Study of some factors affecting intensity/time characteristics of sweetness†

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Intensity/time plots of sweetness produced by D-glucose, D-fructose and sucrose at concentrations ranging from 2·3 to 9·2% (w/v) were recorded for solutions at 15, 22 and 35°C. The intensity (I) and persistence (P) power functions were applied to the results obtained with a potentiometer connected to a chart recorder similar to the sensory measurement unit recording flux (SMURF) device. Increasing the concentration of assessed samples leads to an increase of perceived intensity with a tendency to show a compression for D-fructose and sucrose and an expansion for D-glucose. Persistence increases linearly as a function of concentration for the three sugars. Only very slight modification of intensity and persistence are observed when the temperature is varied from 15 to 35°C.

Intensity/time plots were also recorded at 22°C for solutions containing 5% sucrose or equisweet concentrations of D-glucose or D-fructose brought to apparent viscosities of 5, 15, 25 and 35 mPa s by addition of maltodextrins. It was found that the sweetness intensity decreases as viscosity increases for D-fructose and sucrose solutions whereas it remains constant for D-glucose. The persistence remains almost constant for the three sugars when the viscosity is varied.

The effect of temperature on viscosity coefficients and hydration numbers is measured for the three sugars. A decrease in intrinsic viscosity [η], B-coefficients and hydration numbers is observed with increasing temperature whilst the apparent specific volume is increased.

From the Raman spectra of water and aqueous solutions of sugars, it may be concluded that increasing the temperature leads to a lowering of the rigidity of the hydrogen bonded clusters and an increase in mobility of H_2O molecules. The increase in the size of the sugars derived from apparent specific volume ($\overline{V}_2°$) values reduces their accessibility to the receptor site. This effect is minimised as regards the perceived sweetness by the increased mobility of water. The effects of concentration, temperature and viscosity on the intensity and persistence of the sweet taste of D-glucose, D-fructose and sucrose, together with their physico-chemical properties in dilute solution, suggest that the accessibility of the sweet molecule to the receptor is an important step in the taste chemoreception. This step is followed by a biochemical phenomenon involving opening of ion channels which is sensitive to the mobility of water around the site and the sweetener.

INTRODUCTION

The perception of the sweet taste is controlled by a sequence of chemical, physical and electrophysiological interactions induced by the adsorption of a sweet stimulus on the receptor site. Such a localised excitation

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generates an action potential which, after initiation, moves as a wave over the membrane surface. When it reaches the peripheral nerve cells, it constitutes a neural message. The coding of this message is then processed by the central nervous system. The sweet stimulus goes through different layers of saliva before accession to the receptor membrane. The immediate environment of the site is composed of saliva. Any modification of the vicinal environment of the membrane may provoke specific changes of the membrane itself and alter the response to the stimulus. Therefore, it is of interest to in-

vestigate the influence of solution parameters, (e.g. viscosity, concentration and temperature) on the intensity and persistence of the sweet response for a class of molecules such as carbohydrates.

Each of the physico-chemical parameters may involve a specific perturbation. Temperature increase generally causes the weakening or even the breaking of intramolecular hydrogen bonds in sugars which affects the conformation of the sweet stimulus and modifies its (AH–B, γ) glucophore (Shallenberger & Lindley, 1977). On the other hand, changing the temperature of the vicinal layer of water (or saliva) contributes to a modification of the membrane itself and may lead to an alteration of the message. As the translation of the chemical stimulation into a nerve message proceeds through a number of steps, the influence of the temperature may occur at each of the steps (Bartoshuk *et al.*, 1982). It was observed (Birch & Munton, 1981) that the sweetness intensity for sugars and synthetic sweeteners shows an increase as temperature is increased while persistence decreases for sugars but not for other sweeteners. The behaviour of sugars when temperature is altered is easier to predict than that of artificial sweeteners (Green & Frankmann, 1987).

The perturbation provoked by viscosity change rather concerns the receptor membranes which are hindered in their interactions with the sapid molecules (Pangborn *et al.*, 1973). Viscosity is at the origin of a limitation in the diffusion of stimuli and the viscogen agent may envelop the receptor site and prevents its binding with the sweetener. Except for sucrose, it seems (Pangborn *et al.*, 1973) that the effect of hydrocolloids on the sweet taste depends much more on their physico-chemical properties and the type of sugar or sweetener than on the viscosity. This parameter seems to particularly affect the threshold of detection of the sweet taste (Paulus & Haas, 1980).

The effect of concentration, temperature and viscosity on the intensity/time characteristics of the sweetness response for D-glucose, D-fructose and sucrose in water is now studied. The intensity and persistence power functions were applied to the results obtained with a potentiometer connected to a chart recorder similar to the sensory measurement unit recording flux (SMURF) device of Birch and Munton (1981).

Interpretation of the evolution of sweetness intensity and persistence, as a function of temperature, concentration and viscosity, is based on the solution properties of the sugars and on their effect on water structure.

MATERIALS AND METHODS

The sugars studied were Sigma products or a gift of Roquette Frères (D-fructose and maltodextrin — Glucidex 2B). Water used for the sensory evaluation

was a mineral water, 'Volvic' known for its neutral taste (Sauvageot & Dumas, 1980).

A group of 15 trained panellists from the departmental personnel (males and females aged 20–60 years) were selected for their aptitude to detect and recognize the sweet taste. The assessed solutions were prepared 24 h prior to tasting.

The concentrations of D-glucose, D-fructose and sucrose in water were 2·3, 4·6, 6·9 and 9·2% (w/v) which corresponds to about 5, 10, 15 and 20 times the threshold values (Birch & Munton, 1985). The investigated temperatures were 15, 22 and 35°C. Intensity/time plots were recorded at 22°C for solutions containing 5% sucrose or the equisweet concentrations of D-glucose or D-fructose brought to apparent viscosities of 5, 15, 25 and 35 mPa s by addition of maltodextrin (Glucidex 2B). The intensity and persistence of sweetness were determined by use of a SMURF which consists of a potentiometer connected to a chart recorder. The judges are trained to set the plotter at 50% of the recorder scale when a 5% (w/v) sucrose solution is tasted at 22°C. The evaluation of the sensation consists in moving the lever of the potentiostat proportionally to the intensity of the solution.

