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Selectivity Control in a Sweetness Sensor Using Lipid/Polymer Membranes

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We studied the selectivity control in a sweetness sensor with lipid/polymer membranes. Sweet tasting substances such as sucrose, fructose and glucose are nonelectrolytes and therefore are susceptible to interference from electrolytes and/or adsorptive substances. In this study, we focused on suppressing the adsorption of bitter and astringent substances on a membrane surface. The membrane was designed to be electrically neutral to avoid interference from electrolytes. Additives used in this experiment have a hydroxyl group and are not protonated compounds, which change the hydrophobicity character of a membrane. The results show that *n*-tetradecyl alcohol reduced the responses to bitter and astringent substances.

1. Introduction

A multichannel taste sensor with several lipid/polymer membranes with different characteristics can detect tastes in a manner similar to the human gustatory system. Information from substances producing taste is transformed into electric signals, which are entered into a computer. The sensor output shows different patterns for chemical substances that have different taste qualities such as saltiness and bitterness, while it shows similar patterns for chemical substances with similar taste sensations.(1)

However, the electric responses for nonelectrolytes such as sucrose were one-fifth to one-tenth of those for electrolytic with substance taste. In our previous study, we developed a sweetness sensor with a lipid/polymer membrane.(2) However, the sensor does not have enough selectivity for sweet tasting substances. The sensor responds not only to sweeteners but also to bitter, astringent and umami tastes. In this study, we focused on suppressing the responses to bitterness and astringency. The adsorption of bitter and astringent substances on the membrane surface is generally caused by hydrophobic interactions. We used materials containing hydroxyl groups in order to control the selectivity for sweetness.

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2. Materials and Methods

A schematic diagram of the taste-sensing system SA402B (Intelligent Sensor Technology, Inc.) is shown in Fig.1. The detector of the sensor usually consists of eight kinds of electrodes with different lipids/polymers. The sensor detects taste information which is transformed into electric signals that correlate with membrane potentials. Depending on the substances to be measured, different lipid/polymer membranes are available.

Materials used to prepare a sweetness sensor in this study are listed in Table 1.[2] Lipids were mixed with polyvinyl chloride (PVC) and a plasticizer (DOPP:dioctyl phenylphosphonate) dissolved in tetrahydrofuran and then dried on a glass plate. The lipid/

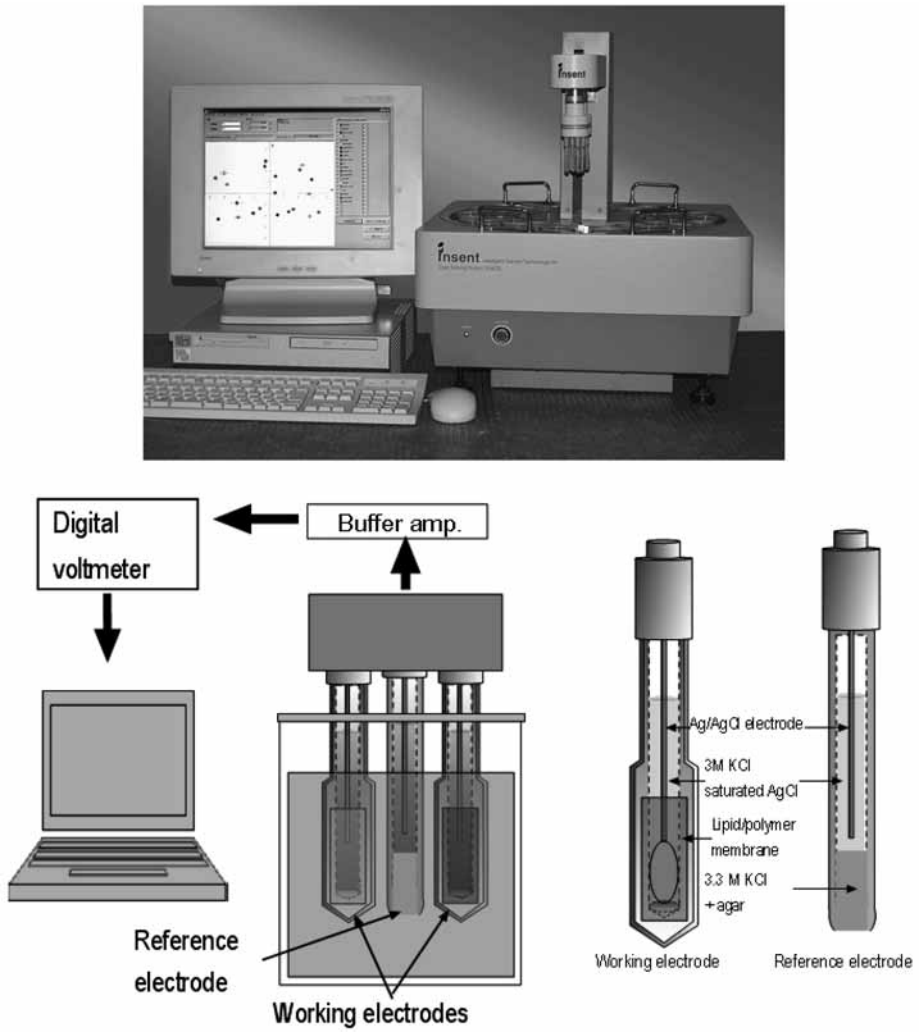

Fig. 1 Schematic diagram of experimental setup.

