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Physicochemical properties of sweeteners in artificial saliva and determination of a hydrophobicity scale for some sweeteners

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Saliva, the first physiological secretion induced by ingestion of food or beverages, plays an extensive role in the oral cavity and in taste perception. The influence of salts and proteins (the major constituents of saliva) on physicochemical properties of sweeteners is studied. Previous findings on the effects of KCl, NaCl and $MgCl_2$ on sweetener properties are now completed with the study of $CaCl_2$. Ca^{2+} modifies the type of hydration of sugars and polyols and has a detergent effect on sweet solutions. As water structure is sensitive to the presence of salts, physicochemical properties of sugars, polyols and intense sweeteners are determined in artificial saliva. Proteins also play a major part in hydration and surface properties of stimuli in saliva. All physicochemical properties determined in this work help in the mechanistic elucidation of sweet taste chemoreception. A scale of hydrophobicity is established for some sweeteners using partition coefficient and contact angle measurements.

INTRODUCTION

Inasmuch as sweetness perception takes place in the buccal environment where obviously water is present, the role of the aqueous solvent should not be neglected. The first stage in the mechanism of sweet taste chemoreception is the solvation of the stimulus. Saliva, composed of salts (1%) and water (99%), is an essential factor for taste mechanisms. It dissolves sapid substances and thus allows them to approach the receptor site (Bradley, 1988). Decrease in salivary flow leads to a reduced gustative perception (Zufrieden, 1986). Solvation implies interactions between solute and water, the structure of which is sensitive to the presence of all dissolved substances and especially electrolytes (Mathlouthi *et al.*, 1996). While the dissolution of organic molecules mainly depends on their polarity, the ionisation and the hydration of salts is a function of their electrostatic configuration. Given that their hydration properties are essential in aqueous solution, the saliva cations were studied in regard to their effect on sweetener properties and accession to receptor sites. The effects of KCl, NaCl and $MgCl_2$ have been reported (Mathlouthi *et al.*, 1996); we now investigate the influence of $CaCl_2$ and that of a mixture of these salts on solution properties of three saccharides (sucrose, D-glucose, D-fructose), four polyols (xylitol, sorbitol, maltitol, lactitol) and four intense sweeteners (aspartame, sodium cyclamate, saccharin Na, acesulfam K). The aqueous solution of salts is completed with proteins (mucin and α-amylase) to reconstitute artificial saliva and physicochemical parameters of sucrose, D-glucose and D-fructose are evaluated in this solvent. From the three-step model of sweetness chemoreception proposed by Mathlouthi *et al.* (1993), it appears that the ease of access to receptor site, and the spreading of stimulus over the hydrophobic protein receptor membrane, are of relevance in the mechanism of sweetness response. Some authors have already made a physicochemical approach to the study of stimulus-receptor interaction. Hansch (1973) found a good correlation for nitroanilines between relative sweetness, Hammett constant σ and hydrophobicity π. Daniel (1989) determined a molecular connectivity coefficient χ_m which takes into account the chiral centre and other physical properties. A comparable procedure was followed by Laffort (1987, 1993) and Patte *et al.* (1982) to link olfactory activity of some molecules and their physicochemical properties such as solubility, polarizability and molar volumes.

Interactions between water and sweet molecules have a major effect on water mobility, which plays an important role in the sweet response by activating the ion transfer across the receptor membrane and initiating the transduction mechanisms (Mathlouthi and Seuvre,

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1988; Lopez Chavez and Birch, 1997). The intensity of sweetness depends on the conformation of a stimulus molecule. Its hydrophobic side repels water molecules and decreases solute-solvent interactions, hence facilitating water-water interaction. Moreover, the hydrophilic side attracts water molecules and establishes hydrogen bonds. The resulting mobility of water is higher if the hydrophilicity and hydrophobicity of opposed sides are more contrasted. In the case of β-D-fructopyranose, the methylene group in 6 position repels water molecules in the vicinity of the sugar and hinders them from establishing hydrogen bonding with the receptor site (Mathlouthi, 1984). We now study the hydrophobicity of sugars (sucrose, D-glucose, D-fructose) and polyols (xylitol, sorbitol, maltitol, lactitol) using the hydrophobic-lipophilic balance (HLB) deduced from partition coefficients between water and isobutanol. A scale of hydrophobicity is also established for sugars, polyols and artificial sweeteners, as well as sweetness inhibitors and bitter substances, based on the contact angle of their aqueous solutions with a hydrophobic surface.