In order to interpret the sensory evaluation results, intrinsic viscosities and apparent specific volumes were determined according to methods previously described (Kemp *et al.*, 1990) as well as the Raman spectra (Portmann & Mathlouthi, 1990) of the studied solutions. The values of the sweetness intensity and persistence derived from the SMURF plot were treated statistically by use of a programme developed by the French Institute of Cereals and recommended (Danzart, 1990) for the treatment of sensory evaluation results. This programme involves inferential and data analysis moduli. It permits variance analysis and comparison of variances with *F*-values from the Fisher's table. A significance level of 0·05 to 0·01 was used.

RESULTS AND DISCUSSION

Intensity and time characteristics

Effects of temperature and concentration on sweetness intensity

The values of sweetness intensity obtained with SMURF plots are reported in Table 1 for the studied solutions of D-glucose, D-fructose and sucrose at 15, 22 and 35°C. The mean values are calculated and standard deviations given in parentheses. In order to show the variation of intensity in terms of concentration at the three temperatures, results are also represented in Fig. 1. Increasing the concentration of the assessed sugars leads to an increase of perceived intensity with a tendency to exhibit a compression for D-fructose and sucrose and an expansion for D-glucose. Such a different behaviour of

Table 1. Sweetness intensity of D-glucose, D-fructose and sucrose at a varied concentrations and at 15, 22 and 35°C

Sugars	Temperature (°C)	Concentration (w/v)			
		2·3%	4·6%	6·9%	9·2%
Glucose	15	7·67	16·08	29·50	48·79
		(2·30)	(3·27)	(2·55)	(9·2)
	22	14·52	25·87	39·50	61·66
		(3·55)	(4·38)	(5·32)	(4·64)
	35	10·03	22·53	33·86	54·35
		(2·07)	(3·89)	(3·40)	(5·19)
Fructose	15	18·32	54·14	75·71	87·40
		(2·31)	(5·17)	(4·06)	(3·44)
	22	28·73	61·68	84·90	89·24
		(3·36)	(7·10)	(3·13)	(3·29)
	35	25·24	49·32	78·12	82·24
		(3·11)	(4·27)	(4·58)	(4·60)
Sucrose	15	18·55	44·60	75·05	82·00
		(2·61)	(6·95)	(5·10)	(5·86)
	22	27·75	54·80	75·00	90·25
		(3·52)	(6·71)	(5·04)	(2·96)
	35	21·28	46·53	72·83	82·87
		(4·13)	(6·86)	(4·01)	(4·15)

Mean values are SMURF values and (SD).

D-glucose when compared to the sweetness of D-fructose and sucrose was also observed by McBride (1986). The initiation of a plateau is observed for D-fructose and sucrose (see Fig. 1) when the concentration reaches 7–9% (w/v), which is not the case for D-glucose at all temperatures investigated. This result may be confirmed by the statistical analysis of variance of the intensities for concentrations 6·9 and 9·2%. Sweetness intensities are found significantly different for D-glucose at 15, 22 and 35°C. Indeed, even with a significance level of 0·01, the F-values obtained from the Fisher table are lower than the calculated F-ratios at all investigated temperatures ($15°C : F(1·23) = 16·54, p < 0·01$; $22°C: F(1·25) = 9·83, p < 0·01$; $35°C : F(1·25) = 10·88, p < 0·01$).

For D-fructose, the difference in sweetness intensity is only significant at 15°C ($F(1·27) = 4·80, p < 0·05$) for solutions at 6·9 and 9·2% whereas no significance is calculated for 22 and 35°C. The intensities for sucrose sweetness are not found statistically different at 35°C. It seems that, for D-fructose and sucrose, a convergence of intensity values exists around 9–10% for all temperatures investigated (see Fig. 1). The level at which no difference of sweetness is detected, even if concentration is noticeably increased, is situated at about 18% of sucrose (Pangborn & Christ, 1966). The slight difference in intensity between D-fructose and sucrose is reduced when the concentration is increased (see Fig. 1) which leads to the conclusion that concentration affects the taste of fructose more than that of other sugars.

On the other hand, analysis of the variance of the
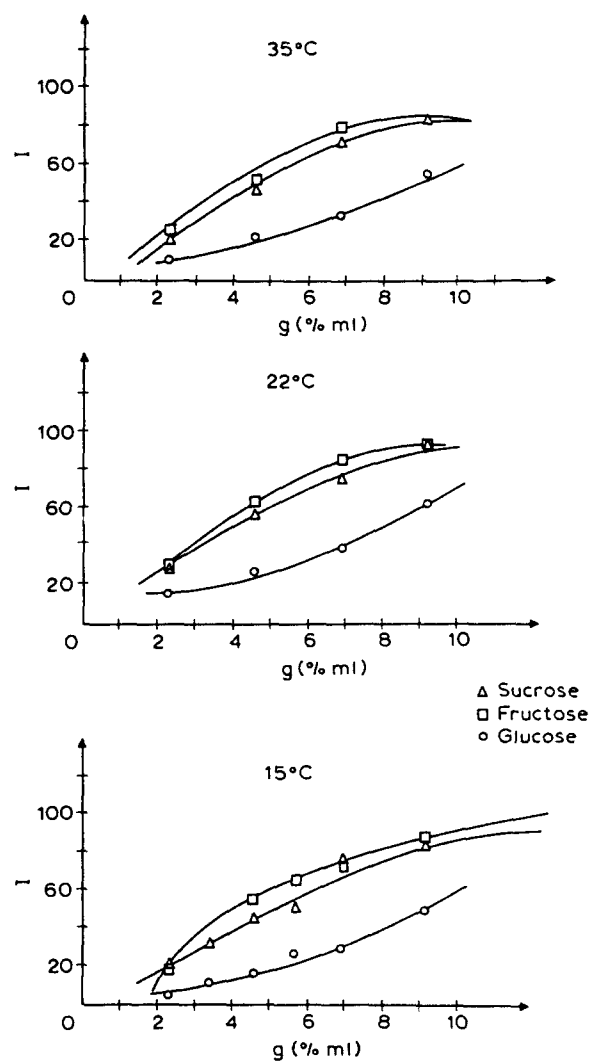
Fig. 1. Variation of sweetness intensity of D-glucose, D-fructose and sucrose in function of concentration (g%ml) at 15, 22 and 35°C.

