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Physicochemical and psychophysical characteristics of binary mixtures of bulk and intense sweeteners

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Correlating psychophysical characteristics with physicochemical properties of sweeteners is of relevance to the understanding of the origin of sweetener synergy, an essential parameter for the food manufacturer. Psychophysical evaluation was carried out on bulk sweeteners (sucrose and maltitol) and intense sweeteners (aspartame, sodium cyclamate, acesulfam-K, alitame) in mixtures. The concentrations of mixtures were calculated to be equisweet to 10% sucrose and sweetness intensity was evaluated by reference to sucrose solutions using a "sip and spit" method. While a positive synergistic phenomenon is observed for sugar/sodium cyclamate and maltitol/acesulfamK mixtures, a significant suppression effect is obtained when aspartame is added to sugars. Additivity is observed for sucrose/alitame and sucrose/acesulfamK mixtures. The origin of these differences lies in the influence of the two molecules on water structure and in the nature of their hydration. From physicochemical properties (intrinsic viscosity, Huggins coefficient, apparent specific volume, hydration number, surface tension and contact angle), alitame and aspartame seem characterised by hydrophobic hydration; sodium cyclamate, as well as the bulk sweeteners, appear more compatible with water structure and possess hydrophilic hydration. ACK is differentiated from other sweeteners by a negative hydration. Synergy occurs when components with identical types of hydration are mixed. This phenomenon is accompanied by an increase in the mobility of water molecules in the proximity of bulk sweeteners (maltitol and sucrose) and a reduction of volume of the hydrated solute molecule. Inversely, suppression and additivity occur when constituents of the mixture possess different natures of hydration, as in sucrose/aspartame mixtures, and when physicochemical properties show a reduction of the mobility of water around the sweeteners. For suppression effects, an increase in volume of the hydration sphere is also observed. Interpretation of the sweetness of mixtures of sugars and artificial sweeteners, in terms of their compatibility with water structure, is of relevance at an economic level in food formulations.

INTRODUCTION

Sweet taste is known to play a preponderant role in food preferences. While the use of sucrose prevails in traditional food industry, numerous nutritive and non-nutritive sweeteners offer new opportunities for the food manufacturer. Development of alternative sweeteners was initiated because of advantages in health (diet and disease) and economy. The benefits of sweetener blends are now unquestioned. They can lead to the formulation of foods and beverages with improvement of sweetness quality, avoiding problems of taste instability. Bulk and intense sweeteners provide some 'body' to reduced calorie beverages and increase the shelf-life of soft drinks, even at quite low pH (Lotz and Meyer, 1994). Moreover, some sensorial properties of synthetic sweeteners are known to limit their use in low-calorie soft drinks; combining different sweeteners can overcome these limitations (van der Tornout *et al.*, 1985). The approval of maltitol and cyclamate in the UK in December 1995, has paved the way for the market

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introduction of these bulk and intense sweeteners. Maltitol has organoleptic and technological properties close to those of sucrose (Portmann and Kilcast, 1996) and it may be used as a sucrose substitute in a wide range of non-cariogenic and/or calorie-reduced food products.

When two sweeteners are blended, the perceived intensity of the mixture may be equal to (additivity), greater than (synergy) or less than (suppression) the sum of the individual sweetness intensities. The complexity of the interactions between the solute and the solvent is probably at the origin of the difficulty of interpretation of the sweetness of the mixtures. Indeed stimuli-receptor interactions, as well as solute-solvent or solute-solute interactions, are of the same nature, namely hydrogen bonding. Moreover, elucidating the sweetness mechanism needs interpretation of the hydrogen bonding in the solvent medium as well as interpretation of solution properties of the aqueous solutions of sweeteners (Mathlouthi and Portmann, 1990; Mathlouthi and Seuvre, 1988). For the explanation of synergistic phenomena, the role of water can be quantified and used to interpret qualitative and quantitative effects (Birch, 1996).

Approval of new sweeteners, combined with the key role played by sweetener blends in the production of foods and beverages, has led us to investigate the psychophysical characteristics of sucrose and maltitol blended with intense sweeteners (aspartame, alitame, acesulfam-K and sodium cyclamate). Alitame is not yet approved in Europe but was investigated in view of its potential importance within the next generation of high potency sweeteners.

Physicochemical results on the binary mixtures of sugars and intense sweeteners are also reported and the observed synergy or suppression of taste is interpreted by reference to their physicochemical properties (intrinsic viscosity, Huggins constant, apparent specific volume, surface tension, contact angle).