MATERIALS AND METHODS

Sucrose, D-glucose, sodium cyclamate, saccharin Na and mineral salts are Sigma products. D-fructose, xylitol and lactitol were donated by Xyrofin France (Paris) and sorbitol and maltitol by Roquette Frères (Lestrem, France). Aspartame and acesulfam K were complimentary and given by Nutra Sweet and Hoechst France (Paris), respectively, all with a purity >99%. Caffeine and quinine sulfate are Prolabo products; the sodium salt of 2-(4-methoxy-phenoxy) propionic acid (Na PMP) was synthesised and provided by Dr M. G. Lindley (Lintech, Reading). The synthesis of α-D-methyl 4,6 dichloro-4,6-dideoxy-galactopyranoside (methyl dichloro gal) was made in our laboratory according to the Bragg (1959), Jones (1960) and Jennings and Jones (1962, 1963, 1965) method. Alitame was supplied by Pfizer Inc. (NY). Sucralose was donated by Professor Plusquellec (ENSC, Rennes, France).

Artificial saliva

Electrolytic solution (saliva A) consisted of a mixture of 5.208 g $NaHCO_3$, 1.369 g $K_2HPO_4.3H_2O$, 0.877 g NaCl, 0.5 g NaN_3, 0.477 g KCl and 0.441 g $CaCl_2$.$2H_2O$ in 1 litre of HPLC grade doubly-distilled water (adjusted to pH 7); saliva B was identical but 2.16 g mucin and 200 000 units α-amylase were added (Van Ruth and Roozen, 1995).

Calcium ion solution

$CaCl_2$ was dissolved in doubly-distilled water with a concentration corresponding to the same number of meq/l (154) as the physiological serum solution.

Intrinsic viscosity [η] results are derived from the time neccessary for a given volume to flow through a capillary at a constant temperature of 25 ± 0.02°C in a semi-automatic Schott AV 400 viscometer. A triple extrapolation procedure was applied for accurate determination of [η] (Mathlouthi *et al.*, 1993). The equation of Huggins (1942) was used to determine the interaction parameter, Huggins constant k'. The apparent specific volumes ($V_2°$) were calculated from density measurements at 25 ± 0.1°C determined with a PAAR densitometer (DMA 45). Estimation of the hydration number h was done according to Herkovitz and Kelley (1973).

Surface tension (γ) results were obtained with a semi-automatic D2000 (Prolabo) tensiometer using a platinum blade wrench method at 25 ± 0.1°C and a buffered slightly mineralized water ('Volvic') for solutions preparation.

Contact angle (θ) measurements were made with a goniometer (type G40, KRÜSS) by determining the affinity of a sweetener solution with a hydrophobic surface (polyethylene). A droplet was placed on a solid polyethylene surface with a syringe; a micro-camera connected to a computer allowed calculation of θ values from the position of the droplet on the support. A hydrophobicity scale was determined by contact angle measurements, starting from apolar hydrophobic substances, e.g. xylene, DMF (Prolabo) which spread on the surface to polar components, especially water. Sugars, polyols and intense sweeteners were studied, as well as sucralose, alitame, bitter substances (quinine sulfate and caffeine) and sweetness inhibitor (Na PMP and methyl dichlorogalactoside).

Further information on hydrophobicity is given by the hydrophobic/lipophilic balance HLB, which is related to partition coefficient P according the equation: (HLB-7) = 0.36 ln(1/P) with P = [sugar in isobutanol]/[sugar in water] (Daniel, 1989). According to Collander (1950), the isobutanol is a fairly good solvent even for hydrophilic substances, such as sugars and amino-acids. Concentrations were determined by use of ionic HPLC (after shaking, for 5 min, aqueous sugar solution (1% w/v) with isobutanol) (Prolabo) (100/150 v/v). Chromatographic analysis of sugars was done on a Carbopac PA-100 (Dionex) anionic column eluted with NaOH (20 to 160 mM) with a pulsed amperometric detection. HPLC grade doubly-distilled water was used for all solutions.

Concentrations used for all measurements were 10% (w/v) sugar, 10% (w/v) polyols and 1% (w/v) intense sweeteners.

RESULTS AND DISCUSSION

We recently have reported the effects of some saliva salts (NaCl, KCl and $MgCl_2$) on the physicochemical properties of sugars and polyols (Mathlouthi *et al.*, 1996). We now investigate the influence of $CaCl_2$ and

that of a mixture of these four salts on the solution properties of sweeteners. To reconstitute artificial saliva, proteins were added to this mixture and solution properties of sweet stimuli were determined in this solvent.

