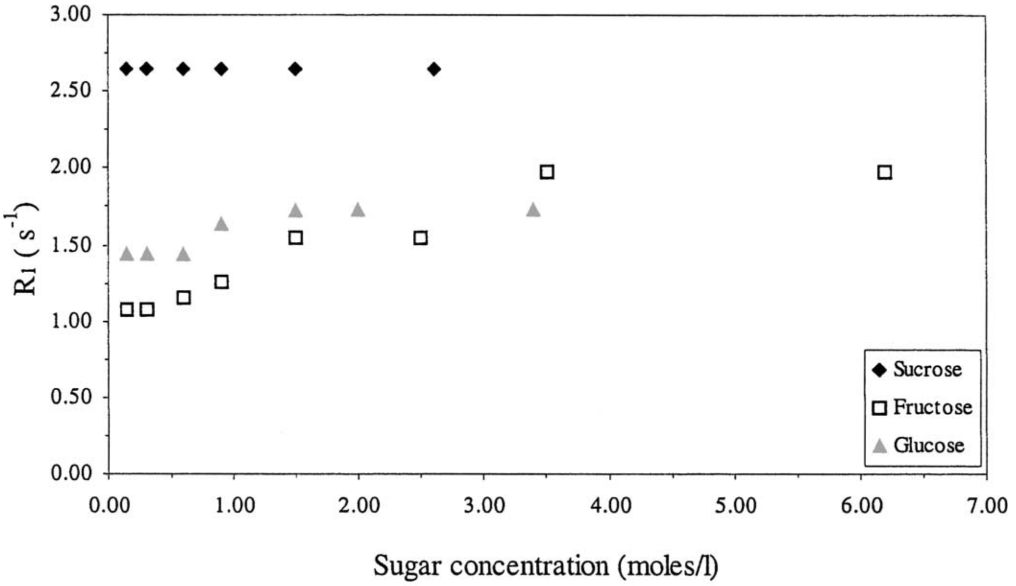

Fig. 3. ^{1}H NMR R_1 values of sucrose, fructose, and glucose molecules in solution vs. sugar concentration (mol/l).

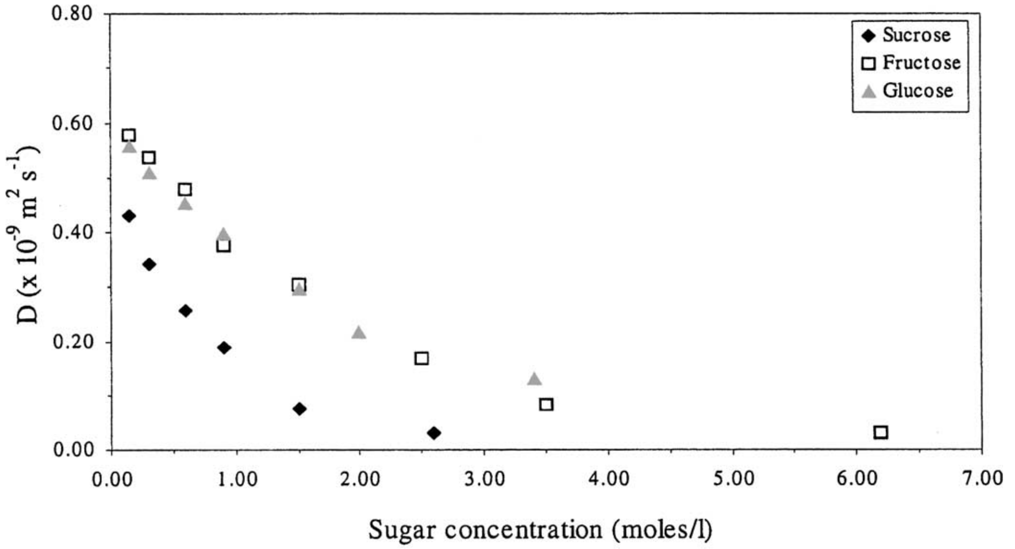

Fig. 4. Self-diffusion coefficient (D) of sucrose, fructose, and glucose molecules in solution vs. sugar concentration (mol/l).

