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## Intrinsic viscosities and other solution properties of sugars and their possible relation to sweetness

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**Abstract.** The intrinsic viscosities, apparent molar volumes, apparent specific volumes and apparent solution parachors of selected sugars were determined with the aim of investigating the possible relationship of these solution properties to the published sweetness values of sugars. For a given sugar, there is no clear relationship between its intrinsic viscosity and other solution properties studied. The values of apparent specific volume and intrinsic viscosity differ greatly even though both parameters reflect the amount of water surrounding 1 g of the solute. There is also no observable correlation between the intrinsic viscosities and the literature values on the relative sweetness of sugars. The intrinsic viscosity of a solute molecule reflects its extent of hydration. It is greatly influenced by the conformation and configuration of the sugar molecules. Equatorial OH groups are more favourable for hydration than the axial OH groups. Thus, the more OH groups in the equatorial position, the higher the intrinsic viscosity.

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### Introduction

Sugar is added to foods because it imparts sweetness and confers desirable textural and rheological properties and also because it acts as a preservative by reducing water activity.

Many attempts have been made to explain the molecular basis of sweetness (Shallenberger and Acree, 1967; Birch, 1976) but much of the work has centred on the stereochemistry of sweeteners, with little emphasis on their physical behaviour in water. This, however, has recently been investigated by Birch and Catsoulis (1985), Birch and Shamil (1986), Shamil *et al.* (1987), Mathlouthi (1984) and Mathlouthi *et al.* (1986). These studies have indicated that the role of water in taste response is important because most insoluble substances are tasteless. The interactions of sapid solutes with water may perhaps influence their accession to, and binding with, receptor sites, thus influencing their taste properties.

Solution properties offer a convenient means of investigating the behaviour of solutes in water. If all taste effects are mediated by water, the solution properties of sugars may provide the most immediate understanding of their potential sapid effects.

This study concentrates on intrinsic viscosities and related solution properties of selected sugars with the objective of investigating whether there exists a relationship between these properties and sweet taste.

### Materials and methods

Sugars investigated in this work were reagent grade chemicals obtained from BDH Chemicals, Poole, Dorset, and The Sigma Chemical Co., Poole, Dorset. D-Glucose was obtained from May and Baker Ltd, Dagenham, Essex, and sucrose was AR grade (Fisons, Loughborough, Leics.).

Water used was 'Hyper Solv' water for HPLC from BDH Chemicals, Poole, Dorset. Reducing sugars were allowed 30 h in the refrigerator (5°C) after dissolution to reach mutarotational equilibrium.

The intrinsic viscosities of sugar solutions were calculated from their relative viscosities. For each sugar, the relative viscosity was determined at several concentrations (between 5 and 10% w/v) using an 'A' type Ostwald's ('U'-shaped) glass capillary viscometer. The measurements were carried out at a constant temperature of 20°C which was achieved by a thermostatically controlled water bath under continuous stirring. The relative viscosities were calculated from the time taken for the test solutions and water to flow through the capillary viscometer and from their densities. The densities were measured on a Parr Densitometer as described by Birch and Catsoulis (1985). The equation used for calculating the relative viscosity, ( $\eta_{rel}$ ), which is the viscosity of test solution over the viscosity of water, is:

$$\eta_{rel} = \frac{d_1 t_1}{d_2 t_2}$$

where  $d_1$  = density of solution,  $d_2$  = density of water,  $t_1$  = time of flow of the solution through the viscometer and  $t_2$  = time of flow of water through the viscometer.

The intrinsic viscosity,  $[\eta]$  (dl/g), was obtained from the plot of reduced specific viscosity against solute concentration to infinite dilution (Barrow, 1966).

$$[\eta] = \lim_{c \rightarrow 0} \left[ \frac{1}{c} (\eta_{rel} - 1) \right]$$

where  $\eta_{rel} - 1 =$  specific viscosity ( $\eta_{sp}$ ), and  $\frac{\eta_{sp}}{c} =$  reduced specific viscosity (in dl/g) ( $c$  = solute concentration in g/dl).