Table 1
Materials used for membranes.

abbr.	composition
TDAB	tetradodecylammonium bromide, 1.50 mg
DOPP	dioctyl phenylphosphonate, 1.0 ml
PVC	polyvinyl chloride, 800 mg

polymer membrane is a transparent, soft film approximately 200 μm thick. Then the membrane was preconditioned with 0.05 wt% tannic acid and 10 mM KOH+30% EtOH+100 mM KCl solution.

A multichannel electrode was connected to eight channels through high-input impedance amplifiers. The electric signal from the sensor was converted to a digital code by an A/D converter and was entered into a computer. The electrode was made of an Ag wire, the surface of which was plated with AgCl, in an internal cavity filled with 3.3 M KCl and some excess AgCl. Then the voltage difference between the multichannel electrode and the Ag/AgCl reference electrode was measured.

Figure 2 shows the procedure of measurement. *V*c is the electric potential of the control, and *V*s is the potential of sample. Therefore, the sensor output should be the value (*V*s–*V*c). The control is composed of 30 mM KCl and 0.3 mM L(+)-tartaric acid. After measuring the sample, washing the membrane removes the adsorbed substance and refreshes the membrane. Table 2 shows the control and the taste samples which consist of typical taste substances used in this experiment.

3. Results and Discussion

Sweet tasting substances such as sucrose, fructose and glucose are nonelectrolytes and are susceptible to interference by electrolytes when membrane potential is measured. Therefore, a membrane was designed to be electrically neutrality to obtain higher sensitivity for sweeteners. Additives used in this experiment are not protonated compounds, and it was expected that they had little interference on membrane potential. Table 3 shows the chemical formulas of lipids added to form membranes. Since they have a hydroxyl group and are not dissociated, it is expected that they do not change the membrane potential and increase the hydrophilicity in order to avoid hydrophobic interactions from other substances that have bitter and astringent tastes.

Initially, we added n-heptyl alcohol (C6), n-tetradecyl alcohol (C13) and stearyl alcohol (C17) into the membrane forming materials listed in Table 1 and examined the response to sucrose. Each additive has a hydroxyl group, which would increase the hydrophilicity, but different lengths of carbon chain. The vertical axis (Fig. 3) shows the electric response obtained from a taste sensor and the horizontal axis is the concentration of additive. As shown in Fig.3, the electric response for sucrose was changed as the amount of additive increased. From this result, 1.50 mg TDAB+100 mg C6, 1.50 mg TDAB+100 mg C13, and 1.50 mg TDAB+30 mg C17 were sensitive to sucrose and were examined for sensitivity to basic tastes listed in Table 2.

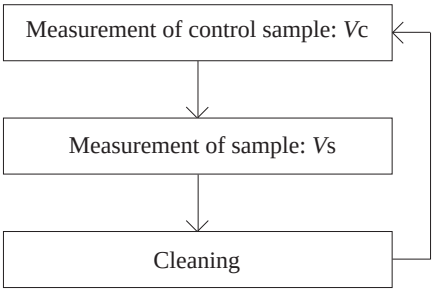

Fig. 2 Measurement procedure.

Table 2
List of sample used in this experiment.

Sample	composition
control	30 mM KCl+0.3 mM L(+)-tartaric acid
saltiness	300 mM KCl+0.3 mM L(+)-tartaric acid
sourness	30 mM KCl+3 mM L(+)-tartaric acid
umami	10 mM MSG+control sample
bitterness 1	0.1 mM quinine+control sample
bitterness 2	0.01 vol% iso α acid+control sample
astringency	0.05 wt% tannic acid+control sample
sweetness	1 M sucrose+control sample

Table 3
Structural formulas of additives to change the hydrophobicity of membranes.

addr.	composition	structural formula
C5	*n*-hexyl alcohol	CH_3-$(CH_2)_5$-OH
C6	*n*-heptyl alcohol	CH_3-$(CH_2)_6$-OH
C13	*n*-tetradecyl alcohol	CH_3-$(CH_2)_{13}$-OH
C17	stearyl alcohol	CH_3-$(CH_2)_{17}$-OH

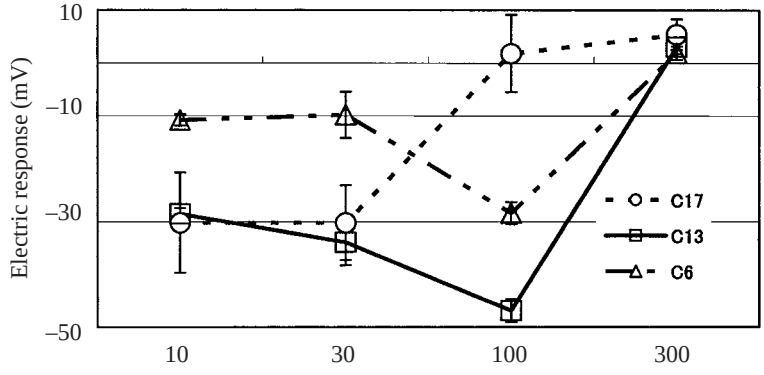

Fig. 3 Concentration of additives (mg).