3.2. Translational mobility of water molecules

The translational mobility of water in the sugar solutions, as monitored by the proton self-diffusion coefficient (*D*), is shown in Fig. 2. In contrast to the rotational mobility of water, fructose solutions had the highest translational water mobility (fastest moving) among the three sugar solutions, followed by sucrose and glucose solutions, respectively. Mathlouthi and Seuvre (1988) measured the intrinsic viscosity, [η], of monosaccharides (fructose, glucose, galactose), disaccharides (sucrose, lactose, maltose), and TGS. The [η] is a shape factor accounting for the hydrodynamic radius of the solvated molecule (Shamil, Birch, Mathlouthi, &

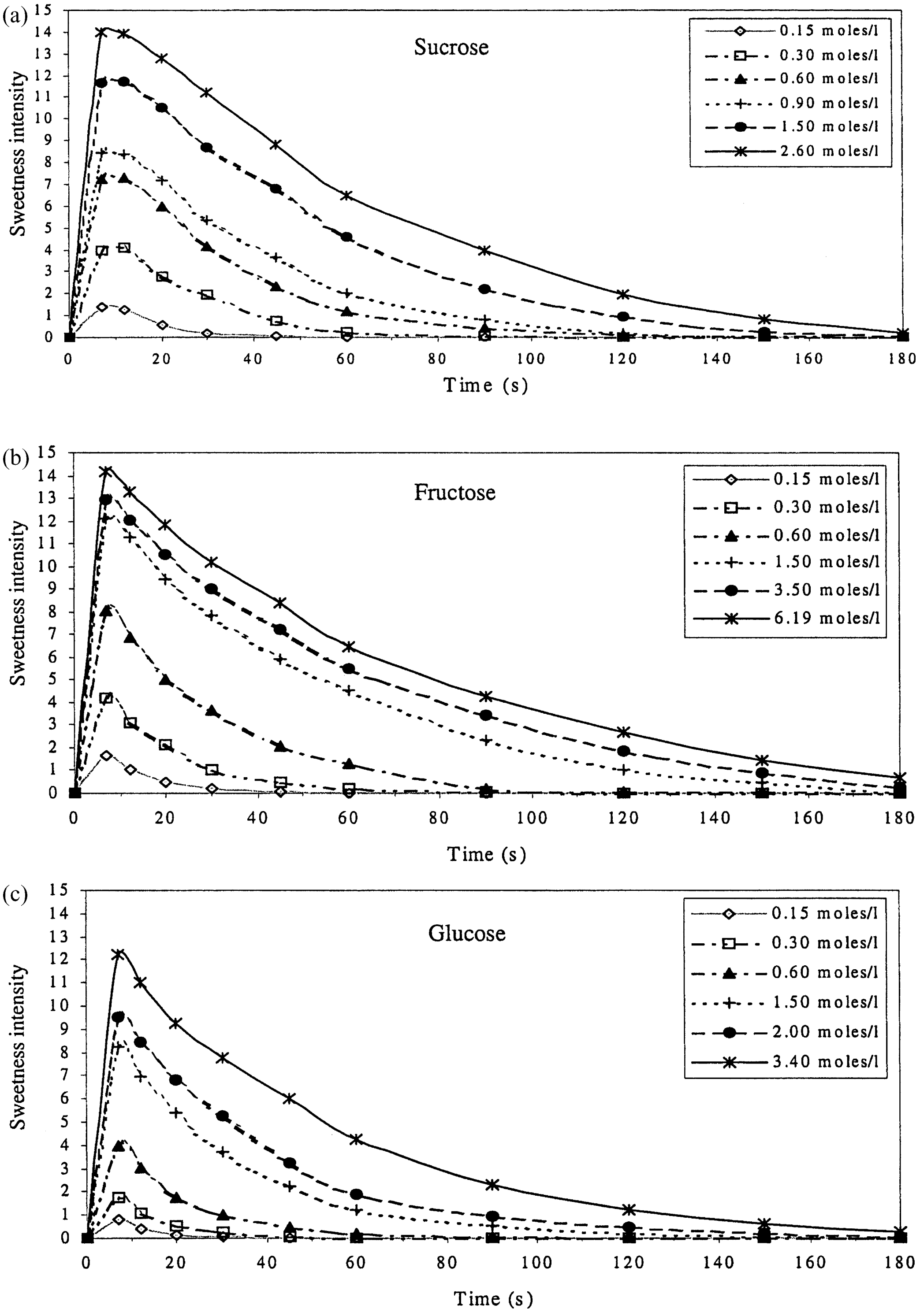

Fig. 5. Time–intensity (TI) plots of (a) sucrose, (b) fructose, and (c) glucose solutions.