results of sweetness intensity at a given concentration does not reveal any significant difference when the temperature is increased from 15 to 35°C (see Table 1 and Fig. 2). The fact that temperature does not cause any noticeable variation of intensity was observed by Stone *et al.* (1969). The authors studied D-glucose and D-fructose between 5 and 50°C by reference to a sucrose solution assessed at 22°C which was also the present reference mark. However, the sweet taste of the three sugars is slightly higher at 22°C than it is at 15 or 35°C (see Fig. 2), which is in agreement with the results obtained by Calvino (1986) for sucrose between 7 and 50°C. Observation of Fig. 2 and analysis of variance show that the intensities at 22 and 35°C are not significantly different for concentrations 6·9 and 9·2% of D-fructose. The lower relative fructose sweetness as concentration and temperature are increased is related to the mutarotational equilibrium of this sugar. Indeed it is well known (e.g. Tsuzuki & Yamazaki, 1953 Shallenberger, 1978) that
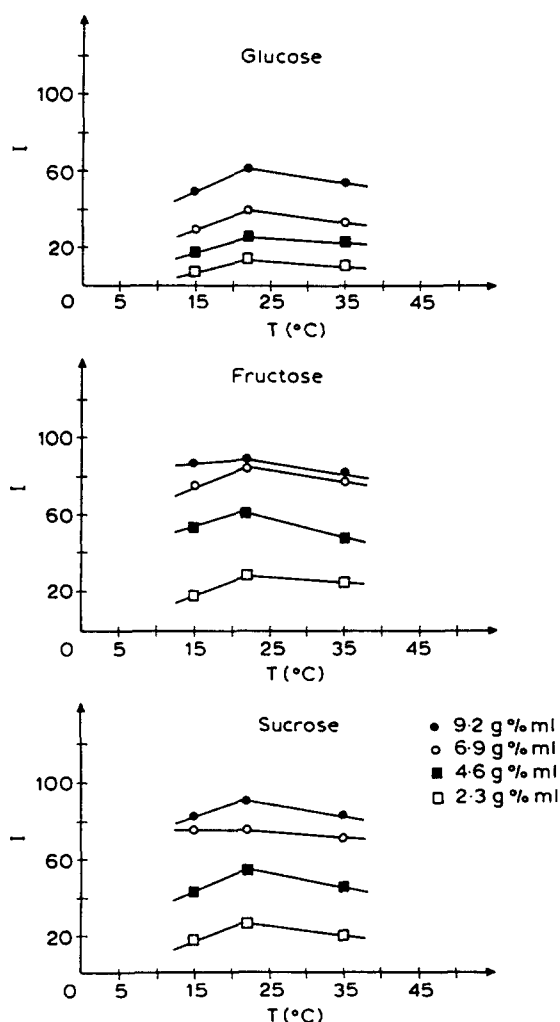
Fig. 2. Variation of sweetness intensity of D-glucose, D-fructose and sucrose in function of temperature and concentration.

the proportion of β-D-fructropyranose, the sweetest isomer, is reduced in favour of β-D-fructofuranose, a less sweet isomer, when temperature is increased.

Effects of temperature and concentration on persistence
Results of persistence estimation from the SMURF plots are reported in Table 2 and Figs 3 and 4. Analysis of variance did not reveal any statistically significant difference in persistence between D-glucose, D-fructose and sucrose solutions when the temperature is varied. Persistence of sweetness increases with concentration for the three sugars (see Fig. 3). However, no convergence of the observed values is found as for intensity. Although not statistically different, the data show, for persistence, the same tendency as for intensity, namely the results at 22°C are higher than at 15 or 35°C. For D-fructose solutions, the lower persistence is found at 35°C which may also be related to the fact that an increase in temperature leads to a decrease in proportion of the sweetest isomer, β-D-pyranose (Shallenberger, 1978).

Table 2. Sweetness persistence of D-glucose, D-fructose and sucrose at varied concentrations and at 15, 22 and 35°C

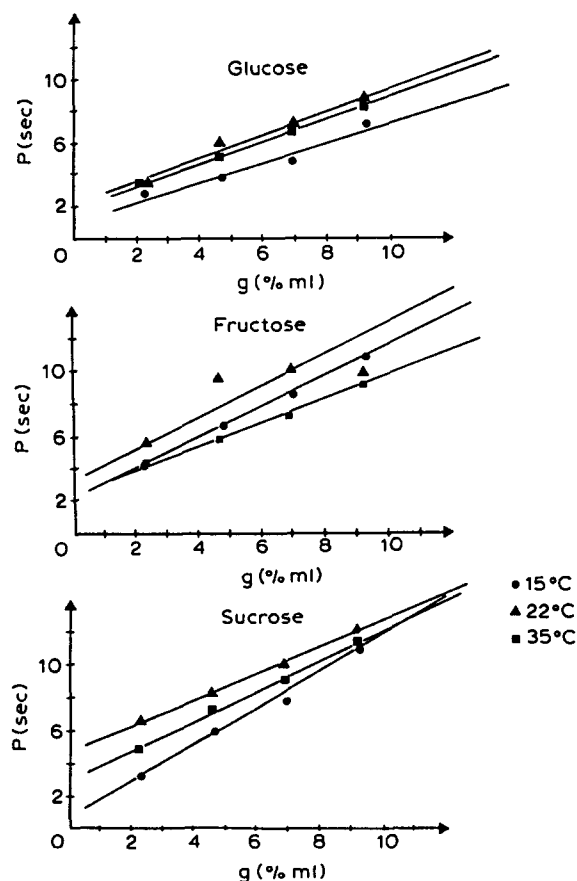| Sugars	Temperature (°C)	Concentration (w/v)			
		2·3%	4·6%	6·9%	9·2%
Glucose	15	2·83	3·71	4·70	6·95
		(0·39)	(0·58)	(0·32)	(0·88)
	22	3·27	5·84	6·92	8·50
		(0·45)	(1·04)	(1·42)	(1·43)
	35	3·34	4·88	6·69	8·07
		(0·53)	(0·95)	(1·24)	(1·16)
Fructose	15	4·25	6·71	8·40	10·80
		(0·66)	(0·70)	(0·82)	(1·50)
	22	5·57	9·50	10·00	9·66
		(0·98)	(1·86)	(2·09)	(1·36)
	35	4·10	5·80	7·15	9·35
		(0·70)	(0·70)	(0·79)	(1·73)
Sucrose	15	3·20	6·00	7·80	10·50
		(0·62)	(1·16)	(1·08)	(1·50)
	22	6·54	8·25	10·00	12·25
		(1·20)	(1·58)	(1·43)	(2·48)
	35	4·75	7·33	9·08	12·37
		(0·72)	(0·90)	(1·23)	(2·00)