MATERIALS AND METHODS

For physicochemical analysis, sucrose and sodium cyclamate were Sigma products. For sensory analysis, sucrose was commercially purchased (Tate and Lyle, London) and sodium cyclamate was a gift of Jan Dekker BV (NL). Other sweeteners were donated by the producers: aspartame (Holland Sweetener Company, NL and NutraSweet, Switzerland), acesulfam-K (Hoescht, UK), alitame (Pfizer, UK) and maltitol (Roquette Limited).

Sensory procedure

Concentrations of intense and bulk sweeteners for each blend were determined from the concentration-sweetness intensity response curves previously established (Portmann and Kilcast, 1996). The approach consisted in developing and evaluating mixtures of sweeteners that were expected to have an equal sweetness intensity. The concentrations of mixtures were calculated to be equivalent in sweetness to 10% w/v sucrose. Five binary mixtures containing, respectively, 0, 25, 50, 75 and 100% of the sweetness originating from the intense sweetener, and the remaining percentage originating from the bulk sweetener, were prepared. Mixture ratios of sucrose-intense sweeteners and maltitol-intense sweeteners are given in Table 1.

The assessed solutions were prepared 24 hours prior to tasting. The tasting was conducted in individual booths; the same trained panel was used to generate the concentration-response (C–R) curves involved in this study. They were 11 female panellists with extensive experience in sweetener assessment. Data acquisition and collection were monitored through a computerised system (Reading Scientific Services, Reading, UK). For each sample, sweetness intensity is evaluated by reference to sucrose solutions using a 'sip and spit' method (Portmann and Kilcast, 1996). A Fisher's LSD (Least Significance Difference) procedure, at a significant level of 5%, was conducted across the samples to compare the intensity ratings of any of the mixtures. Data were analysed with MINITAB 10 (MINITAB, Inc., State College, PA, USA) and STATISTICA (StatSoft, Inc. Tulsa, OK, USA). Visual representations of the synergy effect were drawn with Quattro®Pro5 (Borland International Inc., Scott Valley, USA).

Physicochemical methods

Viscosity results were obtained for mixture solutions at $25 \pm 0.02°C$ using a semi-automatic Schott AVS 400 viscometer. A triple extrapolation procedure was applied for accurate determination of intrinsic viscosity $[\eta]$ (Mathlouthi *et al.*, 1993). Huggins constant k' was determined from the equation of Huggins (1942). Apparent specific volumes ($V_2°$) were calculated from density measurements at $25 \pm 0.1°C$ determined with a PAAR densitometer (DMA 45). Measurements were made for sugar solutions with 2 to 6% (w/v) concentration and intense sweetener solution was used as a solvent in a proportion corresponding to that needed for maximum synergy, suppression or additivity (Table 2). Estimation of the hydration number h was made according to Herkovitz and Kelley (1973) and was calculated with a molecular weight which takes into account the proportion of the intense sweetener in the mixture.

Surface tension (γ) measurements were made with a semi-automatic D2000 (Prolabo) tensiometer using a platinum blade wrench method at $25 \pm 0.1°C$ and solutions were prepared with a buffered slightly mineralised water ('Volvic'). Contact angle (θ) measurements were made with a goniometer (type G40, KRÜSS) placing a drop of solution on a solid polyethylene surface with a micro-syringe. A camera connected to a computer allowed calculation of θ values from the spreading of the

Table 1. Bulk and intense sweetener concentrations in aqueous mixtures

Sweetness contribution	Bulk-intense sweetener mixture ratio				
	0–100	25–75	50–50	75–25	100–0
		Sucrose–alitame			
Sucrose (g % ml)	—	1.92	4.55	7.18	10.00
Alitame (ppm)	63	36	19	8	—
		Sucrose–aspartame			
Sucrose (g % ml)	—	1.92	4.55	7.18	10.00
Aspartame (ppm)	1333	1000	750	522	—
		Sucrose–acesulfam K			
Sucrose (g % ml)	—	1.92	4.55	7.18	10.00
Acesulfam K (ppm)	867	448	248	118	
		Sucrose–cyclamate			
Sucrose (g % ml)	—	1.92	4.55	7.18	10.00
Cyclamate (ppm)	4400	3000	1980	980	—
		Maltitol–alitame			
Maltitol (g % ml)	—	3.55	7.12	10.70	13.50
Alitame (ppm)	63	36	19	8	—
		Maltitol–aspartame			
Maltitol (g % ml)	—	3.55	7.12	10.70	13.50
Aspartame (ppm)	1333	1000	750	522	—
		Maltitol–acesulfam K			
Maltitol (g % ml)	—	3.55	7.12	10.70	13.50
Acesulfam K (ppm)	867	273	2448	118	—
		Maltitol–cyclamate			
Maltitol (g % ml)	—	3.55	7.12	10.70	13.50
Cyclamate (ppm)	4400	3000	1980	980	—

droplet over the surface. For these interfacial properties, mixture ratios of sucrose-intense sweeteners and maltitol-intense sweeteners correspond to that needed for maximum synergy (Table 2).

