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Development of a Flow-Type Taste Sensor

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The purpose of this study is to develop a taste sensor for real-time control of food quality. The main requirement for the taste sensor used for real-time quality control in food production is that it can measure a small amount of the sample, which is extracted automatically. It is considered that a flow-type sensor is more effective than a conventional taste sensor, which is composed of several kinds of lipid/polymer membranes for transforming the information of taste substances into electrical signals. The measurement system was built by combining solenoid valves, a peristaltic pump, some connectors and a computer. Some measurement conditions such as stabilization time and the amount of samples were examined. The flow-type sensing system showed responses to taste substances in a similar way to the conventional batch-type sensing system; i.e., discrimination of taste was successful. Consequently, the ethanol concentration was measured with less than 0.5% error for several brands of sake with different ethanol concentrations. These results suggest that the flow-type taste sensing system is useful for automatic quality control in the fermentation process.

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1. Introduction

Among the five senses of humans, the senses of sight, hearing and touch are physical senses produced by receiving physical quantities; it is relatively easy to detect or reproduce the function of these senses. On the other hand, the senses of taste and smell are chemical senses which function through the reception of a variety of chemical substances. It has been difficult to reproduce these two chemical senses, because the first three senses treat a type of physical quantity such as light, sound or pressure, but the last two senses treat many kinds of chemical substances. In biological systems, these chemical substances are classified into various groups dependent on their taste. Many kinds of chemical substances form one characteristic smell, *e.g.*, of apple, in the olfactory sense. Therefore, it is important to detect them in a way similar to the gustatory or olfactory system.

Several kinds of taste and odor sensors have been developed so far. An odor sensor using a quartz crystal microbalance coated with polymers can discriminate chemical substances with an accuracy as high as that of humans with the aid of neural network;(1) a gas-sensitive field-effect device (electronic nose) is also effective for odor discrimination.(2) A taste sensor (i.e., electronic tongue) using a surface plasmon sensor is studied for control of a fermentation process.(3) A taste sensor using an surface photo-voltage method can discriminate minute differences in the tastes of sweet substances.(4)

A taste sensor developed by us utilizes different kinds of lipid/polymer membranes.(5–8) The lipid/polymer membranes have electric responses to chemical substances depending on their tastes; i.e., the membranes can classify their tastes as five kinds of tastes: saltiness, sourness, bitterness, sweetness and umami. Saltiness is produced by ions such as sodium, sourness by protons, bitterness by quinine, caffeine and many hydrophobic substances, sweetness by sucrose and artificial sweeteners, and umami is the fifth basic taste produced by monosodium glutamate (MSG), disodium inosinate (IMP) and disodium guanylate (GMP). A taste sensor which utilizes the lipid/polymer membranes has been developed by Intelligent Sensor Technology (Insent, Inc.) and has been made commercially available. It is now used in many food and pharmaceutical companies for the purpose of quality evaluation and control of food or medicines.

This system adopts batch-type measurement. If the flow-type measurement system can be constructed, the amount of sample used can be much smaller. This improvement would also enable us to perform automated measurement.

The purpose of the present study is to construct a taste-sensing system for quality control in the fermentation process. We constructed a flow-type sensing system and measured five kinds of taste substances. It was applied for the measurement of ethanol concentration, which is in fact required for the quality control of fermented beverages such as Japanese sake, wine and beer.

2. Materials and Method

2.1 *Lipid/polymer membranes*

The choice of the membranes depends on the samples to be measured. The eight kinds of membranes shown in Table 1 have been used so far for the measurement of food-

Table 1
Lipid components of the sensor membranes for five basic taste substances.

Channel	Lipid component	Channel	Lipid component
DA	Decyl alcohol	5:5	DOP:TOMA=5:5
OA	Oleic acid	3:7	DOP:TOMA=3:7
DOP	Dioctyl phosphate	TOMA	Trioctyl methyl ammonium chloride
9:1	DOP:TOMA=9:1	OAm	Oleyl amine

stuffs.(5,6) In the present study, therefore, these membranes were also used. Dioctyl phenyl phosphonate was used as the plasticizer.