Mean values are seconds and (SD).

Fig. 3. Variation of persistence of sweetness in seconds for D-fructose, D-fructose and sucrose in function of concentration at 15, 22 and 35°C.
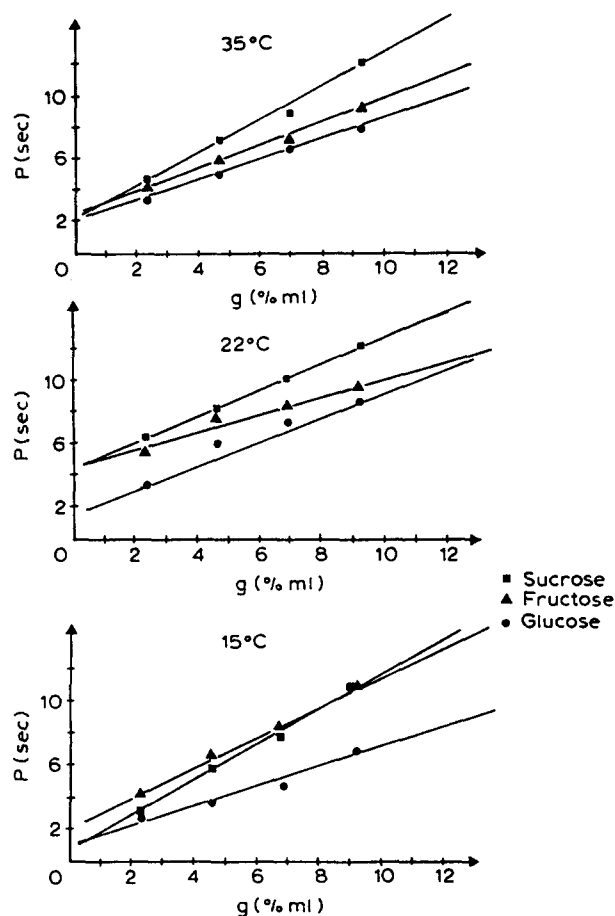
Fig. 4. Variation of sweetness persistence of D-glucose, D-fructose and sucrose in function of temperature and concentration.

Fig. 5. Sweetness intensity of D-glucose, D-fructose and sucrose in function of solution viscosity.

Effect of viscosity on intensity and persistence

The variation of sweetness intensity and persistence in function of solution viscosity is shown in Figs 5 and 6.

Analysis of variance reveals that the addition of maltodextrins to glucose solution does not provoke any statistically significant change in sweetness intensity. On the other hand, the intensity of sweetness is largely decreased for D-fructose and sucrose solutions when the viscosity is increased, especially at the beginning of the curve between 1 and 5 mPa s. (for D-fructose, $F(1 \cdot 19) = 8 \cdot 04$, $p < 0 \cdot 025$; for sucrose, $F(1 \cdot 22) = 7 \cdot 09$, $p < 0 \cdot 28$). For viscosities higher than 5 mPa s, no difference is detected between the intensities of sweetness for the three sugars. These results are comparable to those of Arabie and Moskowitz (1978) who found that the addition of viscogenic bulk agents to sugar solutions contributes to the lowering of their sweet taste. It may also be observed (see Fig. 5) that sweetness intensity of D-fructose solutions decreases and becomes lower than that of sucrose when viscosity is increased, which is in agreement with Pangborn (1963).

No significant difference in sweetness persistence is observed for the three sugars when viscosity is increased (see Fig. 6). Only a slight variation in the general aspect of curves is seen (Fig. 6) at low viscosities
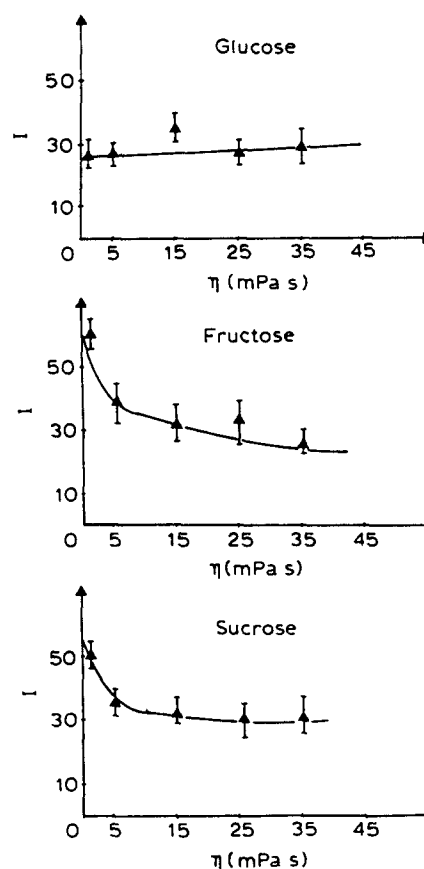
(lower than 5 mPa s). The role of the viscogen agent in masking the receptor site and lowering the differences between the three sugars appears better for persistence than for intensity.