Solutions were prepared in HPLC grade doubly-distilled water except for surface tension measurements which need a buffered mineral water 'Volvic'.

Table 2. Sweetener concentrations used for the determination of physicochemical properties of mixtures

Sweetener mixture	Bulk/intense sweetener blend ratio	% Synergy	Sweetener concentration
Sucrose (g % ml)			1.92
Aspartame (ppm)	25/75	−32.84	1000
Sucrose (g % ml)			1.92
Alitame (ppm)	25/75	−6.61	36
Sucrose (g % ml)			1.92
AcesulfamK (ppm)	25/75	−6	448
Sucrose (g % ml)			7.18
CyclamateNa (ppm)	75/25	12.75	980
Maltitol (g % ml)			3.55
Aspartame (ppm)	25/75	−12.4	1000
Maltitol (g % ml)			7.12
Alitame (ppm)	50/50	−11	19
Maltitol (g % ml)			10.7
AcesulfamK (ppm)	75/25	19.07	118
Maltitol (g % ml)			7.12
Cyclamatena (ppm)	50/50	27.4	1980

RESULTS AND DISCUSSION

Psychophysical functions, such as Beidler's mixture equation (Beidler, 1962, 1971), do not attribute a specific role to the medium (water) in which the sweeteners are dissolved (Frijters *et al.*, 1990). Synergy of sweetener mixtures is a complex phenomenon which, if it exists, involves sugar/water and sugar/receptor interactions and neural mechanisms, or combinations of all three possibilities (Birch *et al.*, 1982). Birch *et al.* (1980) developed the concept of the 'orderly queue hypothesis' in which the taste receptor stimulation results from the activity of the sweeteners and investigation of synergy phenomena must be analysed as a competition between the sweeteners and water molecules for the sweet receptors.

Evaluation of synergism

For each of the binary mixtures, sucrose-intense sweeteners and maltitol-intense sweeteners, the quantitative estimate of % synergy is obtained by the equation:

$$\%\ \text{synergy} = 100^{*}\left[\frac{SE(blendA+B)}{[SE(100\%A)+SE(100\%B)]/2}-1\right]$$

(Carr *et al.*, 1993) where (SE) represents the panel's sweet intensity rating for the sample.

Intensity response rates together with the sem (standard error of the mean) and the % synergy are presented in Table 3 for sucrose-intense sweeteners mixtures and in Table 4 for maltitol-intense sweeteners mixtures. The variation of % synergy, as a function of the % of intense sweeteners in the mixtures, is represented in Figs 1 and 2.

A Fisher's LSD procedure was performed on each binary mixture at each concentration level to determine whether the sweetness intensity rating of each mixture was significantly different from the average sweetness intensity of two 100% solutions. The nature of the mixture interactions is deduced from this statistical analysis. Synergy and suppression occur if the sweetness intensity of the mixture is significantly increased or decreased, respectively. If no statistically significant difference is noticed, additivity is assumed to result. Significant synergy is observed (Tables 3 and 4) for sucrose-cyclamate, maltitol-cyclamate and maltitol-acesulfam-K mixtures. The maximum synergy is obtained when bulk sweetener contributes approximately 50% of the overall sweetness for the maltitol-cyclamate mixture and 75% of the total sweetness for the sucrose-cyclamate and maltitol-acesulfam-K mixtures. These results are in accordance with those of Weikmann *et al.* (1969), van der Tornout *et al.* (1985), Hyvönen *et al.* (1978) and Mathlouthi and Portmann (1994).

Significant suppression is obtained when aspartame is added to bulk sweeteners (sucrose and maltitol) with a contribution of 25% to the total sweetness by sugars.

