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A study of the solution properties of selected binary mixtures of bulk and intense sweeteners in relation to their psychophysical characteristics

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Abstract

The solution properties of binary mixtures of two bulk sweeteners (sucrose and maltitol) and three intense sweeteners (acesulfame K, aspartame and sodium cyclamate) were studied. The parameters measured were apparent specific volumes, isentropic compressibilities and compressibility hydration numbers. An attempt has been made to correlate the solution properties of some of the mixtures with their sweetness responses. Both specific volume and isentropic compressibility data have been used to interpret the possibility of synergism or suppression in the mixtures in terms of the affinity of the solutes for the surrounding solvent structure, and hence the effectiveness of transport of the molecules to their appropriate receptor sites. However, an understanding of the nature and stereochemistry of the individual components in the mixtures is necessary for predicting the packing characteristics of molecules in water and their accession to receptor sites. The intense sweeteners seem to play an important role in modifying the structure of water in solution. Although there is no clear proof of how this affects perception, fine differences in solution behaviour can be observed with isentropic apparent specific compressibility measurements. The study supports the idea that receptors lie at different depths in the lingual epithelium, and also that in two-component systems, one species may alter the packing efficiency of the other in solution sufficiently to effect a change in taste quality or intensity. © 1999 Published by Elsevier Science Ltd. All rights reserved.

1. Introduction

Mixing of sweeteners is commonly practised in food and beverage formulations to achieve optimisation of acceptability, functionality and economics. However, mixing may give sweetness values which are synergistic, additive or suppressive. Extensive research has been carried out on determining the taste intensities and qualities of mixtures of sweeteners, and the tendency now is to mix bulk and intense sweeteners to optimise the mouthfeel characteristics of food products, and to create a "sweet experience" as close as possible to that exhibited by sucrose. The increasing use of intense sweeteners is advantageous for diabetic and dietetic reasons as well as in the manufacture of "tooth-friendly" food and pharmaceutical products. They also offer technological and nutritional advantages. Very little is known, however, about the underlying mechanism of taste, leading to the responses observed by panellists. In the past decade, the emphasis has been laid upon the importance of the aqueous medium to taste perception. Studies on the hydration behaviour of solutes in water are being used to explain the taste of molecules (Birch, Parke, Siertsema & Westwell, 1997; Hutteau, Mathlouthi, Portmann & Kilcast, 1998; Mathlouthi, Bressan, Portmann & Serghat, 1993; Mathlouthi & Portmann, 1994; Parke & Birch, 1999; Parke, Birch & Dijk, 1999; Parke, Birch, MacDougall & Stevens, 1997). In this study, the investigation of the behaviour of the sapid solutes in water may help illuminate, not only the mechanism of taste in general, but also the synergistic tendencies of mixtures.

A previous study of the psychophysical characteristics of binary mixtures of selected bulk sweeteners (sucrose and maltitol) and intense sweeteners (acesulfame K, aspartame and sodium cyclamate) by Hutteau et al. (1998) has reported synergism, additivity and suppression for the mixtures shown in Table 1. The psychophysical responses show that mixtures of the bulk sweeteners with aspartame

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consistently exhibit suppression of sweet taste whereas those with sodium cyclamate or acesulfame K tend to have synergistic or additive properties, respectively. Table 1 also lists the ratio of bulk:intense sweeteners at which these mixtures exhibit synergism, additivity or suppression of sweet taste.

This paper now focuses on the solution properties of the above named mixtures of bulk and intense sweeteners, at the same and different proportions as studied by Hutteau et al. (1998). Using different concentrations in the mixtures will allow the solution effects of each sweetener to be evaluated separately and in the presence of the other sweetener, the concentration of the latter being held constant. The establishment of a possible relationship between synergism, suppression or additivity and the solution properties of the mixture is then attempted. Parameters such as apparent specific volumes (V_2), isentropic apparent specific compressibilities [$K_2(s)$] and compressibility hydration numbers (n_h) are used to explain the state of hydration of the solutes in water and hence their packing characteristics among solvent molecules. The accession of solutes to receptor sites is explained in terms of solute–solute, solute–solvent and solvent–solvent interactions.

The hydration of a solute molecule in water is based on the Frank and Wen (1957) model of solute–solvent interaction which pictures three different solvent-structure regions in the neighbourhood of the solute (Fig. 1). Just outside the solute is a layer of immobilised, compressed water (X) as a result of electrostrictive and other attractive forces exerted by the solute. This layer is surrounded by a slightly less compressed or 'structure-broken' region of water molecules (Y), distantly affected by those forces. The outermost layer is bulk water (Z), which possesses the typical tetra-coordinated hydrogen-bonded structure and is only slightly affected by any of the above forces.

Apparent molar volume (ϕ_v) and apparent specific volume (V_2) are widely used to provide information about the packing characteristics of solute molecules among water molecules. In a non-ideal solution, apparent molar volume is made up of the sum of the intrinsic volume of the solute (van der Waals volume) and the volume contribution due to solute–solute and solute–solvent interactions as shown in the equation below (Galema & Hoiland, 1991).

$$\phi_v = V_{\text{intrinsic}} + V_{\text{solute–solvent}} + V_{\text{solute–solute}}$$

Molecules which are heavily hydrated have smaller apparent molar volumes than those which are not as a result of their greater degree of interaction with water structure. Solute molecules pull in the surrounding water by electrostriction or other attractive forces, compressing the open structure of the solvent as they do so.

This paper uses apparent specific volume (V_2), which distinguishes solutes of different molecular weights, by their apparent volumes per unit mass, and therefore appears to be a more appropriate parameter for comparing the packing characteristics of sugars of different molecular architectures. Shamil, Birch, Mathlouthi and Clifford (1987) have classified apparent specific volumes as broad determinants of taste quality, with the entire human taste range lying within an V_2 range of 0.10–0.93 cm^3 g^{-1}. The V_2 values of sugar molecules lie in the region 0.60–0.64 cm^3 g^{-1}, which is central in the range of sweetness observable for most other sweet-tasting compounds (0.51–0.71 cm^3 g^{-1}). A low apparent specific volume implies extensive solute–solvent interaction, and therefore efficient hydrostatic packing of solutes among solvent molecules (Birch et al., 1997).

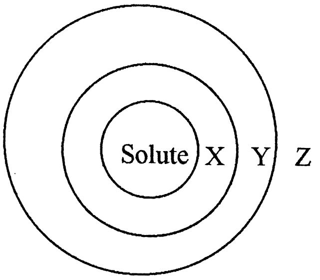

Fig. 1. Hydration model of a solute in water. X: region of immobilization of water molecules; Y: region of structure-breaking more random in organization than normal; Z: structurally normal water (based on the Frank and Wen hydration model, 1957).

Table 1
Psychophysical responses for selected bulk:intense sweetener mixtures[a]

	Bulk:intense sweetener blend	Bulk:intense sweetener ratio[b]	% synergy
Synergism	Sucrose–sodium cyclamate	75:25	12.8
	Maltitol–sodium cyclamate	50:50	27.4
	Maltitol–acesulfame K	75:25	19.1
Additivity	Sucrose–acesulfame K	75:25	−1.20
Suppression	Sucrose–aspartame	25:75	−32.8
	Maltitol–aspartame	25:75	−12.4

[a] Source: Hutteau et al. (1998).

