Sensors and Materials, Vol. 15, No. 3 (2003) 155–163
MYU Tokyo

S & M 0517

Study of Saltiness Using Taste Sensor with Different Lipid/Polymer Membranes

Ronggang Chen, Masaaki Habara and Kiyoshi Toko

Graduate School of Information Science and Electrical Engineering,
Kyushu University
6-10-1 Hakozaki, Higashi-ku, Fukuoka 812-8581, Japan

(Received August 22, 2002; accepted January 9, 2003)

Key words: taste sensor, saltiness, bittern, aftertaste, principal component analysis

We evaluated the saltiness of sodium chloride with coexisting bitterns such as magnesium sulfate, magnesium chloride and calcium chloride. Model samples which consist of sodium chloride and bitterns were measured using a taste sensor with lipid/polymer membranes. Standard data of saltiness were obtained from the sensor output in order to evaluate taste quantitatively and objectively. We proposed an evaluation model of saltiness based on sensor outputs and a human sensory test. The method enabled us to evaluate the taste of salts on the market with a trace of bitterns depending not only on the concentration of Mg^{2+} or Ca^{2+}, but also on their ratio.

1. Introduction

A multichannel taste sensor which has several types of lipid/polymer membranes with different characteristics can detect taste in a manner similar to the human gustatory sensation. Information for substances producing taste is transformed into electric signals using the sensor.[1,2,3]

The taste of salts depends on the manufacturing processes for different compositions. Matsumoto reported that bitterns such as $MgCl_2$, $MgSO_4$, $CaSO_4$ and KCl affect the human gustatory sensation of saltiness.[4] It is, however, difficult to evaluate the effects of bitterns quantitatively because of the small amount of bitterns. Previous studies have shown that the taste sensor can quantify the bitterness elicited by typical bitter substances such as quinine.[5,6,7] As the result of a previous study using the taste sensor,[8] we showed that the taste of salt, which is affected by Ca^{2+}, was correlated to the ionic strength and the results of principal component analysis on the basis of sensor outputs. However, the effect of Mg^{2+} on the taste was not studied owing to the lack of sufficient sensor output information.

In addition to the usual potentiometric measurements carried out in the previous study, therefore, a CPA (Change of membrane Potential caused by Adsorption) measurement which corresponds to the aftertaste of human beings[9] was introduced in the present study. It is very effective for quantifying taste substances such as quinine and monosodium glutamate which were adsorbed onto the electrode when measuring the sample solution. The amount of adsorbed chemical substances on the membranes can be evaluated by the CPA measurement. In this study we proposed the evaluation model for saltiness based on the sensor output and sensory evaluation. In the method, we tried to coordinate the sensor output and sensory evaluation in order to quantify the human gustatory sensation. Application to salts on the market was performed using the evaluation model in order to qualify the method. We measured twelve kinds of salts and evaluated the saltiness by means of the method.

2. Materials and Methods

2.1 *Measurements using taste sensor*

The schematic diagram of the taste-sensing system SA402 (Anritsu Corp.) is shown in Fig. 1. The detecting part of the sensor usually consists of eight electrodes with different lipid/polymer membranes. The sensor detects taste information which is transformed into electric signals of membrane potentials. According to the taste substances to be measured, different lipid/polymer membranes are available. Lipids used for preparing membranes in this study are listed in Table 1. Lipid/polymer membranes from channels 1 to 3 are negatively charged, and those from channels 4 and 5 are positively charged. Each lipid was mixed with polyvinyl chloride (PVC) and a plasticizer (DOPP: dioctyl phenylphosphonate

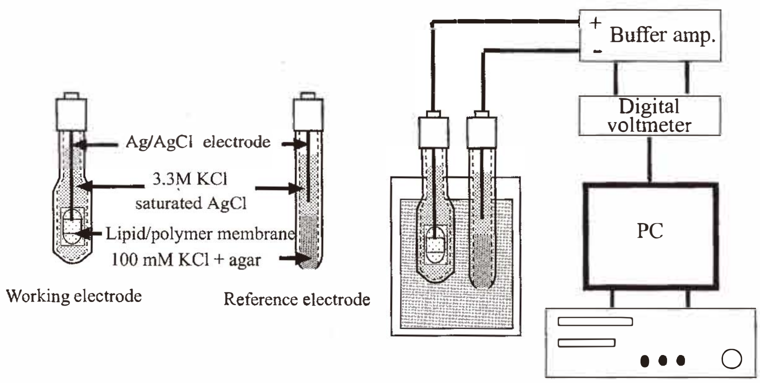

Fig. 1. Schematic diagram of experimental setup.

