Sensors and Materials, Vol. 13, No. 3 (2001) 137–144
MYU Tokyo

S & M 0437

Analysis of Saltiness and Bitterness of Inorganic Salts Using Taste Sensors

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(Received August 22, 2000; accepted March 11, 2001)

Key words: taste sensor, lipid membranes, response pattern, inorganic salts, saltiness, bitterness, adsorption

The response of a taste sensor to inorganic salts was investigated. It is known that in a series of halides of monovalent alkali metals, salts with low molecular weights are salty, whereas those with high molecular weights are bitter. Salty substances such as NaCl changed the potential of positively charged membranes of the sensor in the same way as negatively charged ones. On the other hand, bitter substances affected both membranes differently. For example, inorganic salts which contain iodine ions markedly changed the potential of positively charged membranes. Large changes in membrane potential were induced by the adsorption of taste substances to lipid membranes. Hence a bitter taste seems to be attributable to adsorption.

1. Introduction

Taste can be measured with a multichannel lipid-membrane sensor, which responds to different taste qualities by unique patterns of sensor output signals.[1–7] The sensor utilizes lipid membranes as transducers of tastes and a computer as a data analyzer. The transducers play a role of transforming taste information generated by chemical substances into electric potential changes. As a result, the sensor can easily distinguish the qualities of the five basic tastes, namely sour, bitter, sweet, salty and umami, and also some beverages such as beer, coffee and aqueous drinks. The sensor has sensitivity, durability and reproducibility superior to humans.

All substances with a salty taste are soluble salts composed of positive and negative ions in the solid state which dissolve into water to produce a solution of these ions. The tastes of salts are complicated; they change with concentration. Sodium chloride is the only chemical considered to possess the pure salty taste, although under its threshold concentration it tastes sweet. Other salts display the same phenomenon but yield complex salty tastes at suprathreshold values.[8]

Both the anion and cation contribute to taste quality and to the stimulating efficiency. In a series of sodium salts, the quality of taste elicited varies with the anion. A similar effect can be noted in a chloride series with different cations. It is known that in a series of halides of monovalent alkali metals, salts with molecular weights below 110 are predominantly salty, whereas those with molecular weights over 160 are mainly bitter.[9]

The relationships between the structure of chemical substances and their taste have been studied extensively.[10,11] Bitter substances are generally hydrophobic; hence some of them are considered to adsorb to hydrophobic parts of lipid membranes.[12] Adsorption of bitter substances to lipid membranes has been employed to build up quartz-crystal microbalances.[13] The structures of inorganic salts are simple; hence it is easier to examine their tastes than those of other chemicals. In this study, the responses of a taste sensor to inorganic salts are compared with sensory tests. The bitter taste of inorganic salts seems to be attributable to adsorption to lipid membranes.

2. Materials and Methods

2.1 *Chemicals*

Commercial products were used without further purification. All of the halides of monovalent alkali metals and sodium picrate were obtained from Wako, and quinine hydrochloride was obtained from Sigma. Inorganic salts and bitter substances were dissolved in 1 mM KCl solution.

The lipids are abbreviated as follows: dioctyl phosphate, C; trioctyl methylammonium chloride, T; oleyl amine, N; decyl alcohol, DA; oleic acid, OA. Lipid membranes designated C:T=9:1, C:T=3:7 and C:T=5:5 are mixtures of two lipids for which the ratio shows the molar concentration. In the membranes C:T=3:7, T and N are positively charged, whereas in C:T=9:1, C and OA are negatively charged. The membrane DA is somewhat negatively charged due to the presence of plasticizer.[14] The membrane C:T=5:5 is almost totally neutral.

2.2 *Measurements using a taste sensor*

The taste sensor used in this study was similar to one previously reported.[1–7] The detecting electrode of each lipid membrane was made of Ag wire whose surface was plated with Ag/AgCl and which was embedded in a basal acrylic board 2 mm thick. Another 1-cm-thick acrylic board with eight cone-shaped holes was fixed to the board. The holes were filled with 100 mM KCl solution, and the eight membranes were fitted on the board to cover the holes.

The electric potential across the membrane was detected by a Ag/AgCl electrode filled with 100 mM KCl and a reference electrode (TOA, HS205C). The construction of the measuring system is as follows: Ag/AgCl electrode in 100 mM KCl solution | membrane | reference electrode in taste solution. The membrane potential was changed by applying taste substances, and the electric signal from each membrane was converted into a digital code using a digital voltmeter (ADVANTEST, R6551) through a high-input impedance amplifier and a laboratory-built eight-channel scanner, and recorded in a computer (NEC, PC-9801).