[b] Ratios show the proportions of bulk:intense sweeteners, based on their sweetness contributions to the mixtures, contributing to maximum synergism, additivity or suppression.

Isentropic compressibility measures the extent to which the hydration layer around a molecule can be compressed and therefore reflects directly upon the extent of hydration and interaction of the solute with the surrounding solvent structure. It is a very sensitive parameter and is able to pick up fine differences in the hydration properties of solutes (Parke, Birch & Dijk, 1999; Parke, Birch, MacDougall & Stevens, 1997). The high temperature coefficient of $K_{\phi(s)}$ indicates specific solute–solvent interactions, such as hydrogen bonding, which are very temperature-dependent (Isaacs, 1981). Compressibility measurements measure the changes in the first two layers of solvent around the molecule (i.e. layers X and Y in Fig. 1) and assume that the solutes themselves do not contribute to the measured solution compressibilities. Astley, Birch, Drew, Rodger and Wilden (1998) have shown that these two hydration layers are relevant to the hydrogen bonding of sugars and water.

Solutes vary in their isentropic compressibility depending on the nature of their interaction with water (Fig. 2). Pure water has a molar compressibility of $+8.17\times10^{-4}$ cm^3 mol^{-1} bar^{-1} (Galema & Hoiland, 1991). In the case of hydration of an apolar, hydrophobic solute, the water molecules in the hydration layer will form stronger hydrogen-bonds to each other (hydrophobic hydration) and therefore the structure of the hydration layer becomes less compressible than pure water. When ions are introduced into water, they usually break the water structure by electrostriction, and the water molecules are strongly attracted to the charge centres. This causes the water structure to collapse around the ions, making them very dense and incompressible, leading to typically large negative partial molar compressibilities (-30 to -50×10^{-4} cm^3 mol^{-1} bar^{-1}) (Galema & Hoiland, 1991). The partial molar compressibilities for carbohydrates have intermediate values between those for the ionic structures and the apolar substances. This suggests that the surrounding hydrogen-bonded structure of water is only slightly disturbed in the presence of the carbohydrate. It seems that the more negative the partial isentropic compressibility is, the more the hydration layer will be disturbed compared to pure water, and the less compressible it becomes. Isentropic specific compressibility [K_2(s)] is used to compare the hydration layers of different molecules on a per gram basis.

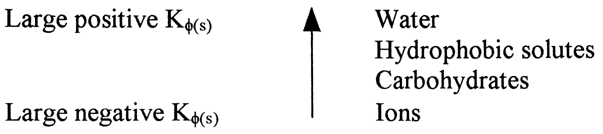

Fig. 2. Trend in isentropic apparent molar compressibility ranges of solutes in water (Source: Galema and Hoiland, 1991).

Compressibility hydration numbers (n_h) are indicative of the number of water molecules that are disturbed by the presence of the solute(s) in solution. They are derived from isentropic compressibility coefficient data and are therefore also associated with the first two layers of water around the solute molecule (refer to Fig. 1). Smaller hydration numbers are found for the solutes which disturb water structure the least. Large hydration numbers reflect extensive solute–solvent interaction. The equation used for calculating hydration numbers assumes that the hydration layer is incompressible, which is clearly not the case in this study. However, it gives an approximate overall picture of the extent of interaction of the solutes with the structure of water. Hydration numbers calculated by other methods cannot be directly compared to compressibility hydration numbers as they measure water disturbance of a single hydration sheath around the solute.

2. Materials and methods

Chemicals used in this experiment were reagent grade. Sucrose, maltitol and sodium cyclamate were obtained from Sigma Chemical Co., Poole, UK; Aspartame and acesulfame K were obtained from Nutrasweet, and Hoechst, Switzerland, respectively. Water used for solution studies was HPLC grade. Sucrose, maltitol and sodium cyclamate were recrystallised three times in ethanol–water mixture. All the substances were dried in a vacuum oven at 60°C, over phosphorus pentoxide as drying agent, for 24 h before being used. Solutions were made up w/w, and all measurements were carried out at 20°C and in duplicate. Reported results are averages of all measurements and are accurate to 4 significant figures.

The density and sound velocity of the mixtures were first measured and these parameters were used to calculate apparent volumes, isentropic compressibilities and hydration numbers.

Density and sound velocity measurements were determined using an Anton Paar Density Sound Analyser (DSA 48) from Paar Scientific Ltd, Raynes Park, London. Temperature was maintained at 20 ± 0.1°C. The density of the sample was measured from the period of oscillation of an oscillating U-tube. The sound velocity was calculated from the propagation speed of ultrasonic pulses in a known distance within the sample in the measuring cell. The instrument was calibrated once using air and distilled water. Density and sound velocity measurements were accurate to $\pm1\times10^{-4}$ g cm^{-3} and ±1 ms^{-1}, respectively.

Apparent specific volume for single solutes, V_2 (cm^3 g^{-1}), is obtained from the following equation

$$V_2 = \phi_v / M_2 \tag{1}$$

where M_2 = molecular weight of solute (g mol^{-1})
ϕ_v is the apparent molar volume (cm^3 mol^{-1}) and is obtained from density measurements using Eq. (2) below:

$$\phi_v = 1000(d_0 - d)/mdd_0 + M_2/d \qquad (2)$$

where d_0 = density of water at one temperature (g cm^{-3})
d = density of solution at the same temperature (g cm^{-3})
m = molality of the solution (mol per kg of water).

Isentropic apparent specific compressibility for single solutes [$K_{2(s)}$ cm^3 g^{-1} bar^{-1}] is obtained from Eq. (3).

$$K_{2(s)} = K_{\phi(s)}/M_2 \qquad (3)$$

where $K_{\phi(s)}$ is the isentropic apparent molar compressibility (cm^3 mol^{-1} bar^{-1}) and is calculated from both density and sound velocity values using Eq. (4):

$$K_{\phi(s)} = 1000(\beta_s - \beta_{so})/md + \beta_s\phi_v \qquad (4)$$

where β_s = isentropic compressibility coefficient of solution (bar^{-1})
β_{so} = isentropic compressibility coefficient of water (bar^{-1})

Isentropic compressibility coefficients are calculated from Eq. (5)

$$\beta_s = 100/u^2 d \qquad (5)$$

where u = sound velocity of solution (m s^{-1}).

Compressibility hydration number for single solutes (n_h) is calculated from the following equation:

$$n_h = (n_w/n_s)(1 - \beta_s/\beta_{so}) \qquad (6)$$

where n_w = number of moles of water (mol per kg solution)
n_s = number of moles of solute (mol per kg solution)

Apparent volumes, isentropic apparent compressibilities and compressibility hydration numbers for binary mixtures are calculated on a total solute basis using Eqs. (1)–(6) above, with the following modifications (Ward & Millero, 1974)

m is replaced by m_T = total molality of the solution (mol per kg water)
M_2 is replaced by M_T = weighted mean molecular weight of solutes (g mol^{-1}) = $\sigma(y_x M_x)$

where M_x is the molecular weight of component x (g mol^{-1})
y_x is the molal weighting factor $= m_x/m_T$, where m_x is the molality of component x in solution (mol per kg of water)
n_s is replaced by n_T = total number of moles of solute (mol per kg solution).