Table 3. % Synergy for sucrose/intense sweetener mixtures

Sweetness contribution	Sucrose-intense sweetener mixture ratio 0–100	25–75	50–50	75–25	100–0
Expected sweetness	10	10	10	10	10
			Sucrose–alitame		
Sweetness intensity (sem)	10.12 (3.0)	9.17 (2.4)	9.27 (1.9)	9.29 (2.3)	9.52 (1.8)
% Synergy	—	−6.61	−5.60	−5.39	—
F-ratio	—	0.11	0.27	0.18	—
p	—	ns	ns	ns	—
			Sucrose–aspartame		
Sweetness intensity (sem)	9.2 (3.1)	6.43 (2.0)	7.66 (2.0)	9.2 (2.8)	9.95 (2.6)
% Synergy	—	−32.84	−20.0	−3.91	—
F-ratio	—	0	0.00014	0.45	—
p	—	***	***	ns	—
			Sucrose–acesulfam K		
Sweetness intensity (sem)	10.7 (3.4)	9.7 (1.9)	10–1.(1,7)	10.2 (1.6)	9.95 (2.6)
% Synergy	—	−6	−2	−1.16	—
F-ratio	—	0.23	0.32	0.91	—
p	—	ns	ns	ns	—
			Sucrose–cyclamate		
Sweetness intensity (sem)	11.7 (3.12)	11.72 (1.9)	12.1 (1.8)	12.1 (1.7)	9.95 (2.6)
% Synergy	—	8.31	11.80	12.75	—
F-ratio	—	0.027	0.0018	0.0009	—
p	—	*	**	***	—

* $p<0.05$ ** $p<0.01$ *** $p<0.001$; ns: not significant.
(sem = standard error of the mean).

Table 4. % Synergy for maltitol/intense sweetener mixtures

Sweetness contribution	Maltitol-intense sweetener mixture ratio 0–100	25–75	50–50	75–25	100–0
Expected sweetness	10	10	10	10	10
			Maltitol–aspartame		
Sweetness intensity (sem)	10.4 (1.8)	8.56 (2.3)	9.4 (2.2)	9.74 (2–2)	9.14 (2.4)
% Synergy	—	−12.40	−3.80	−0.30	—
F-ratio	—	0.009	0.43	0.95	—
p	—	**	ns	ns	—
			Maltitol–acesulfam K		
Sweetness intensity (sem)	9.0 (3.0)	10.2 (3.1)	10.7 (2.5)	10.8 (2.6)	9.14 (2.4)
% Synergy	—	12.45	18.0	19.07	—
F-ratio	—	0.0440	0.0037	0.0017	—
p	—	*	**	**	—
			Maltitol–cyclamate		
Sweetness intensity (sem)	10.55 (3.2)	12.45 (2.3)	12.54 (2.0)	11.64 (2.2)	9.14 (2.4)
% Synergy	—	26.40	27.40	18.20	—
F-ratio	—	0	0	0.0001	—
p	—	***	***	***	—
			Maltitol–alitame		
Sweetness intensity (sem)	10.8 (2.4)	9.88 (2.7)	8.9 (2. 1)	9.01 (3.0)	9.2 (2.4)
% Synergy	—	−1.2	−11.0	−9.9	—
F-ratio	—	0.96	0.05	0.092	—
p	—	ns	ns	ns	—

* $p<0.05$ ** $p<0.01$ *** $p<0.001$; ns: not significant.
(sem = standard error of the mean).

Additivity is obtained for sucrose-acesulfamK, sucrose-alitame and maltitol-alitame mixtures with a maximum obtained when bulk sweeteners contributed 25% of the sweetness except for maltitol-alitame (50%) (Tables 3 and 4).

Solution properties

Synergy, suppression or additivity may have as their origin the effect of each of the solutes on water structure. Sweet taste is known to be mediated by water and