Clifford, 1987) and it can be used to investigate the interaction between solute and water molecules. Mathlouthi and Seuvre (1988) reported that all the monosaccharides (except fructose) and the disaccharides bad similar [η] values. They attributed this to the hydrophilic nature and the quasi-spherical shape of the hydrated sugar molecules. Fructose, on the other hand, had relatively low [η] value. TGS, which is 2000 times sweeter than sucrose, had a remarkably low [η] value. The authors proposed that the low [η] values of fructose and TGS were due to the marked difference between the effects of the hydrophilic and hydrophobic sides of the molecules. The hydrophobic side repels water, resulting in a higher water mobility around the sweetener molecules.

3.3. Rotational mobility of sugar molecules

The rotational mobility of the sugar molecules in aqueous solution was monitored by ^{1}H NMR R_1. ^{1}H NMR R_1, of sugar molecules in solution, as a function of solution concentration, is shown in Fig. 3. The sucrose molecules exhibited the slowest rotational mobility (highest R_1) among the three sugars. Fructose molecules had a slightly higher rotational mobility (lower R_1) than glucose molecules. Birch and Karim (1992) measured the ^{1}H NMR R_2 values of glucose solutions and glucose syrups (5–50%, w/w). The degree of polymerization (DP) of the glucose syrups ranged from 1.6 to 8.3. It was found that for each glucose syrup

Table 1
Time–intensity (TI) parameters of sucrose solutions

TI parameters	Sucrose concentration (mol/l)[a]					
	0.15	0.30	0.60	0.90	1.50	2.60
I_{max}	1.45	4.14	7.45	8.55	11.83	14.21
T_{max}	9.0 s	9.0 s	9.0 s	9.0 s	9.0 s	9.0 s
T_{tot}	45 s	90 s	150 s	150 s	180 s	> 180 s
M_{ads}	0.171	0.488	0.882	1.016	1.406	1.688
M_{des}	−0.039	−0.098	−0.149	−0.142	−0.146	−0.151
AUC_{stim}	4.865	13.895	25.375	29.505	40.775	48.965
AUC_{aft}	20.130	104.735	252.510	339.740	617.675	863.230
AUC_{tot}	24.995	118.630	277.885	369.245	658.450	912.195
AT	4.138	7.538	9.951	11.515	15.148	17.630

I_{max} = Maximum sweetness intensity (intensity units, NONE = 0 and EXTREME = 15). T_{max} = Time to maximum sweetness intensity (s). T_{tot} = Total sweetness duration (s). M_{ads} = Rate of adsorption (positive slope) (intensity units/s). M_{des} = Rate of desorption (negative slope) (intensity units/s). AUC_{stim} = Area under the curve from onset to expectoration (intensity units × s). AUC_{aft} = Area under the curve after expectoration (intensity units × s). AUC_{tot} = Total area under the TI curve (intensity units × s). AT = Aftertaste (AUC_{aft}/AUC_{stim}).

[a] Average values for $n = 13$ panel members.

Table 2
Time–intensity (TI) parameters of fructose solutions

TI parameters	Fructose concentration (mol/l)[a]					
	0.15	0.30	0.60	1.50	3.50	6.19
I_{max}	1.64	4.35	8.31	12.27	13.09	14.32
T_{max}	7.7s	7.7s	7.7s	7.7s	7.7s	7.7s
T_{tot}	45 s	90 s	150 s	180 s	> 180 s	> 180 s
M_{ads}	0.233	0.601	1.151	1.731	1.851	2.026
M_{des}	−0.060	−0.133	−0.157	−0.164	−0.151	−0.153
AUC_{stim}	5.705	14.735	28.210	42.420	45.360	49.630
AUC_{aft}	20.415	72.233	226.270	593.795	740.435	889.260
AUC_{tot}	26.120	86.968	254.480	636.215	785.795	938.890
AT	3.578	4.902	8.021	13.998	16.324	17.918

I_{max} = Maximum sweetness intensity (intensity units, NONE = 0 and EXTREME = 15). T_{max} = Time to maximum sweetness intensity (s). T_{tot} = Total sweetness duration (s). M_{ads} = Rate of adsorption (positive slope) (intensity units/s). M_{des} = Rate of desorption (negative slope) (intensity units/s). AUC_{stim} = Area under the curve from Onset to expectoration (intensity units × s). AUC_{aft} = Area under the curve after expectoration (intensity units × s). AUC_{tot} = Total area under the TI curve (intensity units × s). AT = Aftertaste (AUC_{aft}/AUC_{stim}).