Table 1
Lipid materials used for the membrane forming process.

Channel	Lipid	Plasticizer
1	Palmitic acid	DOPP
2	Phosphoric acid di-n-decyl ester	DOPP
3	Phosphoric acid di-n-decyl ester	NPOE
4	Tetradodecylammonium bromide	DOPP
5	Tetradodecylammonium bromide	NPOE

or NPOE: 2-nitrophenyl octyl ether) dissolved in tetrahydrofuran, and then dried on a glass plate. The lipid/polymer membrane is a transparent, soft film of approximately 200 μm thickness. The electrode was made of an Ag wire, whose surface was plated with AgCl, in an internal cavity filled with 3.3 M KCl and saturated AgCl solution (Fig. 1). The difference in electric potential between the working and reference electrodes was obtained by means of a high-input impedance amplifier. The electric signal from the sensor was converted to a digital code by a digital voltmeter and was fed to a computer.[2,3]

The CPA measurement provides information that is different from that obtained by the conventional measurement. Figure 2 shows the procedure for CPA measurement. The electric potential of CPA is obtained as (Vr'–Vr), which reflects the adsorbed amounts of chemical substances,[9] while the conventional electric potential response is provided by (Vs–Vr).

2.2 *Sample preparation*

We prepared a model sample which consisted of sodium chloride and trace amounts of bitterns such as $CaSO_4 \cdot 2H_2O$, $MgCl_2 \cdot 6H_2O$, $MgSO_4 \cdot 6H_2O$ and KCl. Table 2 shows the concentrations of added bitterns, where the characters S, M, L are assigned to show the corresponding concentrations based on the component ratio of seawater. $MgCl_2 \cdot 6H_2O$ and $MgSO_4 \cdot 6H_2O$ are mixed in the molar ratio 10:3, which is the same ratio as that for seawater. All of these solutions are mixed with 120 mM NaCl.

Table 3 shows twelve kinds of salts on the market from different manufacturers. In some products, the component ratio is not listed on the label. We converted molar concentration to weight percentage which is equivalent to that of Mg^{2+} : Ca^{2+} = LL with 120 mM NaCl.

3. Results and Discussion

3.1 *Discrimination of a trace of bitterns based on electric response patterns*

Initially, we examined response patterns of bitterns as shown in Fig. 3. All of the experimental data are the differences from the electric potential of 120 mM NaCl. Since chs. 1 to 3 are negatively charged membranes, the electric potential changes positively due to cations such as Mg^{2+} and Ca^{2+}. The electric responses of chs. 1 to 3 increase with the Mg^{2+} and Ca^{2+} concentrations. On the other hand, chs. 4 and 5 are positively charged membranes and hence the electric potential changes negatively due to anions such as Cl^-

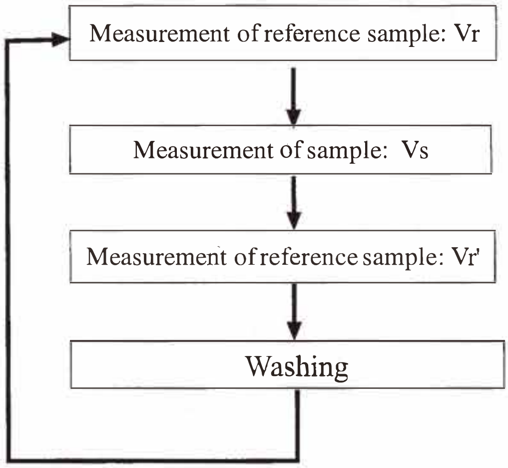

Fig. 2. CPA measurement procedure.

Table 2
Concentration of bitterns examined (mM). The concentration Mg^{2+} is determined by $MgSO_4 \cdot 6H_2O:MgCl_2 \cdot 6H_2O$ =3:10 ratio.