Figure 4 shows the electric responses to basic tastes using membranes such as 1.50 mg TDAB+100 mg C6, 1.50 mg TDAB+100 mg C13, and 1.50 mg TDAB+30 mg C17. By adding C13, the sensitivity to bitterness and astringency was dramatically decreased. The adsorption of bitter and astringenct substances on the membrane surface is generally caused by hydrophobic interactions. From this result, the hydroxyl group and length of carbon chain of C13 has the optimum effect on reducing hydrophobic interactions.

Next we optimized the composition of the membrane in order to obtain a higher output for sucrose. We changed the volume of tetradodecylammonium bromide (TDAB) from 1.0 mg to 2.0 mg and *n*-tetradecyl alcohol (C13) from 10 mg to 300 mg. As shown in Fig. 5, only the composition with 1.50 mg TDAB showed a response to sucrose, and the highest response was for 100 mg C13.

Figure 6 is a comparison of the output of the newly developed sensor with the original one. Although the responses to sourness and umami were increased slightly, the outputs for bitterness and astringency were dramatically reduced. Furthermore, the sensitivity for sweetness was twice as high as for the original sensor.

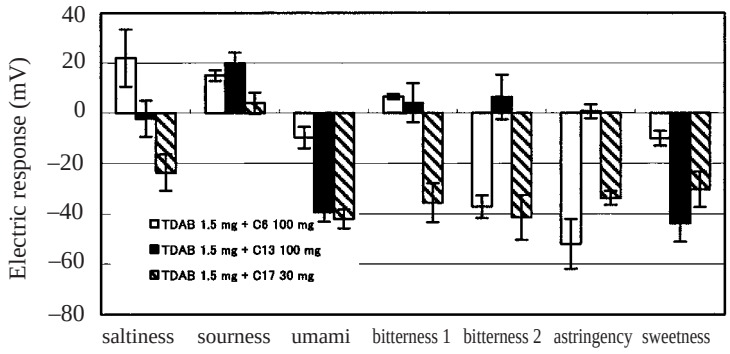

Fig. 4 Electric responses to basic taste substances.

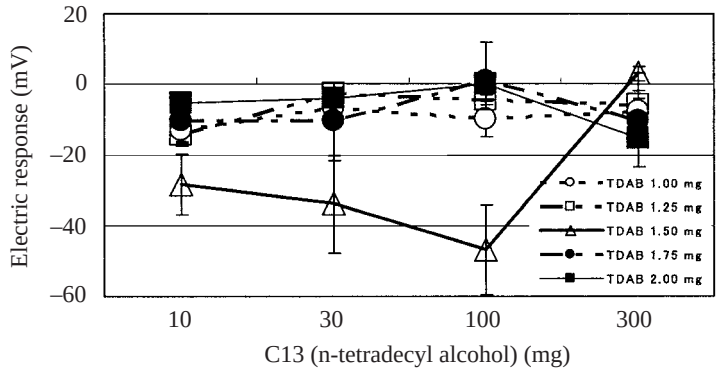

Fig. 5 Sensitivity for sucrose with changing concentration of *n*-tetradecyl alcohol.

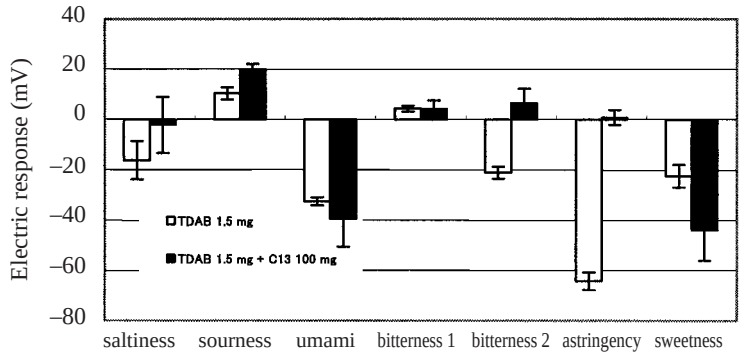

Fig. 6 Electric responses to basic taste substances.

4. Conclusions

We studied the selectivity control for sweetness sensor with lipid/polymer membranes. In this study, we focused on suppressing the adsorption of bitter and astringent substances on the membrane surface. Because the adsorption of bitterness and astringency is generally caused by hydrophobic interactions, we used materials containing a hydroxyl group to change the hydrophobicity of the membrane. From these results, it was found that n-tetradecyl alcohol was useful to control selectivity in measuring sweet tasting substances.

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