Influence of saliva Ca^{2+} on solution properties of sweeteners

Viscometric constants (intrinsic viscosity [η], Huggins constant k'), apparent specific volume $V_2°$ and hydration number h in pure water and in 1% $CaCl_2$ aqueous solutions are listed in Table 1 for simple sugars (sucrose, D-fructose and D-glucose) and polyols (xylitol, sorbitol, maltitol and lactitol).

Intrinsic viscosity, [η], accounts for the hydrodynamic volume of the hydrated solute molecule and describes the effective size of the molecule (Kemp *et al.*, 1990). Compared with pure water, [η] decreases in presence of $CaCl_2$ for all studied solutes, especially for D-fructose and xylitol (see Table 1).

Huggins constant, k', gives a rough estimation of the ease of exchange between hydration water and bulk water. Apart from D-fructose and xylitol, which show an increase in k', other values of k' are found to decrease when $CaCl_2$ solution is used as a solvent, especially for lactitol (1.16 in water to 0.60 in $CaCl_2$ solution) (see Table 1).

Apparent specific volumes ($V_2°$), which measure the hydrostatic volumes of solutes account for the compatibilities of solutes with water structure and have been found to be good discriminators for the four basic tastes. The 0.52–0.71 $cm^3\ g^{-1}$ range was assigned to sweetness while, for bitter molecules, the range was 0.71–0.93 $cm^3\ g^{-1}$ (Shamil *et al.*, 1987). An increase in $V_2°$ is observed in the presence of $CaCl_2$ for sugars and polyols (Table 1). Hydration number h (mole H_2O/mole solute) is defined as the number of water molecules taking part in diffusion movement of solute. Table 1 shows that h is slightly decreased in the presence of $CaCl_2$.

From these physicochemical parameters, the presence of $CaCl_2$ seems to perturb the solvation of sugars and polyols in the aqueous medium. The cation, Ca^{2+}, is known to be highly hydrated (Magat, 1970). Under the influence of the electrostatic field, water molecules orientate themselves around ions, this reducing their degree of freedom in the medium. Consequently, solutes possess a lower volume of hydration sphere (decrease in [η]), transporting less water molecules in their displacement (decrease in hydration number h). Structure of hydration water seems stabilised and water mobility reduced (decrease in k'). From the increased values of $V_2°$, solutes, especially xylitol and sorbitol, appear less compatible with water structure in the presence of $CaCl_2$ by showing values at the upper limit of the sweetness range.

The specific effect of Ca^{2+} on water structure is also illuminated by surface tension (γ) measurements. Results reported in Fig. 1 show that, in a binary water/$CaCl_2$ solution as well as in a ternary water/$CaCl_2$/sugar solution, surface tension, γ, (of water (73.7 mN/m) is slightly increased. This expresses an increase in water molecules' cohesiveness which can be due to the action of the electrostatic field of the calcium ion.

Table 1. Intrinsic viscosity [η], Huggins coefficient k', apparent specific volume $V_2°$ and hydration number (h) for 10% (w/v) sugar or polyols in pure water and in calcium solution at 25°C

	Sucrose		Fructose		Glucose			
	H_2O	$CaCl_2$	H_2O	$CaCl_2$	H_2O	$CaCl_2$		
[η] ($cm^3\ g^{-1}$)	2.37	2.34	2.29	2.18	2.37	2.36		
k'	1.15	0.98	0.89	1.03	1.14	0.97		
$V_2°$ ($cm^3\ g^{-1}$)	0.62	0.63	0.61	0.64	0.62	0.64		
h	6.14	5.80	3.01	2.34	3.26	2.78		
	Xylitol		Sorbitol		Lactitol		Maltitol	
[η] ($cm^3\ g^{-1}$)	2.37	2.22	2.44	2.40	2.48	2.42	2.46	2.37
k'	0.86	0.89	0.89	0.62	1.16	0.60	0.86	0.69
$V_2°$ ($cm^3\ g^{-1}$)	0.66	0.70	0.65	0.67	0.63	0.66	0.63	0.65
h	2.43	1.62	3.31	2.92	6.98	5.90	6.89	5.61

Physicochemical properties of sweeteners in artificial saliva

Saliva A, composed of 1% (w/v) of saliva salts (NaCl, KCl, $MgCl_2$, $CaCl_2$), is used as a solvent for the determination of solution properties of sugars, polyols and artificial sweeteners. Saliva A, completed with proteins (mucin, α-amylase), is used as a solvent (saliva B) for solution properties of sucrose, D-fructose and D-glucose.