	S	M	L
K^+	0.25	0.5	1
Ca^{2+}	0.25	0.5	1
Mg^{2+}	0.75	1.5	3

Table 3
List of salts on the market.

No.	Brand name	No.	Brand name
1	*Honjio*	7	*Nuchimasu*
2	*Syokuen*	8	*Umikaranoyuki*
3	*Hakatanoshio*	9	*Okinawanoshio*
4	*Akounoamajio*	10	*Adannoyume*
5	*Aranami*	11	*Sankaien*
6	*Itoman-irien*	12	*Shizentenpien*

and SO_4^{2-}. It is noted that the response pattern of K^+ with 120 mM NaCl is almost negligible compared with the results of Mg^{2+} and Ca^{2+}. This implies K^+ has little or no effect on saltiness. According to sensory evaluation,[(4)] K^+ has little effect on saltiness either. We therefore investigated the relationship between saltiness and bitterns without K^+ in the following.

We next examined the mixtures of Mg^{2+} : Ca^{2+} with 120 mM NaCl. There are nine combinations with respect to the corresponding concentrations such as LL, LM, ..., SS as listed in Table 2. Figure 4 shows the results of the sensor outputs, which corresponded to the differences from the electric potential of 120 mM NaCl. Each electric response of chs. 1, 2 and 3 in Fig. 4 shows a clear dependence on the Ca^{2+} concentration. Definite groupings are found for L, M and S of Ca^{2+} in chs. 1–3, i.e., top group, LL, ML, SL; middle group,

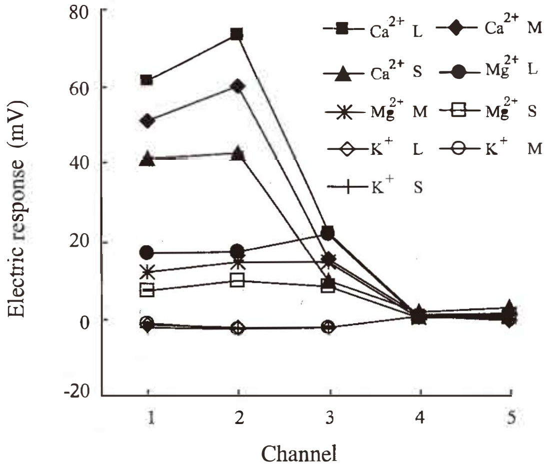

Fig. 3. Electric response patterns for bitterns with 120 mM NaCl.

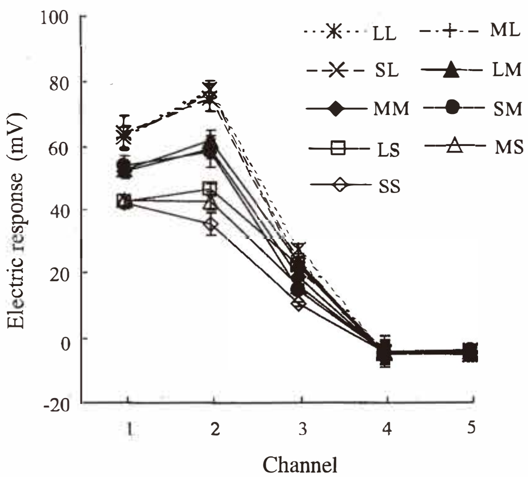

Fig. 4. Response patterns for the mixture of Mg^{2+} and Ca^{2+} with 120 mM NaCl.

LM, MM, SM; bottom group, LS, MS, SS.

Figure 5 shows the response patterns of CPA measurement. The CPA responses of chs. 1 and 2 also depend systematically on the Ca^{2+} concentration, as for the electric responses in Fig. 4. However, ch. 3 shows a systematic dependence on the Mg^{2+} concentration in Fig. 5. The CPA values of the top group are shown by LL, LM and LS; those of the middle group by ML, MM and MS and those of the bottom group by SL, SM and SS. Based on these results, we developed a method for measuring saltiness quantitatively using principal component analysis (PCA).