Partial specific volumes (V_2^0), isentropic partial specific compressibilities [$K^0_{2(s)}$] and partial compressibility hydration numbers (n^0_h) are obtained at infinite dilution by extrapolating the best fit to the curves to zero concentration. Partial values provide information about the hydration of solutes in an ideal solution, that is where no solute–solute interaction is present.

Three sets of solution measurements were performed on the binary mixtures of bulk and intense sweeteners as described below.

2.1. Experiment 1 — binary mixtures of bulk and intense sweeteners

Aim: To relate the psychophysical characteristics (additivity, synergism or suppression) of specific mixtures investigated by Hutteau et al. (1998) to their solution properties.

The concentrations of the mixtures were the same as those investigated by Hutteau et al. (1998). The concentrations of the individual components of the binary mixtures used were such that together they would give a total sweetness intensity equivalent to a 10% sucrose solution. Five different ratios of bulk:intense sweeteners were used (0:100, 25:75, 50:50, 75:25 and 100:0), with each one contributing to that proportion of the total sweetness of the mixture (based on the results of Portmann & Kilcast, 1996). Thirty different mixture combinations were analysed (refer to Table 2).

2.2. Experiment 2—binary mixtures of bulk and intense sweeteners, intense sweetener variable

Aim: To compare the effects of the individual intense sweeteners on the solution while maintaining the concentration of the bulk sweetener constant.

The mid-point of the concentrations reported for each of the bulk sweeteners in experiment 1 is used in conjunction with the range of concentrations for each intense sweetener used (Table 3).

2.3. Experiment 3—binary mixtures of bulk and intense sweeteners, bulk sweetener variable

Aim: To assess the effect of increasing concentrations of bulk sweeteners on the solution while keeping the concentrations of the intense sweeteners constant.

As in experiment 2, the concentrations of the intense sweeteners used are the mid-points of the range tested, and the range of concentrations for the bulk sweeteners is the same as those used in experiment 1 (Table 4).

3. Results

Tables 5 and 6 give the solution measurements for experiments 1–3 for sucrose:intense sweetener and maltitol:intense sweetener mixtures, respectively, calculated on a "total solute" basis.

3.1. Experiment 1—binary mixtures of bulk and intense sweeteners

Results of experiment 1 are displayed in Fig. 3–5. As the proportion of bulk sweeteners goes up and those of the intense sweeteners decrease, there is a general increase in the apparent specific volumes (except for bulk:aspartame mixtures) and isentropic compressibilities. A two-factor ANOVA has, however, not shown any significant differences between the mixtures. The general increase in apparent specific volumes can be accounted for by the corresponding increase in total solute concentration (and therefore, reduced solute-solvent affinity) across the mixtures, largely dictated by the vast molar excess of the bulk over the intense sweeteners.

The apparent specific volumes of all the mixtures lie between 0.60 and 0.64 cm^3 g^{-1}, which Birch (1991) reported for substances exhibiting a clean sweet taste (refer to Fig. 3a and b). The percentage increase in isentropic apparent specific compressibilities across the mixtures from 25:75 to 100:0 bulk:intense sweeteners, are 27.4, 38.0 and 171.2% for mixtures of sucrose with acesulfame K, aspartame and sodium cyclamate, respectively, and 51.6, 92.9 and 113.4% for mixtures of maltitol with the intense sweeteners. The increase in isentropic compressibility shows that the solutes pack into the solvent structure without causing much disruption to the original three-dimensional structure of water. Sodium cyclamate mixtures therefore show the largest change in compressibility, as its concentration is decreased, followed by aspartame (Fig. 4a and b). Sodium cyclamate mixtures are also the least compressible of all the mixtures (-12.36×10^{-6} to -4.556×10^{-6} cm^3 g^{-1} bar^{-1} for sucrose–cyclamate and -9.287×10^{-6} to -4.353×10^{-6} cm^3 g^{-1} bar^{-1} for maltitol–cyclamate, whereas the ranges of specific compressibilities reported

Table 2
Experiment 1: concentrations of bulk:intense sweetener used in binary mixtures

Solution no.	% sweetness contribution (bulk: intense sweetener)	Sucrose (% w/w)	Maltitol (% w/w)	Acesulfame K (% w/w)	Aspartame (% w/w)	Sodium cyclamate (% w/w)
A	0:100	0	0	0.0867	0.12333	0.440
B	25:75	1.92	3.55	0.0448	0.1000	0.300
C	50:50	4.55	7.12	0.0248	0.0755	0.198
D	75:25	7.18	10.70	0.0118	0.0522	0.098
E	100:0	10.00	13.50	0	0	0

Table 3
Experiment 2: concentrations of bulk:intense sweeteners used in "constant bulk/variable intense sweetener" binary mixtures

Solution no.	Sucrose (% w/w)	Maltitol (% w/w)	Acesulfame K (% w/w)	Aspartame (% w/w)	Sodium cyclamate (% w/w)
F	4.55	7.12	0	0	0
G	4.55	7.12	0.0118	0.0522	0.098
H	4.55	7.12	0.0248	0.0755	0.198
I	4.55	7.12	0.0448	0.1000	0.300
J	4.55	7.12	0.0867	0.1333	0.440

Table 4
Experiment 3: concentrations of bulk:intense sweetener used in "variable bulk/constant intense sweetener" binary mixtures

Solution no.	Sucrose (% w/w)	Maltitol (% w/w)	Acesulfame K (% w/w)	Aspartame (% w/w)	Sodium cyclamate (% w/w)
K	0	0	0.0248	0.0755	0.198
L	1.92	3.55	0.0248	0.0755	0.198
M	4.55	7.12	0.0248	0.0755	0.198
N	7.18	10.70	0.0248	0.0755	0.198
O	10.00	13.50	0.0248	0.0755	0.198

for the other intense sweeteners with sucrose and maltitol are -6.286×10^{-6} to -4.556×10^{-6} $\text{cm}^3\ \text{g}^{-1}\ \text{bar}^{-1}$ and -8.397×10^{-6} to -4.353×10^{-6} $\text{cm}_3\ \text{g}^{-1}\ \text{bar}^{-1}$, respectively). The results show varying degrees, and probably types, of interaction between the solutes and water, with sodium cyclamate being most interactive, followed by aspartame and acesulfame K. A fall in hydration number (Fig. 5a and b) is usually associated with increasing solute concentration, and this also provides good evidence of the dominance of the bulk sweeteners in the mixtures.