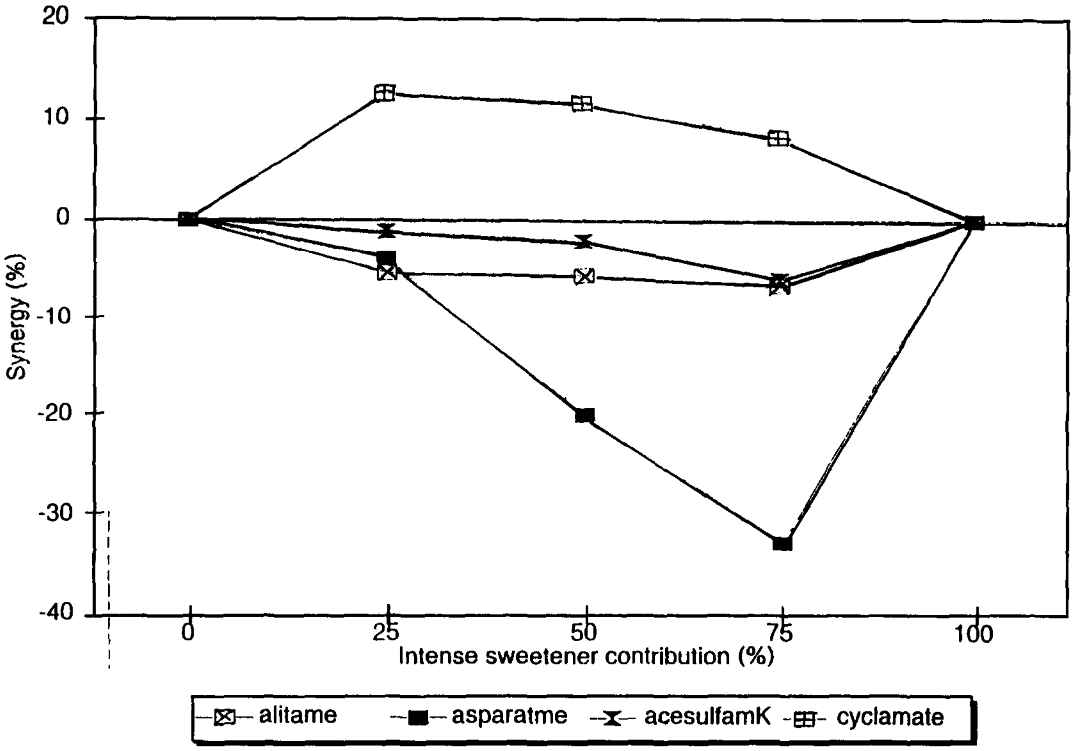

Fig. 1. Synergy in intense sweeteners/sucrose mixtures.

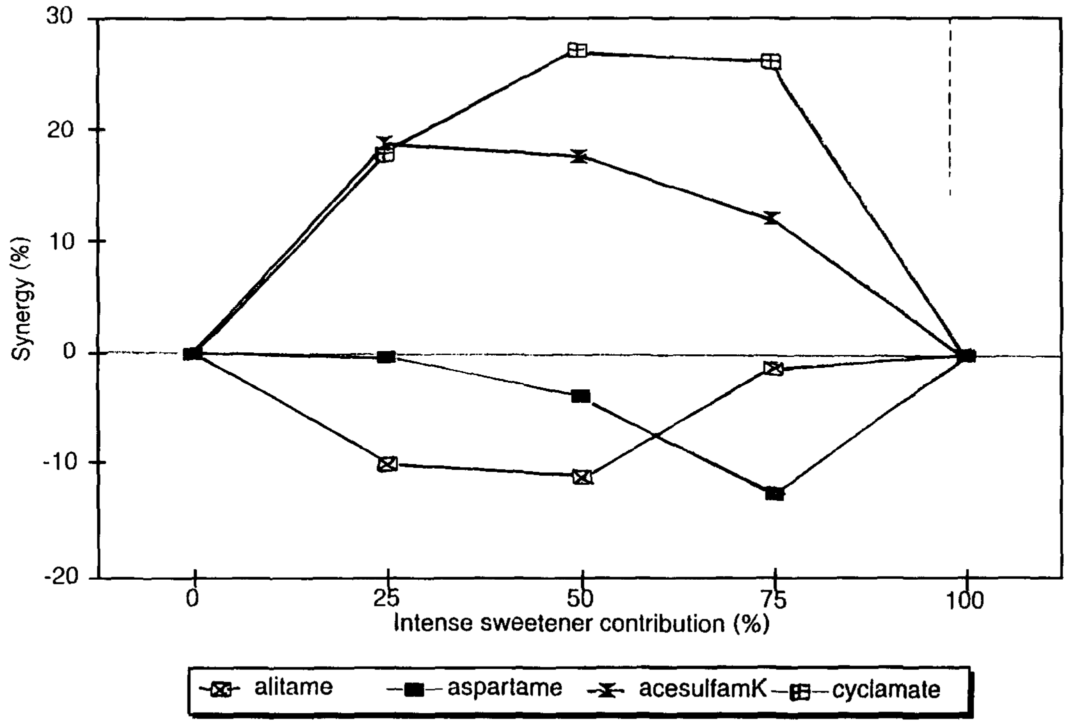

Fig. 2. Synergy in intense sweeteners/maltitol mixtures.

the role of the aqueous medium is far from negligible (Mathlouthi *et al.*, 1996; Birch, 1997). Determination of solution properties (intrinsic viscosity [η], Huggins coefficient k′, apparent specific volume $V_2°$, hydration number h, surface tension γ and contact angle θ) allows estimation of the type of hydration of solutes (Kemp *et al.*, 1990). These properties are reported in Table 5 for sucrose, maltitol, aspartame, alitame and their mixtures.