3.2 *Data analysis based on PCA and sensory evaluation*

As stated in the previous section, we measured model samples and obtained standard data on sensor outputs. We next developed the evaluation model of saltiness based on the results, employing the PCA technique. In Fig. 6 the horizontal axis shows PC1 which shows the analytical results of electric responses in Fig. 4, while the vertical axis shows the results of CPA measurement shown in Fig. 5. The abbreviations LL, LM, ..., SS indicate the corresponding concentrations of Mg^{2+} and Ca^{2+}. In the sensory evaluation report,[4] seven items including salty, aftertaste and bitter were adopted in order to describe taste change with bitterns. Although only the model samples denoted by double circles could be detected by human sensory tests, we could successfully correlate saltiness change and sensor outputs because the measurement errors are very small as shown in Figs. 4 and 5. Model samples, for which panels detected the change of taste from 120 mM NaCl,[4] are denoted by double circles in Fig. 6.

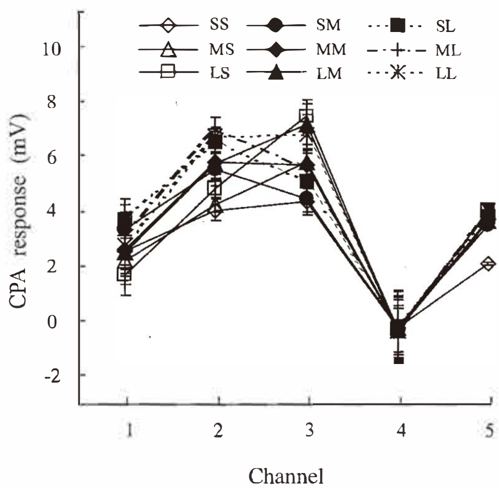

Fig. 5. CPA response patterns for the mixture of Mg^{2+} and Ca^{2+} with 120 mM NaCl.

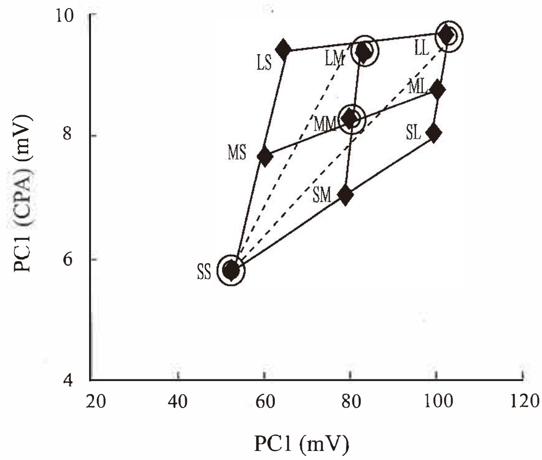

Fig. 6. Evaluation of saltiness on the basis of sensor output and sensory evaluation.

As shown in Fig. 6, the assigned letters such as LS, MS, SS along the longitudinal direction indicate Mg^{2+} concentration dependency, while SL, SM, SS along the lateral direction indicate that of Ca^{2+}. The area bounded by dotted lines in Fig. 6 would indicate the affect on the salty taste. It is noteworthy that saltiness depends not only on the concentration but also on the molar ratio of Mg^{2+} and Ca^{2+} as shown in Fig. 6. The plot of 120 mM NaCl should be the origin of the coordinate axes because all of the experimental data were obtained as the difference from the electric potential of 120 mM NaCl.

3.3 *Evaluation of salts on the market*

In this section, we report the evaluation of salts on the market using the method described above. Figure 7(a) shows the electric responses of twelve kinds of salts from various manufactures. Figure 7(b) shows the response patterns of CPA measurement. All of the results were the difference from the electric potential of 120 mM NaCl. The responses to sample no. 2 are lower than those to other salts in both Figs. 7(a) and (b) because it consists of more than 99% pure sodium chloride. Numbers 7 and 8 show the highest responses in Figs. 7(a) and (b). This is reasonable since they contain the highest amounts of bitterns.