[a] Average values for $n = 13$ panel members.

Table 3
Time–intensity (TI) parameters of glucose solutions

TI parameters	Glucose concentration (mol/l)[a]					
	0.15	0.30	0.60-	1.50	2.00	3.40
I_{max}	0.80	1.80	4.16	8.40	9.73	12.43
T_{max}	7.7s	7.7s	7.7s	7.7s	7.7s	7.7s
T_{tot}	30 s	45 s	90 s	150 s	180 s	> 180 s
M_{ads}	0.113	0.249	0.573	1.176	1.359	1.744
M_{des}	−0.047	−0.063	−0.130	−0.157	−0.164	−0.161
AUC_{stim}	2.765	6.090	14.035	28.805	33.285	42.735
AUC_{aft}	6.360	20.550	72.225	247.825	345.630	597.215
AUC_{tot}	9.125	26.640	86.260	276.630	378.915	639.950
AT	2.300	3.374	5.146	8.604	10.384	13.975

I_{max} = Maximum sweetness intensity (intensity units, NONE = 0 and EXTREME = 15). T_{max} = Time to maximum sweetness intensity (s). T_{tot} = Total sweetness duration (s). M_{ads} = Rate of adsorption (positive slope) (intensity units/s). M_{des} Rate of desorption (negative slope) (intensity units (s). AUC_{stim} = Area under the curve from onset to expectoration (intensity units × s). AUC_{aft} = Area under the curve after expectoration (intensity units × s). AUC_{tot} = Total area under the TI curve (intensity units × s). AT = Aftertaste (AUC_{aft}/AUC_{tot}).

[a] Average values for n = 13 panel members

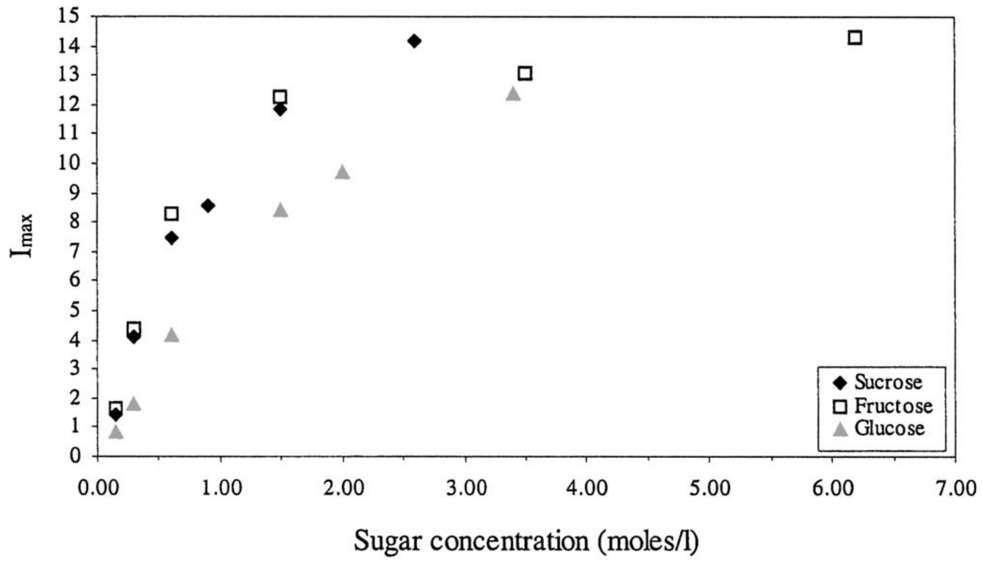

Fig. 6. I_{max} as a function of sugar concentration (mol/l).

(i.e., each DP), the R_2 value increased (decreasing molecular mobility) with increasing concentration. At the same concentration, the ^{1}H NMR R_2 increased (decreasing molecular mobility) with increasing DP. The authors proposed that the larger molecules (i.e., higher DP) had a slower molecular tumbling. The findings in the current study also support this point of view. Sucrose (molecular weight of 342.30) showed a slower rotational mobility than the lighter fructose and glucose molecules (molecular weight of 180.16 each).

3.4. Translational mobility of sugar molecules

The translational mobility of the sugar molecules in aqueous solution, as monitored by proton self-diffusion coefficient (D), is shown in Fig. 4. Similar to the rotational mobility, sucrose, which is a heavier molecule, showed a slower translational mobility (lower D value) than the lower molecular weight fructose and glucose molecules.