Mixing aspartame with sucrose or maltitol yields a suppression of sweetness intensity. The maximum of

Table 5. Viscosimetric and volumetric properties for 10% sucrose and maltitol solutions, 1% intense sweeteners solutions and their mixtures at 25 ± 0.02°C

	[η] (cm³.g⁻¹)	k′	V2° (cm³.g⁻¹)	h
Sucrose/aspartame	2.42	1.02	0.65	6.11
Sucrose/alitame	2.34	0.95	0.63	5.84
Sucrose/acesulfame K	2.36	0.96	0.63	6.03
Sucrose/cyclamate Na	2.28	1.23	0.66	4.71
Maltitol/aspartame	2.47	0.44	0.64	6.72
Maltitol/alitame	2.44	0.81	0.65	6.36
Maltitol/acesulfameK	2.38	1.16	0.64	6.03
Maltitol/cyclamateNa	2.32	1.25	0.65	5.38
Sucrose	2.37	1.15	0.62	6.14
Maltitol	2.45	0.92	0.62	6.85
Alitame	2.41	3.21	0.68	5.97
Aspartame	2.62	2.00	0.72	5.35
CyclamateNa	2.90	0.44	0.61	6.12
AcesulfameK	0.98	0.42	0.55	−1.8

negative synergy (or suppression) is −32.8% for sucrose/aspartame and −12.4% for maltitol/aspartame mixtures. Correlation of these results with solution properties, reported in Table 5, is based on the fact that reduced water mobility leads to reduced perceived intensity. Indeed the increase in intrinsic viscosity [η] and decrease in k′ (see Table 5) mean that more molecules of water are stuck to the solutes and that their exchange with bulk water is hindered, very likely under the hydrophobic effect of aspartame.

Adding alitame to sucrose or maltitol aqueous solutions yields nearly no change in the sweetness intensity of the mixture as calculated from additivity (see Tables 3 and 4). Such a sensory result may be correlated with the stability of intrinsic viscosity ([η]) and the slight decrease in Huggins constant (k′) reported in Table 5. Water mobility around sweeteners is not affected by mixing alitame with sucrose or maltitol and no appreciable synergy is observed.

For sugar/sodium cyclamate mixtures, a positive synergy is obtained at a level of 12.7% for sucrose and 27.4% for maltitol (see Tables 3 and 4). Sodium cyclamate influences the solution properties (increase in k′ and decrease in intrinsic viscosity [η]) in the direction of increased water mobility which seems to be the clue for enhancement of sweet taste.

Potassium acesulfam (ACK) does not show the same behaviour when it is mixed with sucrose or maltitol. Only additivity is observed for ACK/sucrose mixtures whereas positive synergy occurs for ACK/maltitol blends (see Tables 3 and 4). The positive synergy of ACK/maltitol mixtures seems to be a good match for the mobility of water revealed by an increase in k′ and a decrease in [η] (see Table 5). For sucrose/ACK mixtures, sweetness intensity shows an additive behaviour correlated with the comparable values of [η] and a decrease in k′.

For all studied mixtures, a slight increase in V_2° is observed, especially for cyclamate/sugar mixtures (0.66–0.65 cm³.g⁻¹ instead of 0.62 cm³.g⁻¹ for aqueous sugar solutions (Shamil *et al.*, 1987) (see Table 5). Hydration number h is decreased in the mixtures, especially cyclamate mixtures (see Table 5).

Interfacial properties

Results of surface tension, γ, at 20°C are shown in Fig. 3 for sucrose/sweeteners and maltitol/sweeteners at concentrations corresponding to maximum synergy, additivity or suppression (see Table 2). The surface tensions of sucrose (10%), maltitol (10%) and artificial sweeteners (1%) in water are listed in the same table. Sucrose and maltitol do not significantly change surface tension of water (73.9 and 73.8 mN/m against 73.7 mN/m for water). Artificial sweeteners have a noticeable effect on surface tension of water (63.5 and 65.8 mN/m, respectively, for alitame and aspartame). Surface tensions of mixtures are either unchanged or slightly augmented (for ACK/sucrose: γ = 73.6 mN/m and for ACK/maltitol: γ = 74.3 mN/m).

Contact angle (θ) which measures the affinity of a solution for a hydrophobic surface, is related to the hydrophobic character of the solute. The smaller the contact angle, the more the droplet is spread over