Viscosimetric constants

Experimental values of intrinsic viscosity, [η], and Huggins constant, k', in saliva A and B for sugars, polyols and intense sweeteners (aspartame, sodium cyclamate, saccharin Na and acesulfam K) are reported in Fig. 2. For sucrose, D-fructose, D-glucose, aspartame and sodium cyclamate, [η] seems to increase when salts are added to pure water. For polyols, [η] values do not change significantly and for ACK and sodium saccharin, [η] tends to decrease in saliva A. For all compounds studied, apart from saccharin and ACK, the ease of exchange between hydration water and bulk water, as described by Huggins constant (k'), seems to decrease in saliva A (see Fig. 2). When proteins are added to saliva A (saliva B), [η] decreases for sucrose and D-fructose whereas it remains unchanged for D-glucose. k' in saliva B is augmented for the three sugars as compared with k' in water and in saliva A (see Fig. 2).

These results show that in the presence of saliva salts (saliva A), the hydration around sugars, polyols aspartame and sodium cyclamate tends to increase (higher [η] values than in pure water) while the water mobility around these solutes is reduced (lower k' values). This is probably due to the orientation of water molecules in the ionic electrostatic field around cations which reduces their degree of freedom. Saccharin Na and acesulfam K

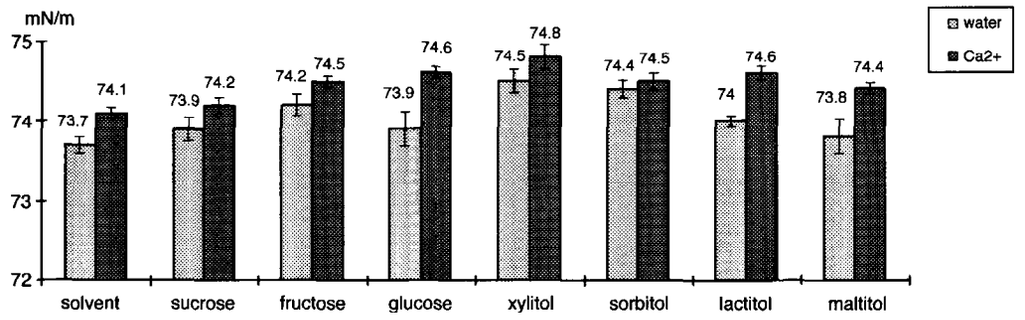


Fig. 1. Surface tension γ (mN/m) for 10% (w/v) sugar (sucrose, D-fructose and D-glucose) in water and in $CaCl_2$ aqueous solution at 20°C.

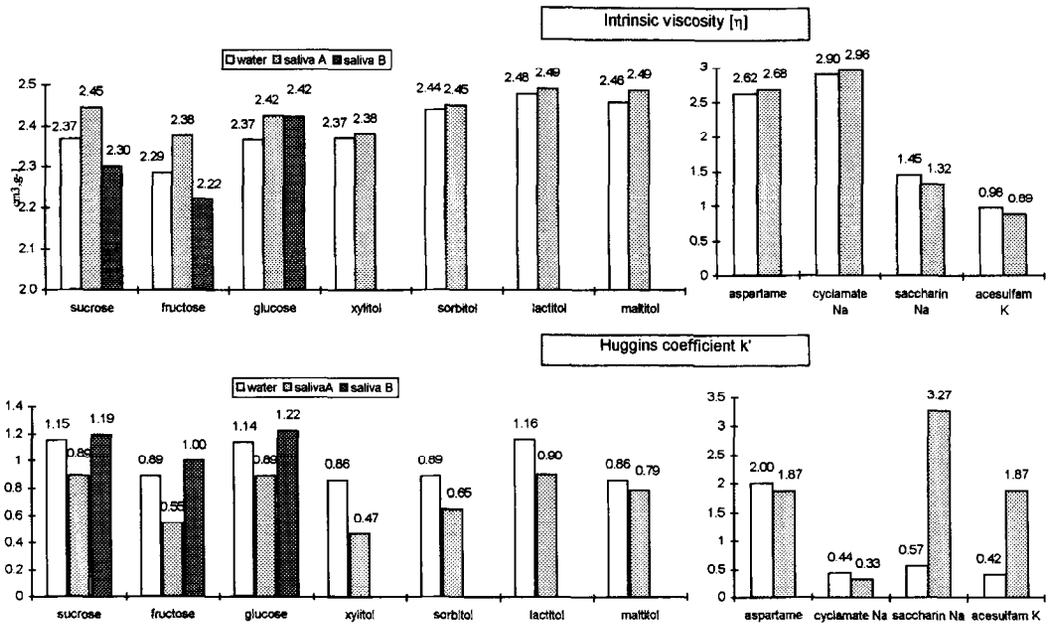


Fig. 2. Intrinsic viscosity [η] and Huggins coefficient k' for 10% (w/v) sugar, polyol or 1% (w/v) intense sweeteners in pure water, saliva A or saliva B.

are weakly marked by hydrophilic hydration (small [η] values). The addition of mucin and α-amylase to saliva salts leads to a reduction of the hydration sphere for sucrose and D-fructose while that of D-glucose remains the same as in saliva A. The presence of proteins facilitates the exchange of water molecules around these sugars (increase of k' values as compared to that in water and in saliva A).