On the other hand, the two kinds of membranes listed in Table 2 were used because these membranes are more hydrophobic than the ones listed in Table 1, and hence they are more suitable for the measurement of alcohol. All these membranes were fabricated by mixing polyvinylchloride, lipids and plasticizer, as reported in previous papers.(5–8)

2.2 *Flow-type sensor cell*

Figure 1 shows a flow-type sensing system. Eight electrodes made of acrylic tubes with a diameter of 3 mm were set into the PVC plate of 3 mm width. The lipid/polymer membranes were pasted on the tip of each electrode using an adhesion bond, 1% PVC tetrahydrofuran solution. The inner solution of the electrode was 3.3 M KCl solution with saturated AgCl, which contains a Ag/AgCl wire.

A silicone-rubber gasket was inserted between the above PVC plate with the electrodes and a 5-mm-wide PVC plate. The measured sample solution flowed in the flow route 3 mm × 30 mm, which was formed in the gasket.

2.3 *Flow-type measuring system*

Figure 2 shows a block diagram of the measuring system. A peristaltic pump was set at the lower position in order to replace the previous solution with a new solution quickly and thoroughly. The flow rate was controlled by switching the solenoid valve using a signal from the computer. A teflon tube of 3 mm diameter was used for the main flow route.

2.4 *Measured samples*

The measured taste substances are as follows: glycine and L-alanine as sweet substances, NaCl and KCl as salty substances, HCl and citric acid as sour substances, quinine-HCl and L-tryptophan as bitter substances, and MSG and IMP as umami taste substances. Measured samples of ethanol are listed in Table 3. Samples of sake with different ethanol concentrations were prepared by diluting one sample of sake on the market containing 16.0% ethanol with distilled water: 16.0, 13.3, 10.7, 8.0 and 5.3%.

2.5 *Measurements procedure*

In the case of the measurement of basic taste substances, 50 mM KCl solution was used

Table 2
Lipid components of the sensor membranes for ethanol.

Channel	Lipid component	Plasticizer
A	Tetradodecylammoniumbromid	NPOE + dioctyl pheny phosphonate
B	Tetradodecylammoniumbromid	DA + dioctyl pheny phosphonate

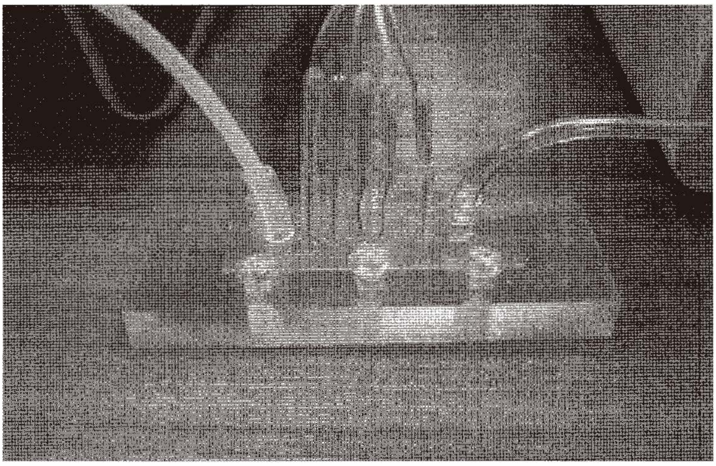


Fig. 1. Flow-type sensing system.

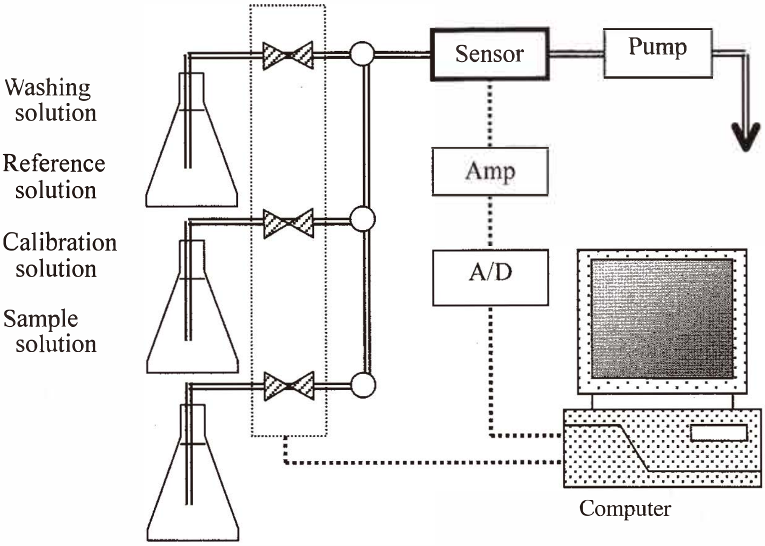


Fig. 2. Block-diagram of measuring system.