Power functions of intensity and persistence

Intensity and persistence may be related to concentration by a power function of the type:

$$S = k_s \, C^{n_s} \text{ and } P = k_p \, C^{n_p}$$

with S intensity, P persistence, n exponent and k constant. Parameters k and n were respectively interpreted as representative of accession (Moskowitz, 1970) to receptor and of interactions between the stimuli and receptor. Efficiency of these interactions were found by Munton and Birch (1985) to depend on the structure of the stimulus. The constants k and n are determined from the graphic representation of log$S$ and log$P$ in function of concentration as the ordinate at the origin and the slope of the line respectively.

Results are reported in Table 3 for sweetness intensity and Table 4 for persistence. It may be observed that n_s is higher for D-glucose than D-fructose or sucrose, which is interpreted (Shamil *et al.*, 1988) as an aptitude for D-glucose to be as sweet or even sweeter than sucrose when concentration is increased. Accession of stimuli
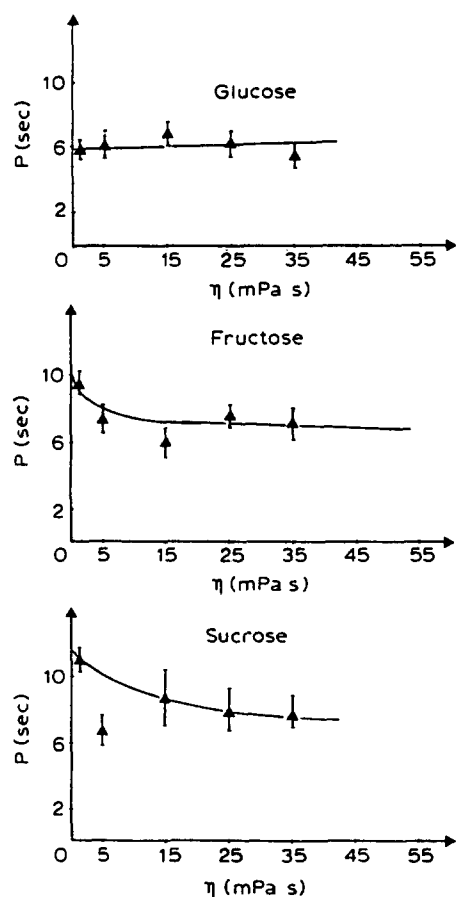
Fig. 6. Sweetness persistence of D-glucose, D-fructose and sucrose in function of solution viscosity.

and their binding to receptor sites seems to be maximum at 22°C, but the efficiency of binding is greater at 35°C than it is at 15°C because k_s (35°C) > k_s (15°C) and n_s (35°C) < n_s (15°C) (see Table 3). The ease of accession to sites is higher for D-fructose and sucrose as well as the efficiency of interactions than it is for D-glucose. The effect of temperature treated by the power function ($S = k_s C^{n_s}$) leads to a result comparable to what was observed in Fig. 1, namely that the behaviour of D-glucose is different from that of D-fructose and sucrose.

Values of k_p and n_p are generally lower than that of k_s and n_s. For a given sugar, persistence has its maximum at higher concentration than that of intensity maximum (Birch & Ogunmuyela, 1980). Differences between their persistence parameters k_p and n_p for the three sugars are less important than what is observed for k_s and n_s (see Table 4). These results show that intensity and persistence very likely proceed from different and independent mechanisms of taste perception.

Power functions were also applied to solutions with increased viscosity and k and n calculated. Their values are shown in Tables 5 and 6 respectively for intensity and persistence. The difference in k_s and n_s values for viscous and non-viscous solutions are very important. Increase of k_s seems to be related to an increase of occupancy of sites by both the sugar and the bulk agent while the decrease in n_s could be ascribed to less efficient binding of sugars to the site. Moreover, this lack of efficiency is the cause of decrease in sweetness intensity. The effect on persistence parameters k_p and n_p, although less important than on intensity (see Table 6) leads to the idea that the enveloping of sites by maltodextrin makes it difficult for stimuli molecules in 'queue' (Birch et al., 1980) to bind to receptor.

Solution properties and Raman spectra

Accession of sugars to receptor sites and their binding propensities to these sites may also be derived from their physico-chemical properties. Parameters such as intrinsic viscosity $[\eta]$, apparent specific volume ($\overline{V_2^\circ}$) and other viscosity coefficients or hydration numbers are of relevance in interpreting taste. Results of determination of intrinsic viscosity, Huggins coefficients and apparent specific and molar volumes are given in Table 7. B-Coefficients and hydration numbers are given in Table 8 for the three sugars at 18, 25 and 35°C.

Although both $[\eta]$ and V_2° are expressed in cm³ g⁻¹ and account, respectively, for hydrodynamic and static volumes of the hydrated sugars (Shamil et al., 1987), it is observed (Table 7) that intrinsic viscosity $[\eta]$ decreases as temperature is increased while apparent

Table 3. Regression line equations for intensity = f(c), correlation coefficient, r, and coefficients k_s and n_s ($S = k_s C^{n_s}$) for sugar solutions at 15, 22 and 35°C

Sugars	Temperature (°C)	Linear equations	r	k_s	n_s
Glucose	15	y = 0·33 + 1·40 ×	0·99	2·13	1·40
	22	y = 0·76 + 1·04 ×	0·99	5·75	1·12
	35	y = 0·56 + 1·19 ×	0·99	3·63	1·25
Fructose	15	y = 0·86 + 1·20 ×	0·97	7·24	1·06
	22	y = 1·17 + 0·87 ×	0·97	15·24	0·85
	35	y = 1·09 + 0·90 ×	0·98	12·60	1·00
Sucrose	15	y = 0·89 + 1·10 ×	0·98	8·00	1·06
	22	y = 1·94 + 0·86 ×	0·99	14·12	0·83
	35	y = 0·96 + 1·02 ×	0·99	10·00	1·00