Apparent specific volumes ($V_2°$)

Experimental results of $V_2°$ in water, saliva A and saliva B are given in Fig. 3. In saliva A, a slight increase in $V_2°$ values is observed for sugars, aspartame and sodium cyclamate ($\Delta V_2° = +0.02\,cm^3\,g^{-1}$). On the other hand, the apparent specific volume, $V_2°$, of sodium saccharin tends to decrease in the presence of saliva A ($\Delta V_2° = -0.04\,cm^3\,g^{-1}$). (see Fig. 3). In saliva B, $V_2°$ values for D-sucrose and D-fructose are slightly lower than in saliva A while that of D-glucose remains constant. The slight increase in $V_2°$ values observed accounts for a slight increase in the volume of the first hydration sphere of the solute.

Hydration number, h, gives an estimation of the number of water molecules strongly bound to solute (H-bond lifetime for water-solute longer than that for water-water). A slight increase in h is observed in Fig. 4 for sugars, maltitol, aspartame and sodium cyclamate in saliva A. This is attributable to a slight stabilization of water molecules around these solutes in the presence of saliva salts. An opposite effect (slight decrease in h) is observed for sorbitol and lactitol and indicates a destabiliziation of water around these molecules.

Negative values of h are obtained for saccharin Na and ACK (Fig. 4). This can be explained by the fact that the opposition between hydrophobic and hydrophilic sides of the solute molecules induces a high mobility of water molecules which do not remain bound to solute molecules (H-bond lifetime for water-solute shorter than in bulk water). Water molecules around solutes are more mobile than in the bulk water (negative hydration).

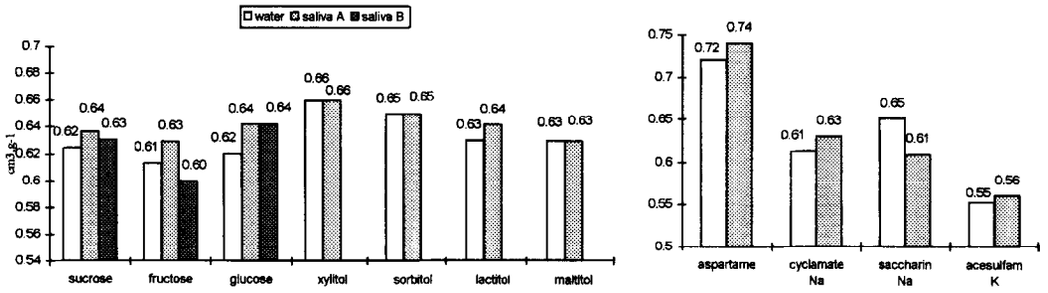


Fig. 3. Apparent specific volume ASV ($V_2°$) for 10% (w/v) sugar, polyol or 1% (w/v) intense sweeteners in pure water, saliva A or saliva B.

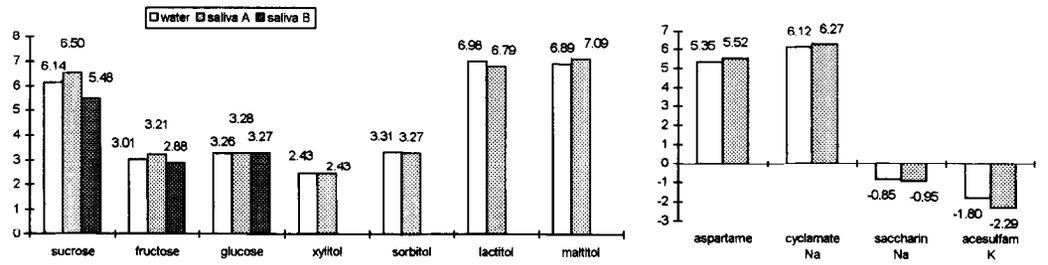


Fig. 4. Hydration number h for 10% (w/v) sugar, polyol or 1% (w/v) intense sweetener in pure water, saliva A or saliva B.

In saliva A, the increased negative values of h indicate a more pronounced negative hydration. In saliva B, the hydration number, h, is slightly decreased for sucrose and D-fructose and unchanged for D-glucose, as compared to saliva A.