3. Results

3.1 *Response of the sensor to salty and bitter substances*

Prior to describing the study of the response of the sensor to halides of alkali metals, let us outline the fundamental properties of the sensor. Figure 1 shows the responses of eight membranes of the sensor to salty NaCl. The responses were measured relative to 1 mM KCl solution. Cations and anions exist homogeneously due to thermal motion in the aqueous solution of NaCl. However, negative charges of the membranes tend to attract cations and repel anions, producing a diffuse electrical double layer. Therefore, the negative charges of C, C:T=9:1, DA and OA membranes are reduced by the shielding

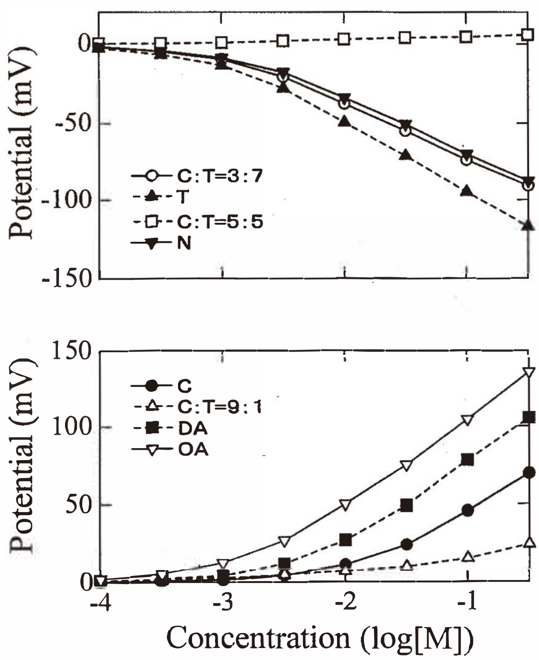

Fig. 1. Response curves of taste sensors to NaCl.

effect of sodium ions, thus increasing the membrane potential. On the other hand, the positive charges of C:T=3:7, T and N membranes are reduced by the shielding effect of chloride ions, thus decreasing the membrane potential. Consequently, salty substances such as NaCl change the potential of positively charged membranes of the sensor in the same manner as negatively charged ones.

Quinine hydrochloride is a typical bitter substance, which has a positive charge and a hydrophobic portion. The response of the sensor to it is shown in Fig. 2. It markedly changed the potential of negatively charged membranes, and its threshold concentration was 0.001 mM. The large potential change is based on the adsorption onto the hydrophobic part of the lipid membrane, as quantitatively shown using electrochemical theory.[15] The Cl^- ion has only a shielding effect. The threshold concentration was about 1 mM.

Another bitter substance is sodium picrate. Picric acid is a strong acid, because it has three electrophilic nitro groups. As shown in Fig. 3, it significantly changed the potential of the positively charged membranes, such as C:T=3:7, T, C:T=5:5 and N; therefore, its conjugate base is effective. At concentrations of 0.3 mM and 3 mM, the potential of the C:T=3:7 membrane changed by approximately 300 mV, which is higher than the theoretical value of 60 mV.

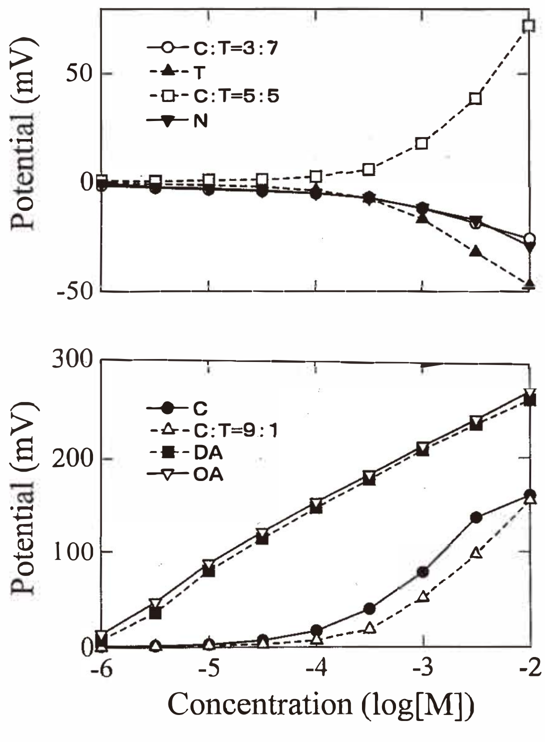

Fig. 2. Response curves of taste sensors to quinine hydrochloride.