Table 4. Regression line equations for persistence $= f(c)$, correlation coefficient, r, and coefficients k_p and n_p $(P = k_p C^{n_p})$ for sugar solutions at 15, 22 and 35°C

Sugars	Temperature (°C)	Linear equation	r	k_p	n_p
Glucose	15	y = 0·15 + 0·63x	0·94	1·41	0·75
	22	y = 0·28 + 0·70x	0·99	2·00	0·87
	35	y = 0·28 + 0·64x	0·99	2·00	0·61
Fructose	15	y = 0·34 + 0·71x	0·98	2·18	0·62
	22	y = 0·62 + 0·42x	0·89	4·16	0·45
	35	y = 0·40 + 0·98x	0·99	2·51	0·55
Sucrose	15	y = 0·23 + 0·81x	0·98	1·70	0·75
	22	y = 0·69 + 0·45x	0·98	4·46	0·46
	35	y = 0·41 + 0·67x	0·99	2·56	0·73

specific volume $(V_2°)$ increases. Change of intrinsic viscosity towards lower values could originate in the rupturing of hydrogen bonds between solute and solvent when the temperature is raised. On the other hand, the increase of apparent specific volume is due to thermal expansion of sugar molecules which is predominant over hydration decrease. The fact that the lowering of intrinsic viscosity is much more related to water structure change around the solutes than to their sizes is derived from B-coefficients (see Table 8). Indeed, the B-coefficient which is generally comparable to $[\eta]$, is the sum of B_{size} and $B_{structure}$, respectively accounting for the size of solute and its effect on solvent structure (Miyajima *et al.*, 1983; Kemp *et al.*, 1990). It seems that temperature affects much more $B_{structure}$ than B_{size} (see Table 8). Moreover, the hydration number, h, also falls rapidly when temperature is increased from 18 to 35°C (Table 8). The number of water molecules with a residence time in the vicinity of sugars longer than in that of another water molecule is given by h. Another parameter which accounts for the rate of exchange of

water molecules around sugar molecules is k'_H. This parameter is generally assigned (Shamil *et al.*, 1987; Kemp *et al.*, 1990) to solute–solvent interactions. Its increase with temperature (see Table 7) is observed for D-fructose and sucrose, and not for D-glucose. Increase of k'_H probably originates from thermal motion except for D-glucose for which the equatorial positions for most OH groups contribute to stabilise the hydration.

Table 7. Intrinsic viscosities $[\eta]$, Huggins coefficients (k'_H), apparent specific volumes $(V_2°)$ and partial molar volumes $(\phi(V_2°))$

Sugars	Temperature (°C)	$[\eta]$ (cm³ g⁻¹)	k'_H	$V_2°$ (cm³ g⁻¹)	$\phi(V_2°)$ (cm³ mol⁻¹)
Glucose	18	2·48	0·88	0·621	111·90
	25	2·42	0·84	0·615	110·80
	35	2·26	0·62	0·629	113·35
Fructose	18	2·39	0·69	0·603	108·66
	25	2·33	0·55	0·603	108·66
	35	2·09	0·99	0·622	112·14
Sucrose	18	2·59	0·65	0·613	209·83
	25	2·48	0·81	0·613	209·83
	35	2·21	0·91	0·676	231·39

Table 5. Coefficients k_s and n_s for sweetness intensity of sugar solutions with and without maltodextrins

Sugars	Without maltodextrins		With maltodextrins	
	k_s	n_s	k_s	n_s
D-Glucose	5·75	1·12	26·30	0·03
D-Fructose	15·84	0·85	53·70	−0·18
Sucrose	14·12	0·83	38·74	−0·10

Table 6. Coefficients k_p and n_p for sweetness persistence of sugar solutions with and without maltodextrins

Sugars	Without maltodextrins		With maltodextrins	
	k_p	n_p	k_p	n_p
D-Glucose	2·00	0·87	6·70	0·035
D-Fructose	4·16	0·45	7·24	0·01
Sucrose	4·46	0·46	6·30	0·10

Table 8. Viscosity B-coefficients and hydration numbers, h, of sugars

Sugars	Temperature (°C)	B (dm³ mol⁻¹)	B_{size} (dm³ mol⁻¹)	$B_{structure}$ (dm³ mol⁻¹)	h
Glucose	18	0·45	0·28	0·17	3·60
	25	0·43	0·28	0·15	3·50
	35	0·41	0·28	0·13	2·74
Fructose	18	0·43	0·27	0·16	3·50
	25	0·43	0·27	0·15	3·30
	35	0·38	0·28	0·10	2·24
Sucrose	18	0·89	0·53	0·36	8·00
	25	0·84	0·53	0·31	6·89
	35	0·75	0·58	0·17	3·80

$\eta_{rel} = 1 + BC + DC^2$
$B_{size} = 2·5 \phi (V_2°)/1000$ and $B_{structure} = B - B_{size}$.
(Miyajima *et al.*, 1983).