In dilute solutions the solute molecules have virtually no interactions between them. There is only solute-solvent interaction and according to the Huggin's (1942) equation:

$$\frac{\eta_{sp}}{c} = [\eta] + k' [\eta]^2 c$$

A plot of reduced specific viscosity against solute concentration gives a straight line where  $k' [\eta]^2$  is the slope ( $k'$  is the interaction coefficient). Extrapolation of the linear plot to zero concentration gives the intrinsic viscosity,  $[\eta]$ , of the substance.

Apparent molar volumes ( $\phi V$ ) were calculated from densities determined with an Anton-Parr Precision Density Meter (DMA 60) and Density Measuring Cell (DMA 602) (Stanton Redcroft, London) equipped with an automatic sampler (SP2) and Anadex printer. Temperature control was achieved with a Hetofrig bath (Heto Birkerod, Denmark) coupled to the density measuring cell. The density meter was calibrated with air and water and the method was as previously described by Birch and Catsoulis (1985). Measurements were carried out at 20°C ( $\pm 0.1^\circ\text{C}$ ). Uncertainty in the density determinations was  $\pm 3.0 \times 10^{-6}$  g/cm<sup>3</sup>.

The surface tensions ( $\gamma$ ) of sugars were determined with a Du Nouy Tensiometer (Cambridge Instrument Co., London), which is a torsion balance method in which a

platinum loop is dipped into the liquid and the force required to separate the loop from the surface is measured directly on a calibrated scale.

The apparent solution parachors [P] were calculated from the surface tensions and apparent molar volumes ( $[P] = \phi V \gamma^k$ ).

## Results and discussion

Apparent molar and apparent specific volumes give direct measurement of displacement or disturbance of water by solutes and thus reflect compatibility of solutes with water structure. Apparent solution parachors represent the apparent molar volume ( $\phi V$ ) of a substance if its surface tension were to remain at unity. Intrinsic viscosity is a measure of the hydro-dynamic volume of the solute molecules and represents the fractional change in the viscosity of a solution per unit concentration of solute at infinite dilution (Elfak *et al.*, 1977). It is an important property that may help to explain the disturbance of water structure as the sapid molecules 'tumble' through the aqueous media in their passage to receptors. Table I lists these solution properties and the published relative sweetness values of 15 sweeteners.

Table I shows no clear trend between the intrinsic viscosities and the reported relative sweetness values of the molecules. This may, however, be due to the literature values on relative sweetness being greatly dependent upon many variables (i.e. purity of compounds tested, concentration range used, psychophysical method employed, etc). Pangborn *et al.* (1973), studying the relationships between viscosity and taste intensity of hydrocolloids, found the taste intensity to be independent of viscosity at low con-

Table I. Solution properties and relative sweetness of sugars.

Sugar	Mol. wt	Apparent molar volume <sup>a</sup> ( $\phi V$ ) (cm <sup>3</sup> /mol)	Apparent specific volume <sup>a</sup> ( $\phi V$ /Mol. wt) (cm <sup>3</sup> /g)	Apparent solution parachor <sup>a</sup> [P]	Intrinsic viscosity [ $\eta$ ] (dl/g) (or $\times 10^2$ cm <sup>3</sup> /g)	Relative sweetness <sup>b</sup>
D-Mannose	180.16	110.8	0.6150	322	0.0165	—
D-Arabinose	150.13	91.35	0.6085	266	0.0170	—
L-Arabinose	150.13	91.84	0.6117	267	0.0171	—
D-Galactose	180.16	109.0	0.6050	316	0.0196	0.6
D-Glucitol	182.18	116.2	0.6378	338	0.0197	0.5–0.6
Glycerol	92.11	70.56	0.7660	201	0.0205	0.8
D-Fructose	180.16	107.3	0.5956	311	0.0222	1.4–1.7
D-Xylose	150.13	93.80	0.6248	268	0.0230	0.7
Lactose	342.30	210.5	0.6150	608	0.0241	0.1–0.2
Sucrose	342.30	208.3	0.6085	604	0.0241	1.0
D-Mannitol	182.18	117.4	0.6444	341	0.0243	0.5–0.6
D-Glucose	180.16	110.8	0.6150	321	0.0247	0.6–0.7
Xylitol	152.15	100.5	0.6605	292	0.0253	0.6–0.7
Palatinose	342.30	219.5	0.6413	632	0.0272	0.4
Maltose	342.30	209.4	0.6117	607	0.0296	0.3–0.4