Intense sweeteners are known to disrupt the homogeneity of water structure profoundly (Mathlouthi et al., 1993; Parke & Birch, 1998), but the introduction of

Table 5
Apparent specific volumes (V_2), isentropic apparent specific compressibilities [$K_{2(s)}$], and compressibility hydration numbers (n_h) of binary mixtures of sucrose (Suc) with acesulfame K (AceK), aspartame (Asp) and sodium cyclamate (Cyc)

	Suc–AceK			Suc–Asp			Suc–Cyc		
Solution no.	V_2 ($\text{cm}^3\ \text{g}^{-1}$)	$K_{2(s)}$ ($\text{cm}^3\ \text{g}^{-1}\ \text{bar}^{-1}$)	n_h	V_2 ($\text{cm}^3\ \text{g}^{-1}$)	$K_{2(s)}$ ($\text{cm}^3\ \text{g}^{-1}\ \text{bar}^{-1}$)	n_h	V_2 ($\text{cm}^3\ \text{g}^{-1}$)	$K_{2(s)}$ ($\text{cm}^3\ \text{g}^{-1}\ \text{bar}^{-1}$)	n_h
Experiment 1. Sucrose increasing, intense sweeteners decreasing									
A	0.529	−1.761E-05	11.19	0.736	−1.201E-05	13.95	0.605	−4.164E-05	17.31
B	0.617	−5.806E-06	13.83	0.623	−6.286E-06	14.25	0.615	−1.236E-05	15.21
C	0.616	−5.448E-06	13.73	0.618	−5.640E-06	13.87	0.614	−7.392E-06	14.15
D	0.616	−4.901E-06	13.45	0.617	−5.017E-06	13.51	0.161	−5.705E-06	13.66
E	0.616	−4.55E-06	13.09	0.616	−4.556E-06	13.09	0.616	−4.556E-06	13.09
Experiment 2. Sucrose constant, intense sweetener increasing									
F	0.616	−5.522E-06	13.92	0.616	−5.522E-06	13.92	0.616	−5.522E-06	13.92
G	0.615	−5.489E-06	13.77	0.618	−5.597E-06	13.86	0.615	−6.680E-06	14.07
H	0.616	−5.448E-06	13.73	0.618	−5.640E-06	13.87	0.614	−7.392E-06	14.15
I	0.614	−5.625E-06	13.73	0.619	−5.625E-06	13.87	0.616	−8.240E-06	14.31
J	0.615	−5.565E-06	13.64	0.620	−5.614E-06	13.86	0.616	−8.982E-06	14.34
Experiment 3. Sucrose increasing, intense sweetener constant									
K	0.528	−1.766E-05	12.68	0.737	−1.214E-05	18.05	0.605	−4.198E-05	17.81
L	0.615	−6.068E-06	13.99	0.624	−6.089E-06	14.12	0.606	−1.103E-05	15.03
M	0.616	−5.448E-06	13.73	0.618	−5.640E-06	13.87	0.614	−7.392E-05	14.15
N	0.617	−4.869E-06	13.43	0.618	−4.945E-06	13.49	0.616	−5.853E-06	13.58
O	0.617	−4.310E-06	13.08	0.618	−4.364E-06	13.13	0.617	−4.849E-06	13.14

Table 6
Apparent specific volumes (V_2), isentropic apparent specific compressibilities [$K_{2(s)}$], and compressibility hydration numbers (n_h) of binary mixtures of maltitol (Mal) with acesulfame K (AceK), aspartame (Asp) and sodium cyclamate (Cyc)

	Mal–AceK			Mal–Asp			Mal–Cyc		
Solution no.	V_2 ($\text{cm}^3\ \text{g}^{-1}$)	$K_{2(s)}$ ($\text{cm}^3\ \text{g}^{-1}\ \text{bar}^{-1}$)	n_h	V_2 ($\text{cm}^3\ \text{g}^{-1}$)	$K_{2(s)}$ ($\text{cm}^3\ \text{g}^{-1}\ \text{bar}^{-1}$)	n_h	V_2 ($\text{cm}^3\ \text{g}^{-1}$)	$K_{2(s)}$ ($\text{cm}^3\ \text{g}^{-1}\ \text{bar}^{-1}$)	n_h
Experiment 1. Maltitol increasing, intense sweeteners decreasing									
A	0.529	−1.761E-05	11.19	0.736	−1.201E-05	13.95	0.605	−4.164E-05	17.31
B	0.626	−6.599E-06	14.43	0.630	−8.397E-06	15.32	0.625	−9.287E-06	14.80
C	0.627	−5.922E-06	14.11	0.628	−6.066E-06	14.20	0.626	−6-508E-06	14.08
D	0.627	−5.155E-06	13.64	0.628	−5.100E-06	13.62	0.627	−5.410E-06	13.65
E	0.629	−4.353E-06	13.22	0.629	−4.353E-06	13.22	0.629	−4.353E-06	13.22
Experiment 2. Maltitol constant, intense sweetener increasing									
F	0.628	−5.565E-06	14.04	0.628	−5.565E-06	14.04	0.628	−5.565E-06	14.02
G	0.628	−5.740E-06	14.07	0.629	−5.796E-06	14.11	0.627	−6/273E-06	14.14
H	0.627	−5.922E-06	14.11	0.682	−6.066E-06	14.20	0.626	−6.508E-06	14.08
I	0.627	−5.822E-06	14.03	0.630	−5.837E-06	14.12	0.627	−6.972E-06	14.15
J	0.627	−5.690E-06	13.93	0.630	−5.748E-06	14.08	0.627	−7.459E-06	14.16
Experiment 3. Maltitol increasing, intense sweetener constant									
K	0.528	−1.766E-05	11.95	0.737	−1.214E-05	18.11	0.605	−4.198E-05	16.67
L	0.626	−6.742E-06	14.56	0.630	−6.706E-06	14.64	0.625	−8.518E-06	14.79
M	0.627	−5.922E-06	14.11	0.628	−6.066E-06	14.20	0.626	−6.508E-06	14.08
N	0.629	−4.917E-06	13.56	0.630	−4.960E-06	13.59	0.628	−5.497E-06	13.63
O	0.629	−4.391E-06	13.19	0.630	−4.373E-06	13.21	0.628	−4.743E-06	13.19

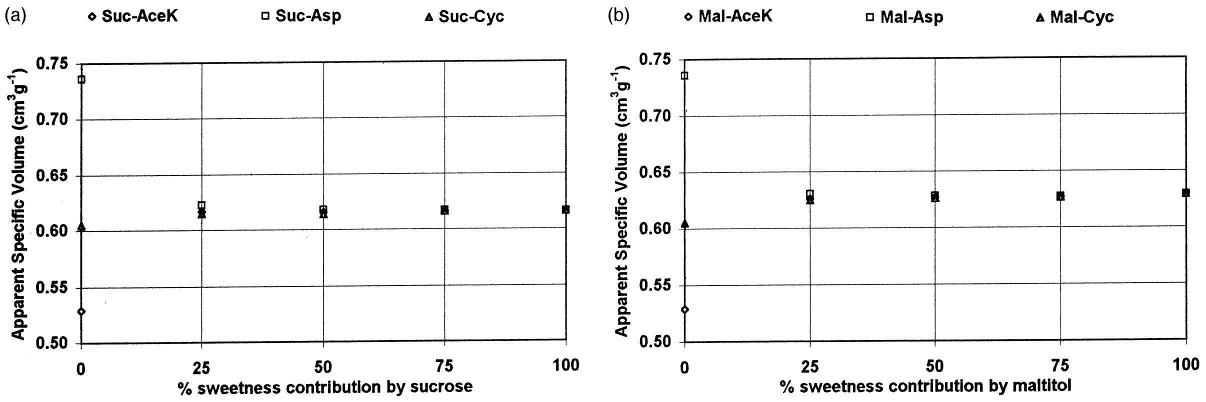

Fig. 3. Apparent specific volumes of binary mixtures of bulk:intense sweeteners (experiment 1).