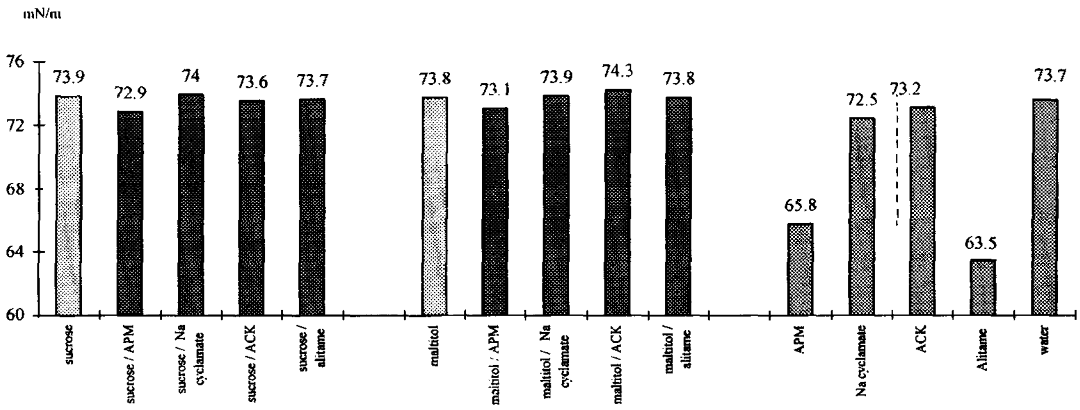

Fig. 3. Surface tension (mN/m) at 20°C for 10% sucrose and maltitol solutions, 1% intense sweeteners and their mixtures.

Table 6. Contact angle (°) of 10% sucrose and maltitol solutions, 1% intense sweeteners solutions and their mixtures at 20°C

Compound	Contact	Angle θ(°)
Sucrose	96.3	±1.4
	97.6	±1.2
Sucrose/aspartame		
Sucrose/alitame	95.9	±1.0
Sucrose/ACK	93.6	±0.6
Sucrose/Na cyclamate	90.8	±1.5
Maltitol	98.3	±1.2
Maltitol/aspartame	98.0	±0.9
Maltitol/alitame	97.5	±0.8
Maltitol/ACK	95.5	±0.8
Maltitol/Na cyclamate	95.3	±1.5
Alitame	73.7	±2.5
Aspartame	89.0	±2.3
ACK	92.1	±0.5
Na Cyclamate	94.5	±0.8
Water	100.0	±1.5

the support. Contact angle measurements on sucrose, maltitol, intense sweeteners and their mixtures are listed on Table 6. From these results, intense sweeteners studied can be classified as a function of their degree of hydrophobicity: alitame ($\theta = 73.7°$) > aspartame ($\theta = 89°$) > ACK ($\theta = 92.1°$) > sodium cyclamate ($\theta = 94.5°$). Sucrose and maltitol show higher values (96.3 and 98.3°) because of their mainly hydrophilic character. Contact angle data are standardized with θ for water = 100°.

For mixtures, the lowest θ values are observed when aspartame and alitame, hydrophobic components, are added to bulk sweetner solution (86.3 and 91.3°, respectively, for sucrose/aspartame and sucrose/alitame mixtures and 88.4 and 88.2°, respectively, for maltitol/aspartame and maltitol/alitame mixtures (Table 6). ACK also involves a decrease in θ but to a lesser extent ($\theta = 92.8°$ for sucrose/ACK mixtures and $\theta = 93.4°$ for maltitol/ACK mixtures). The presence of sodium cyclamate, weakly hydrophobic, does not significantly modify the hydrophobicity of sugar solutions (θ value close to that of the sugar). In this way, intense sweetener/sugar mixtures can be differentiated from each other by their relative hydrophobicity as follows: sugar/alitame > sugar/aspartame > sugar/ACK > sugar/sodium cyclamate. Finally each sweetener assigns to the solution a degree of hydrophobicity which is a function of its concentration and intrinsic properties.

CONCLUSION

Bulk and intense sweeteners can be used in binary combinations to take advantage of both functional properties of the bulk sweeteners and the intense sweetness of the high-potency sweeteners, which bring considerable benefits in terms of taste quality, processing and cost to the food industry. The synergistic effect observed when two components are in a mixture is specific and depends on the compatibility of the hydration of each component and their influence on water structure. This effect is attended by a specific influence of the presence of intense sweetener on physicochemical properties of sugars in solution.

The reduced perceived sweetness intensity observed with sugar/aspartame mixtures can be linked to the reduced water mobility in the medium. Conversely, an increase in water mobility leads to an enhancement of sweet taste (observed for sugar/cyclamate and maltitol/ACK mixtures). By mixing alitame with bulk sweeteners (sucrose or maltitol) and ACK to sucrose, water mobility is not affected and no appreciable synergy is observed (additivity). Investigation of the origin of the mechanisms responsible for the synergy or suppression phenomena is of relevance at an economic level to improve the choice of sugar/intense sweetener mixtures in industrial food formulations. Interpretation of the sweetness of the mixtures of sugars and intense sweeteners, by reference to their compatibility with water structure, may help in predicting the optimal formula and basing the choice on physicochemical parameters.