Surface tension (γ) is an indicator for the detergency of solute in solution. Figure 5a and b gives the values of γ at 20°C for all compounds studied in water and in saliva A and saliva B. All sugars and polyols studied, when they are dissolved in pure water, slightly increase γ and hence the cohesiveness of water molecules (see Fig. 5a). A slight decrease in surface tension of water is observed for artificial sweeteners, especially for aspartame which gives a value of $\gamma = 65.8$ mN/m at a relatively low concentration (0.9%) indicating a high detergent effect.

The addition of a mixture of salts (saliva A) in pure water faintly affects the cohesiveness of water (γ is equal to 73.7 mN/m for water and to 73.3 mN/m for saliva A). For all compounds studied, values in saliva A do not

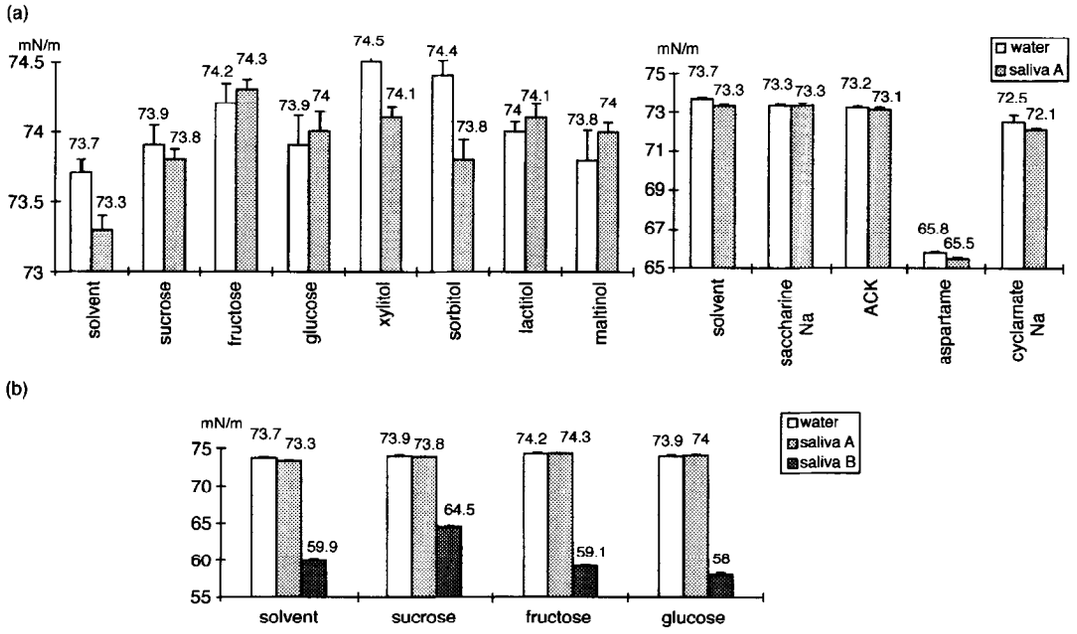


Fig. 5. (a) Surface tension γ for 10% (w/v) sugar and 1% (w/v) intense sweetener in pure water and in saliva A at 20°C. (b) Surface tension ((mN/m) for 10% (w/v) sugar (sucrose, D-fructose and D-glucose) in saliva B at 20°C.

significantly change as compared with that observed in pure water, except for xylitol, sorbitol and aspartame for which γ is slightly decreased (Fig. 5a). The addition of mucin and α-amylase (saliva B) to the medium leads to a pronounced effect on γ (which is decreased in all cases (see Fig. 5b).

Artificial saliva (saliva B) seems to contribute to a noticeable decrease in the cohesion of water which allows the sweetener solution to spread more easily onto the receptor membrane. The surface tension lowering can be related to the hydrophobicity of sweeteners and determined as a degree of affinity with a hydrophobic surface.

Determination of a hydrophobicity scale for sweeteners

Lipophilicity is included amongst the structural parameters needed for QSAR analysis. Several examples show that the biological activity can be related more or less directly to this parameter (Hansch, 1969). Bitterness is also associated with lipophilicity: replacement of an OH group in a sugar with a hydrophobic group leads to an increase of sweetness until a maximum, beyond which increase in hydrophobicity provokes a progressive sweetness disappearance and bitterness is perceived (Daniel, 1989).

Experimentally, lipophilicity is quantified by partition coefficient, P, which is a measure of the relative affinity between the solute and a biphasic system (aqueous and butanolic phases). A solute more soluble in a polar phase ($P < 1$) than in an organic phase is hydrophilic. Partition coefficient measurements for some sugars, polyols and intense sweeteners were carried out and the results are given in Table 2. All these solutes are hydrophilic but D-fructose, sucrose, xylitol and maltitol seem to have a relatively higher hydrophobic character than the other comparable compounds (higher P values).