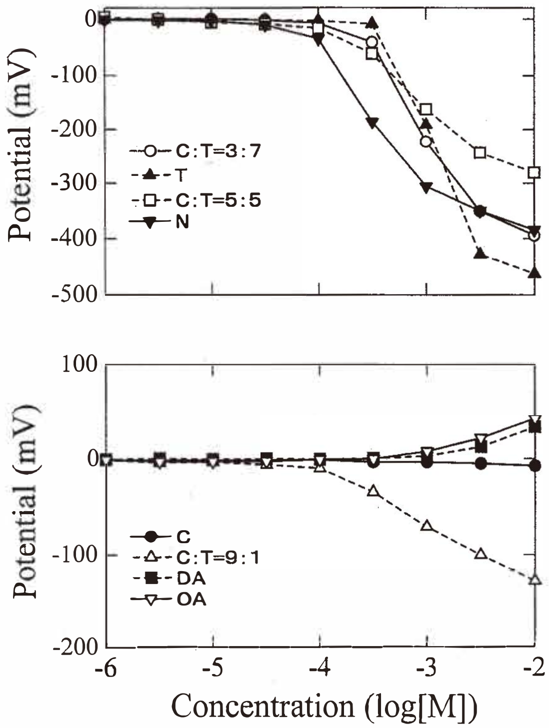

Fig. 3. Response curves of taste sensors to sodium picrate.

The common profile in Figs. 2 and 3, which show the responses to bitter quinine hydrochloride and sodium picrate, may be an imbalanced response between the two charged membranes. In fact, quinine hydrochloride and sodium picrate mainly changed the potential of negatively charged membranes and of positively charged membranes, respectively. This suggests that adsorption rather than the shielding effect plays an important role in the response of the sensor to bitter substances.

3.2 *Response of the sensor to various inorganic salts*

To elucidate the relationship between taste qualities and kinds of inorganic salts, the response of the taste sensor to several salts was measured. Figure 4 shows the responses of negatively charged membranes of the sensor to the chloride series with different cations, namely LiCl, NaCl, KCl, RbCl and CsCl. The result for Na^+ is omitted because it is shown in Fig. 1. The responses of the sensors to monovalent cations were not always the same. The potential change was large in the following order: $Li^+ > Na^+ > K^+ > Rb^+ > Cs^+$, coinciding with the order of decreasing crystal radii.

The responses of the positively charged membranes to the sodium series with different anions, namely NaF, NaCl, NaBr and NaI, are shown in Fig. 5. The responses of the

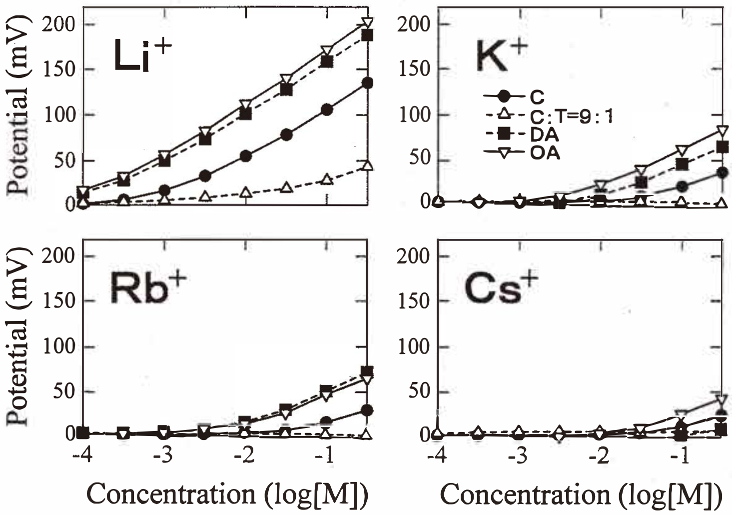

Fig. 4. Response curves of taste sensors to various monovalent cations.

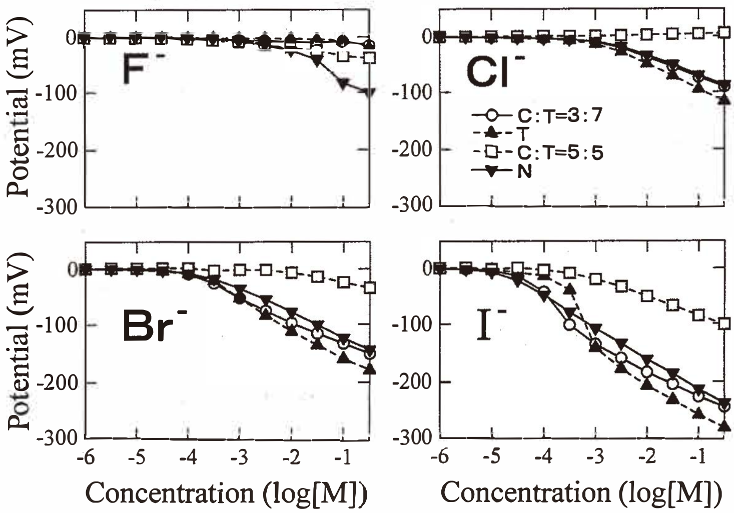

Fig. 5. Response curves of taste sensors to various monovalent anions.