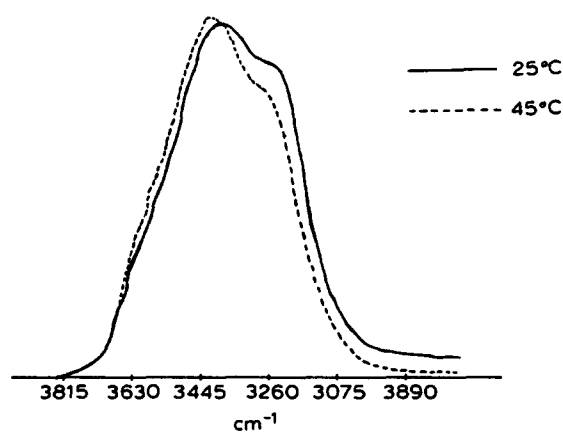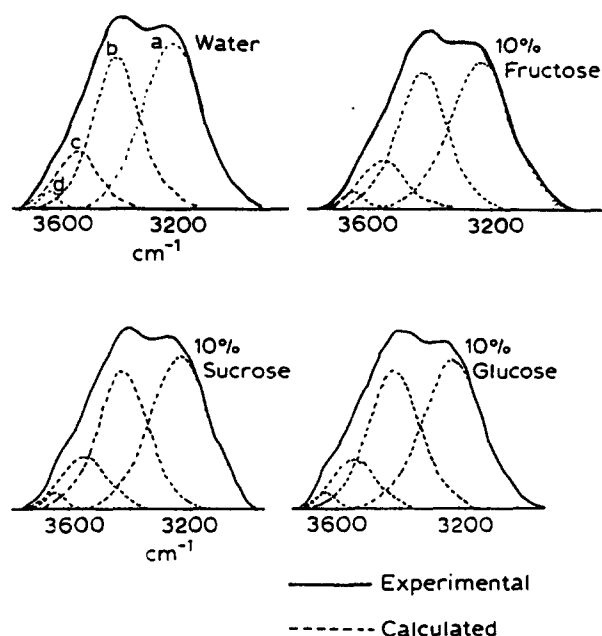Fig. 7. Raman spectra of water at 25 and 45°C.

Fig. 8. Experimental bands and calculated components of the Raman spectra of aqueous solutions (10% (w/w)) of D-glucose, D-fructose and sucrose.

The effect of temperature on viscometric and volumetric properties of D-glucose, D-fructose and sucrose shows that water structure and interactions are more affected than the size of the sugars. It is also observed that D-glucose behaves differently from D-fructose and sucrose. This differentiation was also observed from sensory analysis results. Interpretation of the role of water in sweet taste chemoreception needs investigation at the molecular level, and such can be approached with Raman spectra of water and aqueous solutions of sugars. The effect of temperature on pure water is given in Fig. 7 and Table 9. Decomposition of the Raman band of water in the region of stretching according to a semi-empirical method (Luu et al., 1982) allows four components to be assigned to more or less rigidly hydrogen-bonded species. This model of the structure of water is used as a reference mark when it is desired to follow the evolution of water organisation under the effect of temperature of solutes. Heating water from 25°C to 45°C results in a decrease in the proportion of the quasi-crystalline (a) component in favour of the less organized components (b) and (c) (see Table 9). This result is expected because the overall effect of heat on hydrogen bonding generally provokes loosening of the most rigid species. The energy of binding between

Table 9. Decomposition of Raman spectra of water at 25 and 45°C. Effects of temperature on frequency and intensity (area of band) of vibration

Temperature (°C)	Component	ν (cm^{-1})	Total area (%)	$\Delta\nu$	Δ Area (%)
25	a	3236	55·4	—	—
	b	3421	33·1	—	—
	c	3555	9·6	—	—
	d	3638	1·9	—	—
45	a	3237	50·0	+1	−5·4
	b	3419	37·0	−2	+3·9
	c	3554	11·5	−1	+1·9
	d	3635	1·5	−3	−0·4

water molecules is less perturbed as no noticeable change is observed for the frequencies of the four species of associates (see values of $\Delta\nu$ Table 9).

Raman spectra of aqueous solutions of D-glucose, D-fructose and sucrose (10% (w/w) concentration) are reported in Fig. 8 and the results of decomposition of the observed band summarised in Table 10. The general aspect of the spectra is almost the same for the three sugars. The details of decomposition of Raman bands given in Table 10 show that proportion (percentage of intensity) of each of components is less affected than the energy of hydrogen bonds ($\Delta\nu$). For all three sugars, frequencies shift towards higher values for the most organised water clusters (a) and (b). It is generally accepted that sugars have a 'structure promoter' effect on water. The influence of temperature and that of the addition of sugars on water structure seem to be opposed. This is probably the reason why the expected influence of temperature, namely an easier accession to site because of an increase in mobility of water, is cancelled by the structure-maker effect of sugars. It should also be noted that Raman spectra were recorded at a concentration of 10% (w/w), for which the sensory evaluation results reveal less difference in intensity and persistence than what is observed at 2·3%. Another structural feature of water, which can help in interpreting the higher sweetness intensity observed at 22°C for the three sugars, is the relative stability of water structure revealed by a plateau of resistivity between 18 and 24°C (Mathlouthi et al., 1980). As water structure is less perturbed by solutes at 22°C than at 15 or 35°C, the sensory evaluation is then less influenced by temperature and the perceived intensity is slightly higher.

Table 10. Position of maximum (v) area (integrated intensity), total area (%), shifts in frequencies (Δv) and in % total area (Δ area %) of the Raman band of water and aqueous solutions of sugars

Water

Component	v (cm^{-1})	Area (arb. units)	% total area	Assignments
a	3223	50 384	48·2	Quasi-crystalline
b	3413	39 432	37·7	Solid-like amorphous
c	3547	13 160	12·6	Liquid-like amorphous
d	3638	1593	1·5	Unassociated H_2O, Free OH·

D-fructose 10%

Component	v (cm^{-1})	% total area	Δv (cm^{-1})	Δ Area (%)
a	3229	48·5	+6	−0·3
b	3417	38·0	+4	+0·3
c	3547	11·8	—	−0·8
d	3638	1·6	—	+0·1

D-glucose 10%

Component	v (cm^{-1})	% total area	Δv (cm^{-1})	Δ Area (%)
a	3228	48·2	+5	0
b	3417	38·6	+4	+0·9
c	3544	11·5	−3	−0·3
d	3638	1·7	—	+0·2

Sucrose 10%

Component	v (cm^{-1})	% total area	Δv (cm^{-1})	Δ Area (%)
a	3229	48·6	+6	+0·4
b	3416	37·2	+3	−0·5
c	3544	12·5	−3	−0·1
d	3636	1·7	−2	+0·2

CONCLUSIONS

The effect of temperature on sweetness intensity of D-glucose, D-fructose and sucrose in water between 2·3 and 9·2% (w/v) did not reveal any significant differences. This result is interpreted in terms of solute–solvent interactions and the structure-maker effect of sugars in water. The slight decrease in sweetness of fructose as temperature is raised probably comes from the mutarotation rate which increases and provokes the transformation of the most sweet isomer (β-pyranose) into a less sweet one (β-furanose). Persistence of sweet taste does not change significantly when temperature is increased. A slight decrease of persistence is also observed at 35°C. Increase of viscosity depresses intensity and persistence probably due to a masking of receptor site by viscosity-enhancer maltodextrins.