<sup>a</sup>Values obtained at 3% w/w concentration.

<sup>b</sup>Values obtained from Koivistoinen and Hyvönen (1980), Solms (1971) and other literature sources.

centrations, whereas at high concentrations an increase in viscosity decreased taste intensity. Arabie and Moskowitz (1971) also concluded the taste intensity to be a function of two simultaneously acting variables: concentration of solute and its viscosity.

There appears to be no clear relationship between  $[\eta]$  and the molecular weight of sugars. The disaccharides, however, generally have higher values than the monosaccharides. The sugar alcohols might be expected to have higher intrinsic viscosities than their parent sugars. This is observed in D-mannitol and xylitol, although D-glucitol behaves differently. This might be due to its conformation (Franks, 1987). The high intrinsic viscosities of polyols may be due to their straight chain structures causing a greater resistance to flow than the parent cyclic structures. The presence of a 'bulky' primary alcohol group in a sugar molecule causes a notably higher  $[\eta]$  as observed in the cases of D-glucose/D-xylose, D-galactose/L-arabinose and D-fructose/D-arabinose.

There appears to be a correlation between  $[\eta]$  and the extent of hydration of solute molecules. D-Glucose, containing more equatorial OH groups (favourable for hydration) than D-galactose or D-mannose, is more hydrated (Shamil, 1987) and has the highest intrinsic viscosity of all the monosaccharides tested (Table I). The influence of the position of OH groups on  $[\eta]$  of sugar molecules is further noted in D-xylose and L-arabinose. The much higher  $[\eta]$  of D-xylose (Table I) may be due to the equatorial location of its OH group at C<sub>4</sub>. Maltose, with the highest number of equatorial OH groups, has the highest intrinsic viscosity of all the sugars determined.

The results shown in Table I suggest that  $[\eta]$  is a measure of the extent of hydration of solute molecules in motion;  $[\eta]$  is influenced by the conformation and configuration of the solute molecules in solution. When the molecules are heavily hydrated or exist in open chain forms they experience a greater drag force and hence an increased resistance to flow, which is reflected by a higher  $[\eta]$ .

Table I shows a steady increase in  $\phi V$  and  $[P]$  as the molecular weight of sugars increases. A similar trend was also observed by Birch and Catsoulis (1985). The  $[\eta]$  and  $\phi V$  results cannot be directly compared because they have different units. However, a comparison of the intrinsic viscosity and the apparent specific volume for the same sugar shows a great difference even though both parameters reflect the volume of solvent associated with 1 g of the solute. For each sugar, the  $[\eta]$  value is about two to four times as high as the apparent specific volume. This may perhaps be due to the dynamic status of the solute molecules in solution, being in a stationary status in the case of apparent specific volume determination, but in a tumbling motion in the case of viscosity measurements.

There appears to be no direct correlation between intrinsic viscosity and the other solution properties studied. There is also no observable relation between these solution properties and the published sweetness values. Sweetness may, therefore, be thought of as a sensation involving a complicated interplay of structural and solution properties of sweeteners. An understanding of this interplay may lead to a better understanding of sweet taste chemoreception.

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