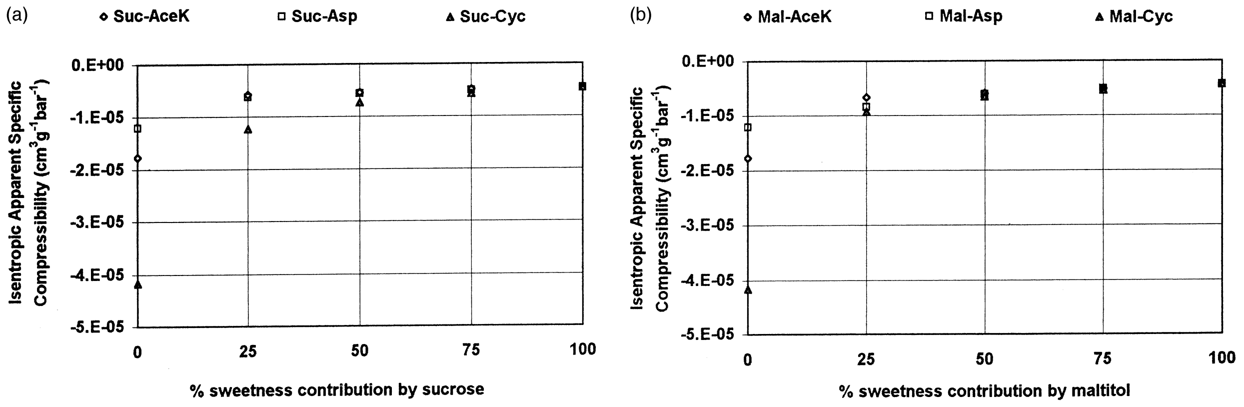

Fig. 4. Isentropic apparent specific compressibility of binary mixtures of bulk:intense sweeteners (experiment 1).

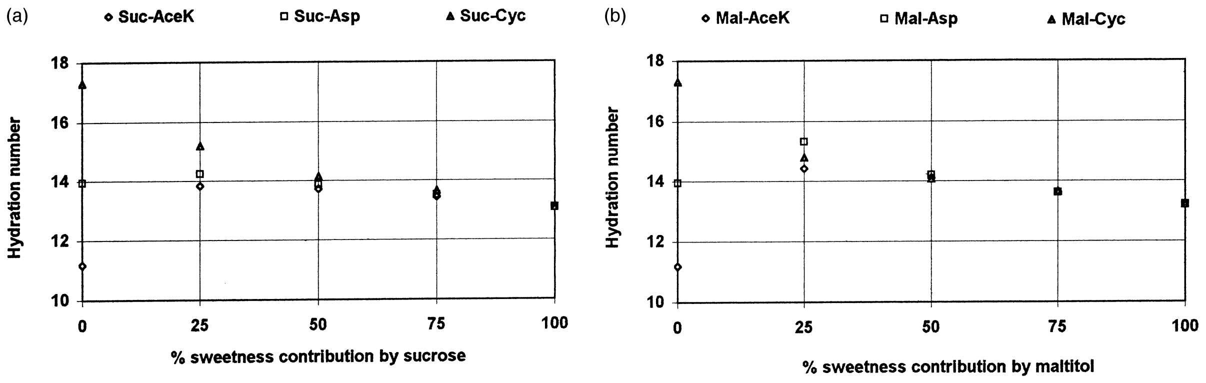

Fig. 5. Compressibility hydration numbers of binary mixtures of bulk:intense sweeteners (experiment 1).

relatively few molecules of intense sweeteners to the mixtures, as opposed to the vast molar excess of bulk sweeteners, would not be expected to affect water structure to a large extent. Therefore the properties of the mixtures of bulk:intense sweeteners seem to be dominated by the properties of the bulk sweeteners, yielding measurements close to the partial values of the latter (Fig. 3–5). When higher concentrations of the intense sweeteners are used (as in the 25:75 bulk:intense sweeteners), the effect on surrounding water structure is more pronounced, as shown by isentropic compressibility data. Acesulfame K and sodium cyclamate are salts which possess small apparent specific volumes and large negative specific compressibilities (refer to Table 7), typical of their ionic structures, and arising from the extensive disturbance of water structure around them from electrostrictive forces. Aspartame has a higher specific volume and is more compressible than the other intense sweeteners, which accords with its hydrophobicity and its hydrophobic mode of hydration.

Both sodium cyclamate and acesulfame K have a

Table 7
Solution properties of mixtures exhibiting maximum psychophysical responses[a]

	ϕ_v (cm^3 mol^{-1})	V_2 (cm^3 g^{-1})	$K_{\phi(s)}$ (cm^3 mol^{-1} bar^{-1})	$K_{2(s)}$ (cm^3 g^{-1} bar^{-1})	n_h
Synergistic mixtures					
Suc–cyc (75:25)	208.9	0.616	-1.934×10^{-3}	-5.705×10^{-6}	13.65
Mal–cyc (50:50)	211.4	0.626	-2.197×10^{-3}	-6.508×10^{-6}	14.08
Mal-aceK (75:25)	215.6	0.627	-1.772×10^{-3}	-5.155×10^{-6}	13.64
Additive mixtures					
Suc–aceK (75:25)	210.7	0.616	-1.676×10^{-3}	-4.901×10^{-6}	13.45
Suppressive mixtures					
Suc–asp (25:75)	211.4	0.623	-2.134×10^{-3}	-6.286×10^{-6}	14.25
Mal–asp (25:75)	216.0	0.630	-2.878×10^{-3}	-8.397×10^{-6}	15.32

[a] Values in brackets represent the proportions of bulk:intense sweeteners at which those maxima occur.

molecular weight of 201.2 g mol^{-1} and they both possess the NH–SO$_2$ moiety postulated by Shallenberger (1993) to be responsible for their sweet taste; therefore a comparison of the solution properties of the mixtures might prove interesting. A two-factor ANOVA has shown that there is no significant difference between the mixtures sucrose–acesulfame K, maltitol–acesulfame K, sucrose–sodium cyclamate and maltitol–sodium cyclamate for their observed apparent specific volumes and isentropic apparent specific compressibilities. But close inspection of the data shows that the specific compressibilities of bulk-sodium cyclamate mixtures are more negative, that is, less compressible, than bulk sweetener-acesulfame K mixtures (see Tables 5 and 6). It is well documented that the potassium ion and the sodium ion affect the structuring of water differently (Parke & Birch, 1999). Surface tension studies by Mathlouthi, Hutteau and Angiboust (1996) have also shown the opposite effects of the sodium ion and the potassium ion on the structure of water. The sodium ion has been shown to disturb water structure, which explains its large negative compressibility values, whereas the potassium ion enhances the structuring of surrounding water molecules and is associated with negative hydration.