3.5. Time–Intensity (TI) sweetness measurement

The TI plots of the sucrose, fructose, and glucose solutions are shown in Fig. 5. The nine parameters obtained from the TI curves are defined and shown in Tables 1, 2, and 3 for sucrose, fructose, and glucose, respectively. Fig. 6 shows a plot of I_{max} vs. sugar concentration.

For each sugar, the maximum intensity (I_{max}) and total duration of sweetness (T_{tot}) increased as the sugar concentration increased. This phenomenon was also

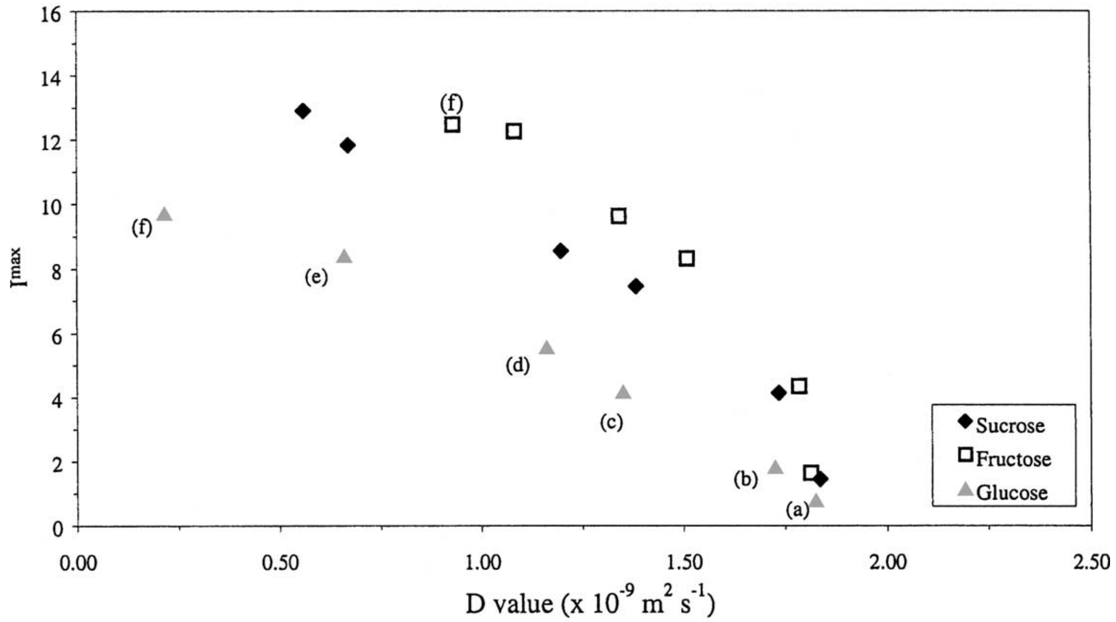

Fig. 7. I_{max} as a function of D values of water in sucrose, fructose, and glucose solutions. Equal molar concentrations are grouped by the following letters: (a) 0.15, (b) 0.30, (c) 0.60, (d) 0.90, (e) 1.50, (f) 2.00 mol/l.

observed in earlier studies by Swartz (1980), Birch and Munton (1981), Dubois and Lee (1983), and Cliff and Noble (1990). Each sugar had the same T_{max} for all concentrations studied. The M_{ads}, AUC_{stim}, AUC_{aft}, AUC_{tot} and AT increased with increasing sugar concentration. At lower concentrations ($\leqslant$0.60 mol/l for sucrose solution, $\leqslant$0.60 mol/l for fructose solution, $\leqslant$1.50 mol/l for glucose solution), M_{des} increased with increasing sugar concentration. However, at higher concentrations M_{des} became similar among different sugar concentrations.

At the same concentration, the three sugars had different I_{max} values. Fructose solutions had the highest I_{max}, followed by sucrose and glucose solutions, respectively. All fructose and glucose solutions had T_{max} at 7.7 s while the sucrose solutions had T_{max} at 9.0 s. Ideally, comparison of TI parameters are most meaningful when done among solutions with equal sweetness intensity since the total duration (T_{tot}, AUC_{tot}), onset characteristics (M_{ads}, AUC_{stim}), and aftertaste characteristics (M_{des}, AUC_{aft}, AT) are functions of I_{max} (Ketelsen et al., 1993).

