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Influence of carboxymethyl cellulose and sodium alginate on sweetness intensity of Aspartame

Xue Han [a], Shu-Zhen Xu [a], Wen-Rui Dong [a], Zhai Wu [b], Ren-Hai Wang [b], Zhong-Xiu Chen [a,*]

[a] Department of Food Quality and Safety, College of Food & Biology Engineering, Zhejiang Gongshang University, Hangzhou, Zhejiang 310035, PR China
[b] Department of Chemistry, College of Food & Biology Engineering, Zhejiang Gongshang University, Hangzhou, Zhejiang 310035, PR China

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ABSTRACT

Sensory evaluation of Aspartame in the presence of sodium carboxymethyl cellulose (CMC-L) and sodium alginate (SA) revealed that only CMC-L showed a suppression effect, while SA did not. By using an artificial taste receptor model, we found that the presence of SA or CMC-L resulted in a decrease in association constants. Further investigation of CMC-L solution revealed that the decrease in water mobility and diffusion also contribute to the suppression effect. In the case of SA, the decreased viscosity and comparatively higher amount of free water facilitated the diffusion of sweetener, which might compensate for the decreased binding constant between Aspartame and receptor. This may suppress the impact of SA on sweetness intensity. The results suggest that exploring the binding affinity of taste molecules with the receptor, along with water mobility and diffusion in hydrocolloidal structures, provide sufficient information for understanding the mechanism behind the effect of macromolecular hydrocolloids on taste.

1. Introduction

The impact macromolecular hydrocolloids have on the sensory properties of food has been the subject of repeated investigations. This is a challenging area because food systems are composed of a complex mixture of protein, polysaccharide and other additives. Aspartame is an artificial, non-saccharide sweetener, widely used as a sugar substitute in foods and beverages. The sweetness intensity of Aspartame is reduced by macromolecules such as hydroxypropylmethyl cellulose, carboxymethyl cellulose, gellan gum, oat gum, and guar gum (Bayarri, Izquierdo, & Costell, 2007; Cook, Hollowood, Linforth, & Taylor, 2002; Mälkki, Heiniö, & Autio, 1993). Current research about the effects of macromolecular hydrocolloids on Aspartame mainly focus on the change of perceived sweetness by sensory evaluation, but the detailed mechanisms involved in the suppression of the sweetness intensity are not fully understood.

The first property of food macromolecules, which could possibly be related to taste suppression, is viscosity. Increasing the viscosity of solutions often results in a decreased taste intensity. For example, Portmann, Serghat, and Mathlouthi (1992) found that the sweetness intensity decreased as the viscosity of D-fructose and sucrose solutions increased. Christensen (1980) showed that CMC-L-thickened solutions produced little or no suppression of perceived taste intensity in sucrose solutions whereas viscous CMC-H solutions produced significant reductions in perceived saltiness and sweetness. Moskowitz and Arabie (1970) found that the increase in viscosity of the aqueous solvent by sodium carboxymethyl cellulose resulted in decrease in taste intensity for glucose, citric acid, sodium chloride and quinine sulphate. Besides the viscous hydrocolloidal solution, the structural change of the macromolecules might also account for the suppression effect. When the concentration of macromolecules is higher than its coil-overlap concentration (C^*), the chains are forced to interpenetrate and form an entangled network which results in inefficient mixing of the tastant in solution (Baines & Morris, 1987; Cook et al., 2002; Koliandris, Lee, Ferry, Hill, & Mitchell, 2008). Sweetness also decreased with increasing gel hardness. Boland, Delahunty, and van Ruth (2006) observed that increased gel rigidity resulted in decreased perception of odour, strawberry flavour and sweetness. Bayarri, Durán, and Costell (2003) found that the decrease in sweetness which comes from increased concentration of hydrocolloid was greater in gellan than in carrageenan gels. Texture–taste interactions, which focused on the binding of tastants to the thickener, were further proposed to elucidate the effect of macromolecules on the taste by correlating the sensory perception with analytical measurement of the physical or chemical properties of

* Corresponding author. Tel.: +86 13735802398; fax: +86 571 88071024.
E-mail address: zhxchen@ustc.edu (Z.-X. Chen).

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food (Burseg, Camacho, & Bult, 2011; Cook, Hollowood, Linforth, & Taylor, 2005; Yven, Guichard, Giboreau, & Roberts, 1998). Salivation effects were also considered to be influenced by hydrocolloids since the sweet stimulus goes through different layers of saliva before it can access to the receptor membrane (Portmann et al., 1992; Stokes, Boehm, & Baier, 2013). These work provided useful information for understanding the initiation of the suppression of taste by macromolecular hydrocolloids from different aspects, such as solution environment, structure of macromolecules, texture–taste interactions and salivation effects. However, it appears there has been little reported on the change of binding affinity of sweetener with the taste receptor induced by food hydrocolloids.