The concept of partition coefficient, P, can be related to hydrophilic/lipophilic balance HLB by a simple equation: $(HLB\text{-}7) = 0.36 \ln(1/P)$ (Davies, 1957). There is no simple relationship between HLB and relative sweetness but HLB can affect the distribution of the sweetener from an aqueous environment (saliva) onto a lipophilic receptor and then the distribution of sweet ligand from the lipophilic receptor to the hydrophilic environment of the saliva. HLB values lie usually between 1 (non polar compounds) and 20 (polar compounds). Under the previous experimental conditions, sugars and polyols show intermediate HLB values of 7.62–8.38 (Table 2). The difference in HLB between components is relatively low. However D-fructose and sucrose can be differentiated from D-glucose and sorbitol by a lower hydrophilic/ lipophilic balance.

Another physicochemical parameter, contact angle θ, can help in understanding the diffusion of sweet molecules onto the lipophilic surface of the receptor site. The experiment was done on a hydrophobic polyethylene surface. θ measurements were obtained on solutions of sugars, polyols and intense sweeteners and results are given in Table 3. In order to establish a scale of θ values, hydrophobic organic solvents (xylene, DMF) were studied, together with a polar solvent (water) and aqueous solutions of sugars and polyols including isomaltulitol (isomalt is a mixture of maltitol and isomaltulitol). Bitter substances (quinine sulfate and caffeine), sweetness inhibitor components (methyl-dichlorogalactoside and NaPMP) were also analysed (see Table 3). The lower the θ value, the more the droplet is spread over the holder and the higher the solute hydrophobicity. The studied substances could be classified according to their hydrophobic character: sugars and polyols < intense sweeteners (sodium saccharin, sodium cyclamate, ACK

Table 2. Partition coefficient (P) and hydrophilic/lipophilic balance (HLB) for sugars and polyols

	[Sugar] in water (g/l)	[Sugar] in isobutanol (g/l)	P	HLB
D-Fructose	1.87	0.34	0.18	7.62
Sucrose	3.43	0.60	0.17	7.63
Maltitol	3.28	0.40	0.12	7.76
Xylitol	1.39	0.17	0.12	7.76
Lactitol	2.19	0.17	0.08	7.93
Sorbitol	1.87	0.11	0.06	8.00
D-Glucose	2.47	0.05	0.02	8.38

Table 3. Contact angle θ (°) of 10% (w/v) sugars and polyols and 1% (w/v) intense sweeteners, inhibitor and bitter substances and their quality of taste

Component	Contact angle (°)	Quality of taste
Xylene	0	
DMF	51.82 ± 1.34	
Alitame	73.71 ± 2.50	Clean intense sweet taste, lingering
Sucralose	82.59 ± 2.10	Instant onset of clean sweet taste
Methyl diCl gal	83.63 ± 0.82	Inhibitor
NaPMP	84.11 ± 1.50	Inhibitor
Caffeine	86.13 ± 1.20	Bitter
Aspartame	88.98 ± 2.30	Lingering sweet-bitter aftertaste
Quinine sulfate	90.69 ± 0.90	Bitter
Acesulfam K	92.10 ± 0.52	Lingering bitter and chemical synthetic aftertastes
Sodium cyclamate	94.50 ± 0.82	Lingering sweet-sour aftertaste
Sodium saccharin	94.74 ± 1.69	Bitter-metallic-astringent aftertastes
Erythritol	94.93 ± 2.15	
Fructose	95.85 ± 1.35	
Sucrose	96.30 ± 1.44	
Xylitol	96.96 ± 0.66	
Isomaltulitol	98.19 ± 0.75	
Maltitol	98.25 ± 1.22	
Isomalt	98.33 ± 0.78	
Lactitol	99.29 ± 2.20	
Sorbitol	99.33 ± 0.84	
Glucose	99.39 ± 1.48	
H_2O	100	