Sugar solutions with similar I_{max} exhibited similar T_{max} and T_{tot}. The sucrose solution had a slightly lower M_{ads} and slightly higher AUC_{aft}, AUC_{tot} and AT than the other two sugars. M_{des} and AUC_{stim} were comparable among the sugar solutions (data not shown; see Mahawanich, 2000). Similar onset and aftertaste characteristics among equisweet solutions were also reported for sucrose and fructose by Ketelsen et al. (1993).

Because the sugar solutions had different I_{max} and the T_{tot}, AUC_{tot}, M_{ads}, AUC_{stim}, M_{des}, AUC_{aft}, AT are a function of I_{max}, the I_{max} was selected to be the most suitable parameter for comparing the perceived sweetness with the NMR molecular mobility parameters.

3.6. Molecular mobility and the perceived sweetness

According to Shallenberger and Acree's sweet unit model (Shallenberger & Acree, 1967), sweetness originates in an AH/B glucophore on the sweet molecule. This AH/B glucophore couples with another AH/B unit on the receptor site of the taste bud to elicit a sweet taste sensation. Kier (1972) proposed that a third hydrophobic X site plays a role in amplifying the sweetness intensity. Van der Heijden (1993) reported that derivatives of galactosucrose with chlorine substitution at position 4, 1′, 4′, or 6′ are all intensely sweet. This is mainly because these positions are located on the side of the molecule opposite to the hydrophilic moiety (the AH/B system). Water molecules are repelled by the hydrophobic side. According to Sybesma (1977), upon its dissociation, this highly mobile water is believed to help facilitate the Na^+/K^+ transport across the membrane of the tongue, increase the membrane potential, and thus enhance the perceived sweetness intensity.

In this study, it was found that the only NMR parameter that exhibited the same trend as I_{max} (fructose > sucrose > glucose) was the translational mobility of the water molecule, as monitored by the proton self-diffusion coefficient (D). In general, all of the other NMR parameters (rotational mobility of water, and rotational and translational mobility of sugar molecules) exhibited the trend that glucose was similar to fructose and both exhibited a higher mobility than sucrose (glucose≈fructose > sucrose), as previously discussed under the various NMR results and discussions sections.

Fig. 7 shows a plot of I_{max} as a function of the D values of water. Equal molar concentrations, in Fig. 7, are grouped by the following letters: (a) 0.15, (b) 0.30, (c) 0.60, (d) 0.90, (e) 1.50, and (f) 2.00 mol/l. Even though the translational mobility of water and the I_{max} of the sugar solutions showed the same trend, the plot of I_{max} vs. D values of water shows that the relationship between the I_{max} and D values of water was sugar-dependent. There appears to be no overall correlation between I_{max} and D values of water.

Mathlouthi (1984) proposed that the hydrophobic side of the fructose molecule repels water molecules, resulting in a higher water mobility. Mathlouthi and Seuvre (1988) studied the effect of traces amounts of sucrose, fructose, and glucose (10^{-6}, 10^{-4}, and 10^{-2} g sugar/g water) on the structure of water using laser Raman spectroscopy. They reported that fructose caused an increase in the vibrational energy of the water while sucrose and glucose did not cause a noticeable change in the Raman spectrum of water. This supports their hypothesis that fructose has a breaking effect on the water structure.

To investigate the hydrophobicity of sweeteners (sucrose, fructose, glucose, and TGS), Mathlouthi and Hutteau (1999) measured the adhesion force (W_{ls}) of the sweetener solutions to the hydrophobic (polyethylene) surface. It was proposed that this interfacial property might help explain the diffusion of a sweet molecule onto the hydrophobic surface of the receptor site. In an earlier study, Hutteau and Mathlouthi (1998) reported that there was a high correlation between the W_{ls} value and the perceived sweetness. A molecule with higher published relative sweetness value exhibited a higher W_{ls} value.

4. Conclusion

Contrary to the proposed hypothesis that at equal molar concentrations, the higher the perceived sweetness of a sugar, the more mobile is the water associated with the sugar, there appears to be no direct correlation between the perceived sweetness and the NMR molecular mobility parameters measured in this study. Perhaps the lack of direct correlation between the perceived sweetness and the NMR molecular mobility parameters is due to the complex nature of sweet taste perception and the challenges associated with correlating molecular level parameters with macroscopic sensory data.

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