Table 3
Measured samples of ethanol and sake.

EtOH / KCl					
5%/0 mM	5%/10 mM	5%/100 mM,	10%/0 mM	10%/10 mM	10%/100 mM,
15%/0 mM	15%/10 mM	15%/100 mM,	20%/0 mM	20%/10 mM	20%/100 mM

as the standard solution and washing solution, as usual.(9) The response potential Vs is defined as the difference of the electric potential for the standard solution (V_0) and that for the sample (V): $Vs=V-V_0$.

Figure 3(a) shows the measurement procedure for basic taste substances. The electrodes are stabilized in the standard solution, and then the standard electric potential is measured. Then, the sample solution flow is introduced into the measuring cell to obtain the response potential, and the electrodes are washed in the washing solution. This process is one rotation procedure for measuring one sample. The same procedure is carried out for the following sample.

Figure 3(b) shows the measurement procedure for ethanol. We used two equations for response potentials, which show linear relationships with ethanol and chloride concentrations, for estimating the ethanol concentration. The values of six coefficients in the equations were determined using the solutions for calibration, as shown in Fig. 4. Three kinds of calibration solutions which contain KCl and ethanol were used.

3. Results and Discussion

3.1 *Measurement of KCl*

KCl solutions of three different concentrations (10 mM, 100 mM, 1 M) were measured using two kinds of membranes made of DOP and TOMA, which are typical lipid/polymer membranes negatively and positively charged, respectively. The stability of the response potential was compared between two methods: one involves measuring the response potential of a flowing sample and the other is for nonflowing samples.

The stability was found to be better for the nonflowing sample than for the flowing sample. The sample of over 0.1 ml, which amounts to the sample volume, was made to flow into and out of the cell. It was found from the sensor response that the KCl solution over 3 ml is sufficient for the complete replacement of the previous sample. This implies that an amount of sample as little as 3 ml is required for measurements, whereas 30–50 ml of the sample was required in the conventional batch-type taste sensor.

Figure 4 shows electric responses of the TOMA membrane to KCl with different concentrations. The same value of response is obtained in the repeated measurements.

3.2 *Response characteristics for basic taste substances*

Figures 5(a) and 5(b) show response patterns for saltiness and sourness, respectively. The response potentials of membranes made of DA, OA and DOP increase, while those of

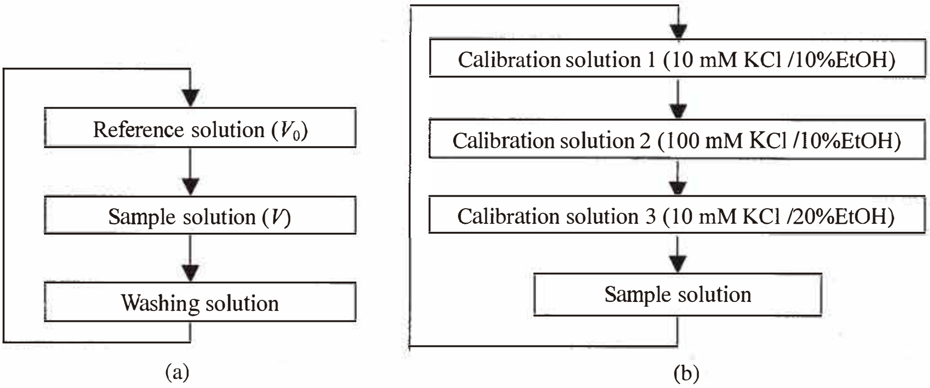


Fig. 3. Measurement procedures for five basic tastes (a) and ethanol (b).