3.2. Experiment 2—binary mixtures of bulk and intense sweeteners, intense sweetener variable

By keeping the bulk sweetener constant and increasing the concentration of the intense sweetener (Tables 5 and 6), the percentage changes reported for the apparent specific volumes from 25:75 to 100:0 bulk:intense sweeteners are less than 1% for all the mixtures. These small changes across each bulk:intense sweetener mixture result from the low concentration ranges of intense sweeteners used.

In mixtures of aspartame with the bulk sweeteners, increasing the concentration of aspartame increases apparent specific volumes, as in experiment 1, possibly as a result of the hydrophobic effect of aspartame. Water around hydrophobic solutes undergoes a structural rearrangement with strong intermolecular hydrogen-bonds, which increases the overall volume of the solution (Parke & Birch, 1999). At the same time, the bulk solutes are likely to aggregate (Mora-Guitierrez & Baianu, 1989) and also contribute to the increase in volume. When acesulfame K and sodium cyclamate are mixed with the bulk sweeteners, apparent specific volumes tend to decrease. Being ionic in nature, they are expected to dissociate in solution (Cohn, McMeekin, Edsall & Blanchard, 1934), and through electrostrictive effects, pull the hydration water closer to them. Increasing concentrations of these intense sweeteners also increased the packing efficiency of the solutes in solution.

There is, however, a larger change observable for isentropic specific compressibilities than for specific volumes. In general, isentropic compressibility values tend to become more negative with increasing concentration of intense sweetener, and the biggest change reported is for sodium cyclamate [$K_{2(s)}$ values decrease from -5.522×10^{-6} to -8.982×10^{-6} cm^3 g^{-1} bar^{-1} (62.7%) for sucrose mixtures, and -5.565×10^{-6} to -7.459×10^{-6} cm^3 g^{-1} bar^{-1} (34.0%) for maltitol mixtures]. Isentropic compressibilities for the other mixtures with sucrose and maltitol vary from -5.522×10^{-6} to -5.614×10^{-6} cm^3 g^{-1} bar^{-1} and -5.565×10^{-6} to -5.748×10^{-6} cm^3 g^{-1} bar^{-1}, with a percentage change of 1.7 and 0.8% for mixtures of sucrose with aspartame and acesulfame K, and 3.3 and 2.2% for mixtures of maltitol with the intense sweeteners, respectively. Bulk:sodium cyclamate mixtures still exhibit the largest negative compressibility values (i.e. the smallest compressibilities) ($p < 0.001$). As explained earlier, the sodium salt disrupts water structure to a large extent, making the hydration layer around it less compressible. Because of the low molality ranges of acesulfame K and aspartame used, the changes in their solution properties, as their concentrations are increased, are fairly small compared with sodium cyclamate.

Generally we would expect a fall in compressibility hydration number as the concentration of solutes in solution is increased. However, as the concentration of sodium cyclamate is increased, the number of water molecules that are disturbed also increases, showing the disruptive effect of the cyclamate molecules on the surrounding water structure. In the case of acesulfame K and aspartame, the concentrations used are too small for any significant changes to be detected.

This experiment clearly demonstrates the effect of concentration of intense sweetener on the mixtures: the higher the concentration used, the more marked the effect on water structure, that is, disruption of water–water hydrogen bonding. However, it is important to note that the large change accompanying the concentration change of sodium cyclamate is not solely the result of the high concentrations used, but the inherent nature of sodium cyclamate (Parke & Birch, 1999). Sodium cyclamate has been shown to have an unusually low isentropic apparent specific compressibility (-43.24×10^{-6} cm^3 g^{-1} bar^{-1}) compared to the other intense sweeteners studied (aspartame -12.31×10^{-6} and acesulfame K -17.68×10^{-6} cm^3 g-1 bar $^{-1}$). Mixtures of the bulk sweeteners with aspartame are significantly different from the others ($p<0.001$) in that they have bigger apparent specific volumes which is expected from the size and hydrophobicity of aspartame.

3.3. Experiment 3—binary mixtures of bulk and intense sweeteners, bulk sweetener variable

When the intense sweeteners are kept constant and the bulk sweeteners varied (Tables 5 and 6), results once again show that mixtures with sodium cyclamate are much less compressible than with the other intense sweeteners. Compressibilities decrease from -11.03×10^{-6} to -4.849×10^{-6} cm^3 g^{-1} bar^{-1} for sucrose:cyclamate and -8.518×10^{-6} to -4.743×10^{-6} cm^3 g^{-1} bar^{-1} for maltitol:cyclamate mixtures (overall range reported for mixtures of the other intense sweeteners with sucrose is -6.090×10^{-6} to -4.310×10^{-6} cm^3 g^{-1} bar^{-1}, and -6.74×10^{-6} to -4.37×10^{-6} cm^3 g^{-1} bar^{-1} for maltitol mixtures). These represent 127.4 and 79.6% changes in specific compressibility across sucrose:cyclamate and maltitol:cyclamate mixtures, respectively (compared with approximately 40.0 and 50.0% change for the other mixtures of intense sweeteners with sucrose and maltitol).

Increasing the bulk sweetener concentration also reduces compressibility hydration numbers, showing fewer water molecules being disturbed as the solute–solvent interactions are replaced by solute–solute interactions. The hydration numbers for all the mixtures are comparable, as they are governed by the molar excess of bulk sweeteners.

The apparent specific volumes of the mixtures of bulk sweeteners with acesulfame K and sodium cyclamate, increase as the concentration of the bulk sweetener is raised whereas, when aspartame is used, the apparent specific volume is found to decrease slightly. As a result of the low concentrations of the intense sweeteners used, the effects of the intense sweeteners on the solution behaviour of the bulk solutes are not very pronounced. The bulk sweeteners therefore dominate the properties of the mixture. The unusual behaviour of bulk:aspartame mixtures can again be attributed to the hydrophobicity of the intense sweetener.

This experiment shows that the higher concentrations of bulk sweeteners dictate the properties of the solution, a phenomenon also demonstrated by Mathlouthi and Portmann (1994) in mixtures of bulk and artificial sweeteners. No significant difference has been found among the mixtures with respect to any of the parameters studied.

4. Discussion

A brief summary of the results of experiments 1 to 3 is given below:

- Experiment 1—Increasing bulk sweeteners and decreasing intense sweeteners

  The properties of the bulk sweeteners dominate the properties of the mixtures. Slight differences in solution properties are attributable to the influence of the intense sweeteners on the bulk sweeteners. Mixtures containing sodium cyclamate seem to have the most profound influence on the structure of water.
- Experiment 2—Bulk sweetener constant, increasing intense sweetener concentrations

  The effects of the intense sweeteners on the solutions become apparent, especially when higher concentrations are used. Sodium cyclamate mixtures show significantly different isentropic compressibilities from the other mixtures.
- Experiment 3—Intense sweetener constant, bulk sweetener concentration increasing.