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REFERENCES

Beidler, L. M. (1962) Taste receptor stimulation. *Progress in Biophysics* **12**, 109–151.

Beidler, L. M. (1971) Taste receptor stimulation with salts and acids. In. *Handbook of sensory physiology*, Vol. 4, Chemical Sense—Part 2: Taste, ed. L. M. Beidler, pp. 200–221. Springer Verlag, New York.

Birch, G. G., Latymer, Z. and Holloway, M. R. (1980) Intensity-time relationships in sweetness: evidence for a queue hypothesis in taste chemoreception. *Chem. Senses* **5**, 63–78.

Birch, G. G., Ogunmoyela, G. and Munton, S. L. (1982) Synergism and the sweet response. In *Nutritive sweeteners*, eds G. G. Birch and K. J. Parker, pp. 275–285. Applied Science Publishers Ltd, Barking, UK.

Birch, G. G. (1996) Towards an improved understanding of sweetener synergy. *Trends in Food Sci. Technol.* **7**(12), 403–407.

Birch, G. G. (1997) The hydrostatic and hydrodynamic volumes of polyols and their sweet taste. *Chemical Senses* **22**, 149–161.

Carr, B. T., Pecore, S. D., Gibes, K. M. and Dubois, G. E. (1993) Sensory methods for sweetener evaluation. In *Flavour measurement*, eds C. T. Ho and C. H. Manley, pp. 219–256. Marcel Dekker Inc., New York.

Frijters, J. E. R., van der Klaauw, N. and Kranen, B. (1990) The use of molal concentration in a psychophysical investigation of Beidler's mixture equation. *Chem. Senses* **5**(6), 659–669.

Herkovitz, T. T. and Kelley, M. (1973) Viscosity studies of aqueous solutions of alcohols, ureas and amides. *J. Phys. Chem.* **77**, 381–388.

Huggins, M. L. (1942) The viscosity of dilute solutions of long chain molecules. IV Dependence on concentration. *J. Am. Chem. Soc.* **64**, 2716–2718.

Hyvönen, L., Kuekela, R., Koivistoinen, P. and Ratilainen, A. (1978) Fructose-saccharin and xylitol-saccharin synergism. *J. Food Sci.* **43**, 251.

Kemp, S. E., Birch, G. G., Portmann, M. O. and Mathlouthi, M. (1990) Intrinsic Viscosities and Apparent Specific Volumes of Amino-Acids and Sugars. "Effective Size" and Taste of Sapid Molecules. *J. Sci. Food Agric.* **51**, 97–107.

Lotz, A. and Meyer, E. (1994) Sweeteners in beverages: new development. *Int. Food Market. Technol.*, 4–9.

Mathlouthi, M. and Seuvre, A. M. (1988) Solution properties and the Sweet Taste of Small Carbohydrates. *J. Chem. Soc., Faraday Trans. I* **84**, 2641–2650.

Mathlouthi, M. and Portmann, M. O. (1990) Hydrogen bonding and the sweet mechanism. *J. Mol. Struct.* **237**, 327–338.

Mathlouthi, M., Bressan, C., Portmann, M. O. and Serghat, S. (1993) *Role of water structure in sweet taste chemoreception.* eds M. Mathlouthi, J. A. Kanters and G. G. Birch, pp. 141–174. Elsevier Applied Science, London, UK.

Mathlouthi, M. and Portmann, M. O. (1994) Solution properties and the sweet taste of the mixtures. In *Synergy*, eds G. G. Birch and G. Campbell-Platt, pp. 63–80. Intercept Ltd., UK.

Mathlouthi, M., Hutteau, F. and Angiboust, J. F. (1996) Physicochemical properties and vibrational spectra of small carbohydrates in aqueous solution and the role of water in their sweet taste. *Food Chem.* **56**(3), 215–221.

Portmann, M. O. and Kilcast, D. (1996) Psychophysical characterisation of new sweeteners of commercial importance for the EC food industry. *Food Chem.* **56**(3), 291–302.

Shamil, S., Birch, G. G., Mathlouthi, M. and Clifford, M. N. (1987) Apparent Molar Volumes and Taste of Molecules with more than one Sapophore. *Chem. Senses* **12**(2), 397–409.

van der Tornout, P., Pelgroms, J. and van der Meeren, J. (1985) Sweetness evaluation of mixtures of sucrose with saccharin, aspartame or acesulfam-K. *J. Food Sci.* **50**, 469.

Weikmann, F., Weikmann, H, Fincke, K., Weikmann, F. A. and Huber, B (1969) Deutsche Laevosan-Gesellschaft Boehringer, Mannheim German Patent 1961769.