sensors to monovalent anions also differed depending on the species. The change in the membrane potential increased in the following order: $I^- > Br^- > Cl^- > F^-$. In contrast to cations, this is the same as the order of increasing crystal radii.

3.3 *Adsorption of inorganic salts to lipid membranes*

Anions of high atomic weights such as Br^- and I^- markedly changed the potential of positively charged membranes; therefore, they may adsorb to hydrophobic membranes. If the ions adsorb to the membranes, their after-effect will be detected when the membranes are washed with 1 mM KCl solution.

Table 1 summarizes the potentials of positively charged membranes which were dipped into 100 mM NaF, NaCl, NaBr and NaI, and the potentials of the membranes which were transferred into 1 mM KCl. If these ions have only a shielding effect, the potential values in 1 mM KCl should be zero, because the data were obtained relative to 1 mM KCl. But as shown in the table, the effect of pretreatment with inorganic salts remains even in 1 mM KCl solution, indicating the occurrence of adsorption.

4. Discussion

Eight lipid membranes of the taste sensor are roughly divided into negatively charged membranes and positively charged ones. Salty NaCl affected both membranes equally by the shielding effect (Fig. 1). Contrary to this, negatively charged membranes unilaterally responded to bitter quinine hydrochloride. In the same manner, positively charged membranes markedly responded to bitter sodium picrate. The large potential changes exceed the theoretical value of 60 mV; hence this effect may be attributable to adsorption. The responses of the two types of membranes to bitter substances are not balanced because of adsorption. Moreover, two types of bitter substances must exist: one which mainly affects negatively charged membranes; the other, positively charged ones.

It is known that in a series of halides of monovalent alkali metals, salts with low molecular weights are salty, whereas those with high molecular weights are bitter.[9] Therefore, the responses of the taste sensor to various inorganic salts were investigated. It was revealed that the responses to cations are large and occur in the order $Li^+ > Na^+ > K^+ > Rb^+ > Cs^+$, whereas those to anions are large and occur in the order $I^- > Br^- > Cl^- > F^-$ (Figs. 4 and 5). Combinations of cations with high atomic weights, such as Rb^+ and Cs^+, and anions with high atomic weights, such as Br^- and I^-, may primarily affect the positively

Table 1
After-effect of various inorganic salts on positively charged membranes (unit, mV).

Treatment \ Membrane	C:T=3:7	T	C:T=5:5	N
100 mM NaF	−1.6	−12.2	−31.1	−58.2
1 mM KCl	2.6	4.1	2.5	2.6
100 mM NaCl	−89.3	−93.9	−10.2	−82.5
1 mM KCl	−5.6	−11.7	−9.2	−13.1
100 mM NaBr	−148.9	−161.1	−25.7	−140.9
1 mM KCl	−21.8	−34.3	5.4	−23.7
100 mM NaI	−221.9	−263.6	−94.4	−219.1
1 mM KCl	−82.4	−59.4	−18.5	−98.4

charged membranes of the sensor, which resembles the effect of sodium picrate. This result explains why inorganic salts with high molecular weights are bitter.

The order of responses of the sensors to cations and anions were opposite with respect to the crystal radii. Electronegativity seems to explain the order of responses best. Electronegativity values for anions are as follows: I^- 2.5, Br^- 2.8, Cl^- 3.0, F^- 4.0.[16] The value for a positively charged membrane is unknown; the value of Na^+ 0.9 could be substituted tentatively for that of the membrane. Differences in electronegativity between cations and anions in NaI, NaBr, NaCl and NaF are 1.6, 1.9, 2.1 and 3.1, respectively. The difference in NaI is the smallest; hence, the interaction between Na^+ as the positively charged membrane and I^- must be stronger than the others.

The taste sensor was developed to evaluate the tastes of foods objectively. Moreover, it was very useful in revealing that the taste of astringency consists of both chemical and physical stimulations, whereas the sense has been recognized as a physical one, namely a tactile sensation.[3] The further application of the sensor to the study of taste is expected.

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