  The concentrations of intense sweeteners are too low for their effect on the hydration of the mixtures to become significant. Therefore, the bulk sweeteners dominate the properties of the mixtures.

Table 7 summarises the solution properties of binary mixtures of sucrose or maltitol with intense sweeteners (experiment 1), at the proportions producing maximum synergy, addition or suppression.

In experiment 1, the following two points explain the dominance of the bulk sweetener properties upon those of the mixtures:

1. The vast molar excess of bulk sweeteners over the intense sweeteners used.
2. The low molality ranges of acesulfame K and aspartame (0.001–0.005 mol kg^{-1}) as opposed to the higher concentration range of sodium cyclamate (0.005–0.02 mol kg^{-1}) used.

However, across all three experiments, there are distinct patterns of hydration of bulk:intense sweeteners as shown by their solution properties. At the low concentrations of use of the intense sweeteners, the differences in hydration from using one intense sweetener to the other may not be highly significant, but slight differences in their behaviour can be observed on close comparison of their solution property trends. It has also been shown that in mixtures, the presence of one species may alter the solution properties of another sufficiently to affect the overall properties of the mixture as well as effect a taste change (Birch, Ogunmoyela & Munton, 1982; Parke, Birch, MacDougall & Stevens, 1997). The properties of the mixtures dealt with in this study, although dominated by the bulk sweeteners, seem to be influenced by the nature of the intense sweetener present.

The apparent specific volumes of the mixtures in all three experiments tend to decrease in the order bulk, aspartame, acesulfame K and sodium cyclamate. For both sucrose:intense sweetener and maltitol:intense sweetener mixtures, apparent specific volumes generally increase from synergistic to suppressive mixtures (Table 7), implicating the size of the component solute fraction in the psychophysical response achieved (refer to Table 8 for molecular weights of substances). The idea of an optimum apparent specific volume for sweetness has previously been discussed by Birch (1991). Presumably, the smaller the V_2, the more efficient is the recruitment of receptors, and the deeper in the lingual epithelium molecules may travel. This results from prolonged interaction with water, which is the vehicle to receptor access.

Experiments 1 and 2 show that isentropic specific compressibilities become larger negative (that is, mixtures become less compressible) from aspartame to acesulfame K and sodium cyclamate, as would be expected from the hydration of the pure intense sweeteners in solution (Table 7). This shows the influence of the individual intense sweeteners on the hydration of the solutes in solution. Compressibility hydration numbers across all the experiments also fall, in mixtures of bulk sweeteners with cyclamate, aspartame and acesulfame K, and in that order. Bulk:sodium cyclamate mixtures seem to be more interactive with water structure than the other bulk:intense sweetener mixtures whereas mixtures containing acesulfame K seem to show the least disturbance of the surrounding water structure, as is expected from their opposed hydration properties conferred by the sodium and potassium ions, respectively (Parke & Birch, 1999).

Table 8
Partial specific volumes (V_2^0), isentropic partial specific compressibilities [$K_{2(s)}^0$] and partial compressibility hydration numbers (n_h^0) of selected bulk and intense sweeteners[a]

	Mol wt (g mol^{-1})	V_2^0 (cm^3 g^{-1})	$K_{2(s)}^0 \times 10^{-6}$ (cm^3 g^{-1} bar^{btl1})	n_h^0
Maltitol	344.3	0.627	−6.874	14.86
Sucrose	342.3	0.615	−6.370	14.33
Acesulfame K	201.2	0.529	−17.27	10.28
Aspartame	294.3	0.738	−12.31	16.70
Sodium cyclamate	201.2	0.605	−43.24	17.31

[a] Source: Parke and Birch, 1999.

Although the general trends in isentropic compressibilities are as elucidated in the experiments above, across both sucrose:intense sweetener and maltitol: intense sweetener mixtures producing either synergy, additivity or suppression, specific compressibility values are the lowest (that is, solutions are the least compressible) for suppressive mixtures, and largest for those exhibiting additivity (as shown in Table 7). Synergistic mixtures have compressibilities in between those two. Hydration numbers seem to follow the same pattern as compressibilities, as they show the number of water molecules that are displaced as a result of the change in the hydration layer around the solutes. The range of values of isentropic apparent specific compressibilities reported for the mixture with the same psychophysical characteristics is given below.

Synergistic mixtures	-6.508×10^{-6} to -5.155×10^{-6} cm^3 g^{-1} bar^{-1}
Additive mixtures	-4.917×10^{-6} to -4.135×10^{-6} cm^3 g^{-1} bar^{-1}
Suppressive mixtures	-8.397×10^{-6} to -6.286×10^{-6} cm^3 g^{-1} bar^{-1}

A more highly compressed hydration layer may not necessarily result in enhanced sweetness. These results seem to show that there is an optimum range of compressibility in which the hydration layer must reside to produce synergistic taste responses. However, the nature of the molecules in solution seems highly significant, for instance, their size and shape will affect their accession to and fit onto the receptor; their hydrophobicity and hence their ability to partition between saliva and the taste receptor membrane will affect their orientation onto the receptors.

Maltitol:intense sweetener mixtures have larger apparent volumes and more negative compressibility values than sucrose:intense sweeteners (Table 7). Chavez Lopez and Birch (1997) have shown that polyols have "structure-breaking" effects resulting from their open linear structures (compared to the cyclic structures encountered in sugars). This presents a large surface area to the solvent for interaction to occur and therefore affects packing characteristics in water.

4.1. Proposed mechanisms of transfer of taste molecules to receptors

Table 8 shows that pure aspartame has the biggest apparent specific volume, and is the most compressible, of the intense sweeteners studied, and this emanates from its hydrophobic mode of hydration in water. In binary mixtures with bulk sweeteners, the solutions also show the largest apparent specific volumes and most compressible hydration layers. These results are in agreement with Mathlouthi and Portmann (1994), who have also shown that traces of aspartame raise the partial specific volumes of bulk sweeteners in solution. The hydrophobicity of the aspartame molecules is thought to enhance aggregation of the hydrophilic sugar molecules (Mora-Guitierrez & Baianu, 1989), which then form strong intermolecular hydrogen bonds between themselves. Water around the aspartame molecules is also expected to take part in hydrophobic hydration. This interplay of hydrogen-bonds between sugar–water and water–water gives rise to larger apparent specific volumes and smaller compressibility values than for the pure bulk sweeteners in solution. Because of the vast molar excess of the bulk sweeteners used over the aspartame, the properties of the mixtures are effectively governed by the way the bulk sweeteners pack within water. The solution properties observed for the mixture are therefore inevitably closer to those of the pure bulk sweeteners than to the pure intense sweeteners used.

Sodium cyclamate has a stronger disruptive effect on water structure as shown by its low apparent specific volume and isentropic compressibility (Table 8). And its influence on the hydration behaviour of mixtures is high. The Na^+ ions attract water of hydration by electrostriction and cause the collapse of the solvation layer around the solute molecules. The net result is a reduction in volume and a more compressed solvation layer around the molecules. Very little hydrophobic hydration is thought to take place as a result of the hydrophilic nature of sucrose and maltitol, and the essentially hydrophilic behaviour of the sodium cyclamate.