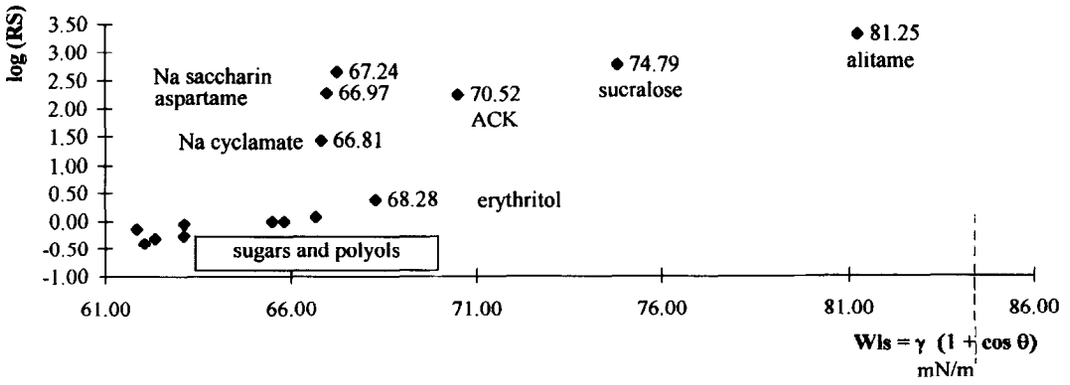
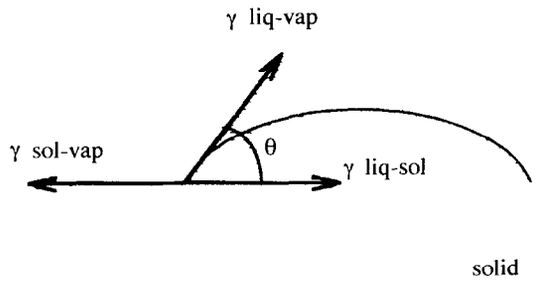


Fig. 6. Relationship between relative sweetness (RS) and adhesion force Wls.

and aspartame) < sweetness inhibitors and bitter components < sucralose and alitame.

In 1805, Young defined a relation which describes the interfacial tension at the 3-phases contact line:



$$\gamma_{sol\text{-}vap} = \gamma_{liq\text{-}sol} + \gamma_{liq\text{-}vap} * \cos(\theta) \Rightarrow \gamma_{liq\text{-}sol} = \gamma_{liq\text{-}vap}(1+\cos\theta) = Wls$$

with $\gamma_{liq\text{-}vap}$, the interfacial tension of the liquid. $\gamma_{liq\text{-}sol}$ is assimilated to the adhesion force on the support and noted Wls. When log(RS) (RS: relative sweetness) is plotted as a function of Wls, a correlation is found which permits classification of sweeteners into different categories (see Fig. 6):

- hydrophilic low intensity sweeteners (sugars and polyols) with a relatively low adhesion force;
- hydrophobic high intensity sweeteners (sucralose and alitame) with a clean sweet taste showing the largest values of W_{ls};
- hydrophobic high intensity sweeteners (aspartame, ACK, sodium saccharin and sodium cyclamate), with defined aftertastes (see Table 3), showing an intermediate values of W_{ls}. Erythritol is found in this category although its sweetness is low (this short chain alcohol behaves like glycerol as concerns its increased hydrophobicity).

CONCLUSION

Salts, because of their specific ionic hydration, can modify the type of hydration of sweeteners (simple sugars and polyols) and the organization of water molecules in aqueous solutions of sweeteners. This modification influences the accession of stimulus to receptor site and then the perception of sweet taste. Our physicochemical approach needs to be completed with the sensory evaluation of the sweeteners in the presence of salts to evaluate their effect on sweet taste intensity and persistence. It was recently (Barisias *et al.*, 1995) shown that small amounts of NaCl enhance sucrose sweetness. Kurihara *et al.* (1990) found that monovalent salts and Ca^{2+} prolonged the response to D-fructose for the dog and suggested that the presence of salts on the tongue surface may increase the interaction between receptor domains for sugars, perhaps stabilizing the active complex with the receptor. Van der Heijden *et al.* (1983) showed that sub-threshold levels of salts can enhance or suppress the perceived sweetness and the degree of this effect depends on the type of salt and sweetener.

In saliva, each salt has a specific effect on water structure. The resultant effect of a mixture of salts on solution properties is a loss of mobility of water molecules around the solute and a perturbation of the cohesiveness of water. Proteins also play a major part in hydration, the most pronounced being the detergent effect (lowering of surface tension) on sweet solutions. Surface properties of stimulus solutions seem to be relevant for the understanding of taste perception. A well known taste modifier, gymnemic acid, has been shown to have surfactant properties comparable to sodium lauryl sulfate (De Simone, 1980). Partition coefficient and contact angle seem to be good discriminant factors to estimate hydrophobicities of sweeteners. Among sugars and polyols, erythritol and D-fructose are differentiated by a relatively high hydrophobicity. Intense sweeteners, as well as bitter substances and sweetness inhibitor components, show pronounced hydrophobic character.

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