We obtained the PCA plot as shown in Fig. 8 on the basis of the results in Figs. 6 and 7. According to the results of Fig. 7, sample no. 2 is very close to the origin. This indicates that sample no. 2 tastes like pure salt; sodium chloride. On the other hand, sample no. 7 which is denoted around an extension of the dotted line contains the highest amounts of Mg^{2+} and Ca^{2+}, and hence is farthest away from the origin. However, it is apparently difficult to express the change of taste using only five basic taste qualities because the

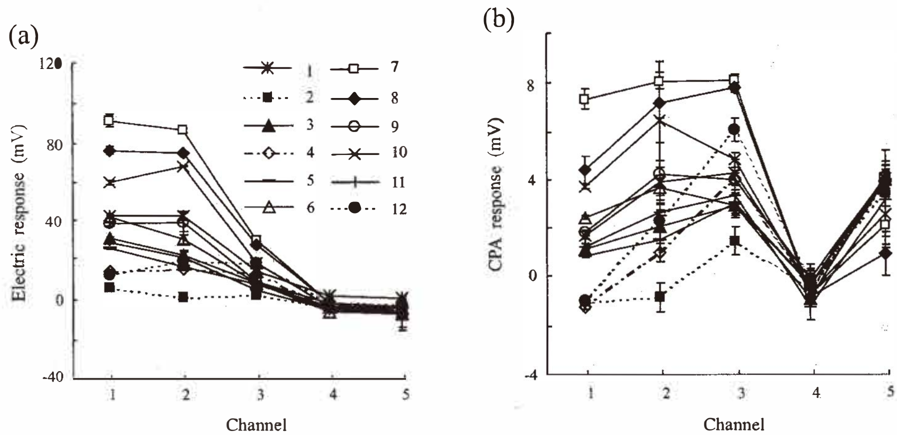

Fig. 7. Electric response (a) and CPA response (b) patterns of the salts on the market.

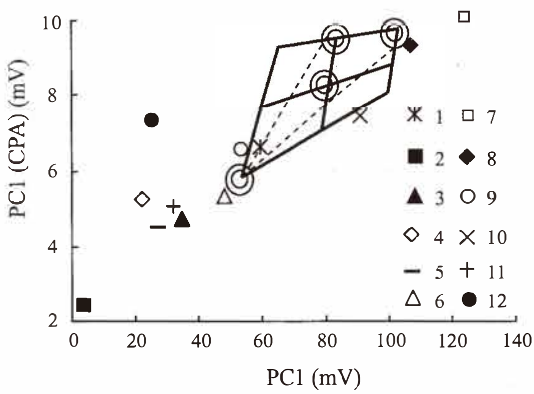

Fig. 8. Evaluation of saltiness based on the sensor output and sensory evaluation.

amount of bitterns is quite small compared to that of sodium chloride, and is almost at the limit of perceptible taste.

4. Conclusions

We examined the correlation between salt with bitterns and saltiness using a multichannel taste sensor. We obtained a taste map constructed from a Ca^{2+} dominant axis and a Mg^{2+} dominant axis. Based on the sensor outputs and sensory evaluation for model samples, we developed a quantitative measurement method for saltiness which contains a trace of bitterns. The method enabled us to evaluate small quantities of bitterns sufficiently and accurately. Application of the method to the salts on the market verified its reliability. This method has significant advantages over human sensory evaluations, since it can evaluate taste quantitatively and also predict the taste of unknown samples.

Acknowledgement

This study was partly funded by the Salt Science Research Foundation.

References

1 K. Toko: Biomimetic Sensor Technology (Cambridge University Press, Cambridge, 2000).
2 K. Toko: Meas. Sci. Technol. **9** (1998) 1919.
3 K. Toko: Sens. Actuators B**64** (2000) 205.
4 N. Matsumoto: The Salt Science Research Foundation Annual Research Report 1991, Physiology Food Science (1991) 61 (in Japanese).
5 S. Takagi and K. Toko: Digest Tech. Papers Transducers'99 (1999) 1638.
6 S. Arita, R. Takamatsu, S. Takagi and K. Toko: Tech. Dig. 18th Sens. Sympo. (2001) 413.
7 S. Takagi, K. Toko, K. Wada and T. Ohki: J. Pharm. Sci. **90** (2001) 2042.
8 M. Habara and K. Toko: IEICE Trans. Electronics E83–C (2000) 1040.
9 H. Ikezaki, A. Taniguchi and K. Toko: Trans. IEE of Japan **118**–E (1998) 506 (in Japanese).