The effects of sugars and temperature on water structure are revealed by the viscometric and volumetric parameters on the one hand and by the decomposition of Raman spectra in the region of OHs on the other.

Perturbation of water structure may be deduced from both methods of analysis. Accession of sugars to receptor sites seems to depend on water structure around the sugar and the site.

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REFERENCES

Arabie, P. & Moskowitz, H. R. (1971). The effect of viscosity upon perceived sweetness. *Perception and Psychophysics,* **9**(5), 410–12.

Bartoshuk, L. M., Reinnert, K., Rodin, J. & Stevens, J. C. (1982). Effect of temperature on the perceived sweetness of sucrose. *Physiol. and Behav.*, **28**, 905–10.

Birch, G. G. & Munton, S. E. (1981). Use of the 'SMURF' in the taste analysis. *Chem. Senses*, 6(1), 45–52.

Birch, G. G. & Munton, S. L. (1981). Evidence for a constant number of available sweet receptor sites at threshold concentrations of sugars. *Experientia*, 37, 839–40.

Birch, G. G. & Ogunmoyela, G. (1980). Effect of surfactants on the taste and flavour of drinking chocolate. *J. Food Sci.*, 45(4), 981–4.

Birch, G. G., Latymer, Z. & Hollaway, M. (1980). Intensity/time relationships on sweetness: evidence for a queue hypothesis in taste chemoreception. *Chem. Senses*, 5(1), 63–78.

Calvino, A. M. (1986). Perception of the sweetness: the effect of concentration and temperature. *Physiol. and Behav.*, 36, 1021–8.

Danzart, M. (1990). In *Evaluation Sensorielle*, ed. SSHA and ISHA. Technique et Documentation, Lavoisier, Paris, pp. 250.

Green, B. G. & Frankmann, S. P. (1988). The effect of cooling and the perception of carbohydrate and intensive sweeteners. *Physiol. Behav.*, 43, 515–19.

Kemp, S. E., Birch, G. G., Portmann, M. O. & Mathlouthi, M. (1990). Intrinsic viscosities and apparent specific volume of amino acids and sugars. 'Effective size' and taste of sapid molecules. *J. Sci. Food Agric.*, 51, 97–107.

Luu, C., Luu, D. V., Rull, F. & Sopron, F. (1982). Raman effect study on structural perturbation of liquid water by a foreign substance. *J. Mol. Struct.*, 81, 1–10.

Mathlouthi, M., Conry, M., Jaillant, G. & Maitenaz, P. C. (1980). Water vapor sorption of gruyere cheese. *Lebensm.-Wiss.u.-Technol.*, 13, 264–8.

McBride, R. L. (1986). Sweetness of Binary Mixtures of Sucrose, Fructose and Glucose. *J. Exp. Psych. Human Perception and Performance*, 12(4), 584–91.

Miyajima, K., Sawada, M. & Nakagaki, M. (1983). Studies on aqueous solutions of saccharides II. : Viscosity-B-coefficients; apparent molar volumes and activity coefficients of D-glucose, maltose and maltotriose in aqueous solutions. *Bul. Chem. Soc. of Japan*, 56, 1954–7.

Moskowitz, H. R. (1970). Ratio scales of sugar sweetness. *Percept. Psychophys.*, 7, 315–22.

Munton, S. L. & Birch, G. G. (1985). Accession of stimuli to receptor. I. Absolute dominance of one molecular species in binary mixtures. *J. Theoret. Biol.*, 112, 539–51.

Pangborn, R. M. (1963). Relative taste intensities of selected sugars and organic acids. *J. Food. Sci.*, 28, 726–33.

Pangborn, R. M. & Christ, R. B. (1966). Gustatory response to anomeric sugars. *Experientia*, 22, 612–15.

Pangborn, R. M., Trabane, I. & Szczerniak, A. (1973). Effect of hydrocolloids on oral viscosity and basic taste intensities. *J. Text. Struct.*, 4, 224–41.

Paulus, K. & Haas, E. M. (1980). The influence of solvent viscosity on the threshold of primary tastes. *Chem. Senses*, 5(1), 23–33.

Portmann, M. O. & Mathlouthi, M. (1990). Hydrogen bonding and sweet taste mechanism. *J. Mol Struct.*, 237, 327–38.

Sauvageot, F. & Dumas, P. (1980). Propriétés organoleptiques d'une nouvelle formule d'édulcorant à base d'aspartame et de xylitol. *Annales Nutrition & Alimentation*, 34, 121–30.

Shallenberger, R. S. (1978). Intrinsic chemistry of fructose. *Pure & Applied Chem.*, 50, 1409–20.

Shallenberger, R. S. & Lindley, M. G. (1977). A lipophilic–hydrophobic attribute and component in the stereochemistry of sweetness. *J. Food Science*, 30, 560.

Shamil, S., Birch, G. G., Mathlouthi, M. & Clifford, M. N. (1987). Apparent molar volumes and taste of molecules with more than one sapophore. *Chem. Senses*, 12(2), 397–409.

Shamil, S., Birch, G. G., Jackson, A. A. S. F. & Meek, S. (1988). Use of intensity–time studies as an aid to interpreting sweet taste chemoreception. *Chem. Senses*, 13(4), 597–605.

Stone, H., Oliver, S. & Kloehn, J. (1969) Temperature and pH effects on the relative sweetness of suprathreshold mixtures of dextrose–fructose. *Percept. Psychophys.*, 5, 257–60.

Tsuzuki, Y. & Yamazaki, J. (1953). On the sweetness of fructose and some other sugars, especially its variation with temperature. *Biochemische Zeitschrift, Bd*, 323(S), 525–31.