The initial event of sweetness sensation generally involves stereospecific binding of sweet compounds to the sweet taste receptor. The degree of sweetness depends on how well the receptors in our tongue interact with sugar molecules. Therefore, the relationship between the chemical structures of sweeteners with various sweetness intensities and their interactions with the taste receptor has always been a subject of keen interest to chemists. In an effort to rationalise these relationships, biochemists attempt to express the taste receptor and provide functional analysis of ligand binding. Chemists have built models for the sweet taste receptor theoretically because the crystal structure of the receptor protein is not known. Early models were exclusively pharmacophore models, but receptor models, even computational models with predictive capability, were subsequently developed (DuBois, 2011). We believe that chemical interactions are the cornerstone of biological function. Simplified models have often been used to investigate aspects of more complex biological systems. In recent years, we tried to use an artificial sweet taste receptor model for the biomimetic research of the interaction between sweeteners and receptors. We have found that thermodynamic parameters correlate well with the sweetness intensity of some sweeteners (Chen, Guo, & Deng, 2009). The thermodynamic basis for the initiation of sweetness inhibition as well as that for the sweetness differences between the enantiomers of amino acids and monosaccharides were also proposed recently (Chen, Wu, Zhang, & Deng, 2011; Dong, Chen, Chen, & Deng, 2013). The continued interest in the thermodynamics of sweet taste sensation motivated us to study the increased viscosity on binding constants, which likely accounts for taste suppression. In this paper, we used sodium carboxymethyl cellulose (CMC-L) and sodium alginate (SA) with similar viscosity to produce the hydrocolloidal solution for investigating the change of the binding affinity of Aspartame and the thermodynamic parameters involved in the binding process. Water mobility as well as diffusion was further investigated with the aim of finding the possible mechanism behind the effect of macromolecular hydrocolloids on the sweetness intensity of Aspartame.

2. Materials and methods

2.1. Materials

Aspartame (98%), sodium alginate (90%), sodium carboxymethyl cellulose (M.W. 9000, DS = 0.7, 50–100 mPa s), and anhydrous sodium sulphate (>99%) were purchased from Sigma–Aldrich. Potassium ferrocyanide (>99.5%) was obtained from National Pharmaceutical Group Corporation of China. C_{60} was provided by from the Henan Yongxin Company of China. Polyhydroxylated fullerene was synthesized according to a reported method (Li et al., 1993). Deionized water was used for preparation of all solutions.

2.2. Sensory evaluation

Sensory evaluation was conducted to estimate the sweet intensity level of Aspartame in viscous systems. A panel of eight subjects (four males and four females, aged 23–24 years), was recruited from the graduate students of the Department of Food Science and Technology at Zhejiang Gongshang University. The students were provided training in the use of the labelled magnitude scales to evaluate the sweet intensity. Six scales were designated in the experiment, namely strongest imaginable, very strong, strong, moderate, weak, barely detectable. The sweet intensity of solution was recorded on a scale of 0–100.

A series of concentrations of Aspartame (125, 250, and 500 mg/L) in different viscous solutions of SA (0, 0.4, 1.2, and 3.6 g/L) or CMC-L (0, 2.5, 7.5, and 15 g/L) were evaluated. Sample solutions (5 mL) were transferred to a plastic beaker. All the samples were placed in a predetermined sequence. Assessors were asked to place the test compound into their mouths for 2 s, and then expectorate and rinse their mouths with distilled water. Subjects were asked to pause 10 s after each test. Each sample was tested in triplicate.

2.3. Isothermal titration calorimetry

An isothermal titration calorimeter (VP-ITC, Microcal Inc., Northampton, MA) was used to measure enthalpies from titrations of Aspartame into fullerenols with CMC-L or SA and sodium chloride (NaCl) at 298 K. In an individual experiment, the cell (1.43 mL) was loaded with fullerenol solution (0.1 mmol/L) and the syringe with Aspartame solution (5 mmol/L). The reference cell was filled with deionized water. All of the solutions were prepared in deionized water and were degassed before use. The first injection was set to 1 μL, and then the titration was performed in 27 injections of 10 μL at intervals of 360 s. The duration of each injection was 2 s, and the solution in the titration cell was stirred at a speed of 307 rpm throughout the experiment. Control experiments included the titration of Aspartame into water/hydrocolloid and water/hydrocolloid into fullerenol solution. The first titration point was removed from the data before fitting. The data were fitted without subtracting the control due to the negligible heat released in the dilution. Raw data were obtained as a plot of heating rate (μcal s^{-1}) versus time (min). Integration was carried out to obtain a plot of observed enthalpy change per mole of injected fullerenols (ΔH, kcal mol^{-1}) against molar ratio (Aspartame/fullerenols). Equilibrium association constants and enthalpies were determined by fitting the data with a sequential binding site model by MicroCal Origin 7.0 software using a nonlinear least-squares approach (Levenberg–Marquardt algorithm).

2.4. Cyclic voltammetry measurements

Cyclic voltammetry (CV) measurements were carried out with cyclic voltammeter CHI 103 electrochemical working station (Chenhua Instrument Co., China). Three electrode systems consist of a working gold electrode, a reference saturated KCl (3.5 mol/L) electrode, and the counter electrode (a Platinum wire). The working electrode was polished with alumina powder (slurry) and washed thoroughly with deionized water before each measurement. The working electrode area was determined using cyclic voltammetry experiments on a reversible system (4 mmol/L $K_4Fe(CN)_6$ in 0.1 mol/L Na_2SO_4). By use of the diffusion coefficient $D = 6.3 \times 10^{-10}$ m^2 s^{-1}, the electrode area was determined as $A = 2.54$ mm^2, which is consistent with the reported value (Chen, Deng, & Li, 2008). The potential was scanned between −1 and +1 V, and the sweep rate range used was 0.05–0.5 V s^{-1}. Potassium ferrocyanide (4 mmol/L) was used as the electroactive probe and Na_2SO_4 (0.1 mol/L) was used as the supporting electrolyte in each sample. Sample solutions of SA or CMC-L at a series of concentrations were prepared and reported data are an average of three measurements.