Bulk sweetener combinations with acesulfame K show additivity in their sweetness response. The concentration of acesulfame K used is comparable to that of aspartame, and the use of very small amounts of acesulfame K will not be expected to affect the hydration of the bulk solutes in solution to any significant extent. Acesulfame K has both hydrophilic and hydrophobic sides on its molecule; Mathlouthi and Portmann (1994) classify acesulfame K as being essentially hydrophilic from its relatively high solubility and the presence of the CH_3 group in its formula. When acesulfame K is mixed with bulk sweeteners, some hydrophobic hydration is expected leading to very little clustering of the bulk sweeteners (because of the relatively few acesulfame K molecules in solution as well as their small hydrophobic character). Specific hydration of both intense sweetener and the remaining bulk sweetener molecules also takes place. The collapse of water caused by the K^+ ions in solution is less than by Na^+ ions, because K^+ ions have been associated with negative hydration (in which the lifetime of hydrogen bonds in the vicinity of the ion is less than around a water molecule, resulting in an increase of water mobility). Negative hydration actually promotes water structure. The overall effect on water is an increase in the size; hence there is a more compressible hydration layer around the solutes in solution, compared to the hydration layer of the pure bulk sweetener alone.

The dissociation of sodium cyclamate causes increased interaction of the solutes with water structure as explained earlier. Sodium cyclamate molecules, on their own, are expected to reach the sweet receptors, because of their strong interaction with water. Pure bulk sweeteners, which have large compressibility values compared to intense sweeteners, would not be expected to travel far into the lingual epithelium, but would instead occupy receptors corresponding to the middle of the sweetness range ($0.61\ cm^3\ g^{-1}$). But in mixtures with sodium cyclamate, if indeed their compressibility is reduced (implying they become more interactive with water), they would travel deeper and to more receptors. Because of the effect of the salt structure on the bulk sweetener, the sweetness intensity is higher than what would be perceived if the sweeteners were tasted individually. This may give synergism.

Because of the effect of hydrophobic hydration of aspartame on the bulk sweeteners, which causes clustering of the sugars, we can expect fewer "free" sugar molecules to be available to reach the receptors. The aspartame molecules are also bulky and hydrophobic (and are the most compressible of all the intense sweeteners), and hence may not travel too far into the epithelium. They will preferentially remain near the surface. This means less sweet receptors being occupied and, as a result, suppression occurs. Moreover, the high apparent specific volume of aspartame approaches the bitter-sweet borderline for taste and non-sweet tastes are known to interfere with sweet perception.

Acesulfame K, being ionic, may be expected to collapse water around some of the bulk sweeteners, so that more molecules are able to reach the receptors. However, the concentrations used are much lower than those of the sodium cyclamate, and therefore the electrostrictive effects would be expected to be less. In addition, both solutes are more hydrophobic than the cyclamate, which would affect their packing characteristics in water. The result is an additive response with sucrose and synergism with maltitol.

These deductions are based on the interpretation of sweet taste chemoreception, which assumes that the sweet receptors lie at different depths in the lingual epithelium (Birch, Karim, Chavez & Morini, 1993). Receptors lie deeper in the tongue surface in the order bitter to sweet to sour to salty. Only those molecules which have the optimum stereochemical structure, size, shape and the right hydrophile–lipophile balance will be able to travel far enough to initiate the sweet response. If more sweet taste molecules access more taste receptors, this could result in an increase in the sweetness intensity perceived. Rapidity of accession is also dependent on the extent and type of interaction with water. The more hydrophilic sweeteners which interact more with water structure (that is, they have fairly low compressibilities) may be expected to be rapidly transported to the deeper receptors. The effective size of aspartame as a sweet solute presumably limits its depth of accession, and therefore it would be expected to occupy the shallower receptors where fewer sweet receptors are located.

A possible explanation for synergy may be an increase in the number of receptors recruited as well as decodification at the neural level. Mixing two solutes affects water structure and the accession of the solutes to the receptors (Munton & Birch, 1985). If more molecules are available to bind to the receptors, and more receptors are made available to the solutes (by the transport of the energetically- and structurally-favoured molecules to deeper layers of the tongue), the result will be an enhancement of sweetness, as in bulk:sodium cyclamate mixtures. The removal of free molecules of the bulk sweeteners from solution by the effect of aspartame, reduces sweetness, and hence suppression results. In mixtures, the effect of one solute on the hydration behaviour of the other is also important, as one solute may either hinder or enhance the accession of another to the receptor by modifying the solvation layer around it and hence its interaction with the surrounding water structure.

As an extension to the findings of Pfaffman, Bartoshuk and McBurney (1971), Moskowitz (1974) and Bartoshuk (1977), where they suggest that mixtures of similar tasting compounds show synergism whereas dissimilar-tasting substances show suppression, we have shown that, when both compounds exhibit marked hydrophilic character, their response is likely to be synergistic. Suppression is exhibited by compounds which do not interact well with water. Our findings also agree with those of Hutteau et al. (1998).

5. Conclusion

From a comparison of the solution properties of the mixtures producing synergy, additivity and suppression, and a knowledge of the chemical structures of their components in solution, it is possible to make some general observations.

1. Hydrophobicity of the molecule—Aspartame is the most hydrophobic of all and exhibits suppression in mixtures with the bulk sweeteners in solution. Sodium cyclamate is the most hydrophilic and shows synergism. Acesulfame K is of intermediate hydrophobic–hydrophilic character and shows additivity in its sweetness response with bulk sweeteners.
2. Size of molecule—Aspartame is the largest molecule (mw 294.3 g mol^{-1}) and exhibits suppression. Sodium cyclamate is smaller in size (mw 201.2 g mol^{-1}) and is synergistic with sucrose and maltitol. Acesulfame K is also 201.2 g mol^{-1} and has been found to be synergistic with maltitol and additive with sucrose.
3. Isentropic apparent specific compressibility—Mixtures of bulk sweeteners with aspartame at the concentrations exhibiting maximum suppression, have the lowest compressibilities. Sodium cyclamate mixtures have slightly larger compressibilities than aspartame mixtures. Bulk:acesulfame K mixtures have the highest compressibilities and exhibit additivity.
4. Ionic character of solute—Mixtures of the salts of the intense sweeteners with bulk sweeteners exhibit either synergy or additivity, whereas the non-ionic intense sweetener, aspartame, shows suppression.

The results suggest that, in binary mixtures, the presence of one substance may influence the fraction of presented molecules of a second substance that successfully recruits receptors, also shown by Birch et al. (1982). The effect of one solute on the other will depend on its concentration and nature, as well as the concentration and nature of the other component. At low concentrations, the pure solutes may pack differently from the way they do at higher concentrations, and the same applies to mixtures. The nature of the solutes will also dictate the type of packing in water, and therefore the availability of free solutes to access receptors.

Solution properties, and a knowledge of the nature of the sapid molecules, may therefore be used to help explain the ease of transport to receptors and probably the recruitment of taste molecules by the appropriate receptors. Although there is no proof that solution properties directly affect taste perception, experiments to date show that they certainly do play an active role in the taste mechanism.

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