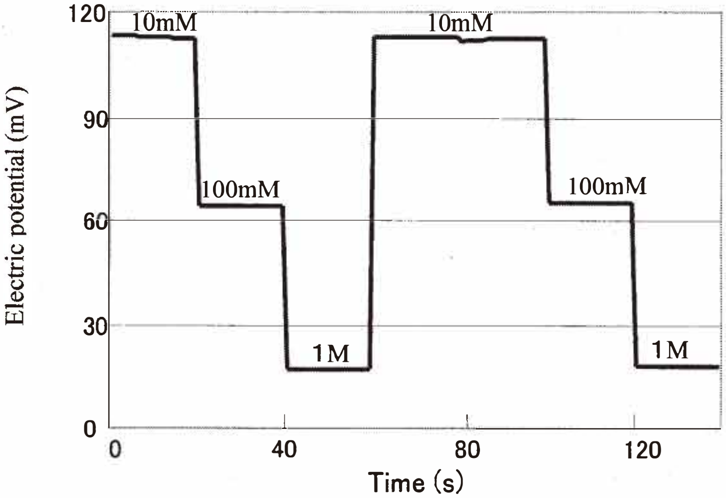
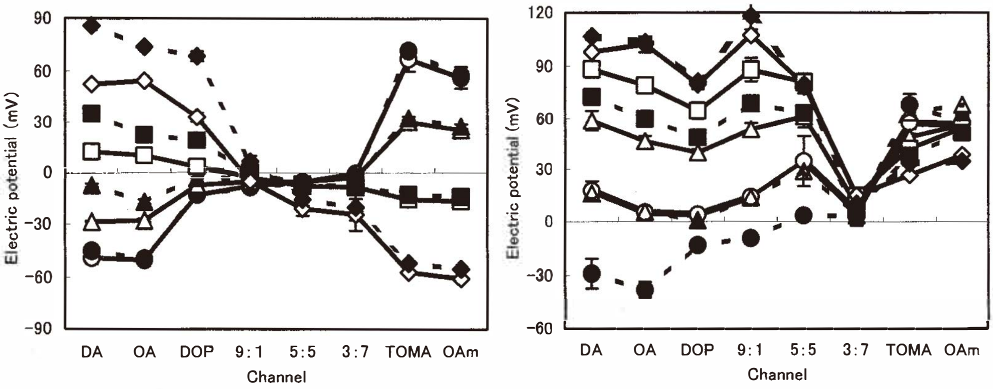


Fig. 4. Response of the TOMA membrane to KCl.

3:7, TOMA and OAm, decrease with increasing concentrations of KCl and NaCl. The 9:1 and 5:5 membranes showed an increase in the response potential in addition to the above change in the case of sour tastes such as HCl and citric acid.

It is noticeable that the response potentials are similar for the same kind of taste, while they are different for different taste qualities; i. e., the patterns for NaCl are similar to that for KCl, but are much different for those for HCl and citric acid. The response magnitudes are a little smaller than those for the conventional batch-type taste sensor,(5–7) but the same characteristics are obtained.



(a) (b)

Fig. 5. Response potential of the taste sensor for KCl and NaCl (a) and HCl and Citric acid (b). (a) ○ 1 mM, △ 10 mM, □ 100 mM, ◇ 1 M for KCl and ● 1 mM, ▲ 10 mM, ■ 100 mM, ◆ 1 M for NaCl. (b) ○ 0.1 mM, △ 1 mM, □ 10 mM, ◇ 100 mM for citric acid and ● 0.01 mM, ▲ 0.1 mM, ■ 1 mM, ◆ 10 mM for HCl.

Figure 6 shows responses to five basic tastes (saltiness, sourness, bitterness, sweetness, umami taste). We can see that the response patterns are different for different tastes, while they are similar for two kinds of chemical substances having the same taste.

3.3 *Estimate of ethanol concentration*

Two kinds of lipid/membranes listed in Table 2 were used for estimating the ethanol concentration. The responses of these two membranes were proportional to chloride and ethanol concentrations. This result agrees with those obtained previously.(10,11) The change in electric potential due to ethanol concentration was suggested to be brought about by a change in the activity coefficient of chloride ions due to ethanol.

On the basis of this suggestion, the following equations hold for the response potentials E_1 and E_2:

$$E_1 = S_{11}\log[\mathrm{Cl}^-] + S_{12}[\mathrm{EtOH}] + E_{01},$$

$$E_2 = S_{21}\log[\mathrm{Cl}^-] + S_{22}[\mathrm{EtOH}] + E_{02}, \tag{1}$$

where $[\mathrm{Cl}^-]$ and [EtOH] are the concentrations of chloride anions and ethanol, respectively, and the six coefficients S_{11}, S_{12}, E_{01}, S_{21}, S_{22} and E_{02} are determined using the calibration solution in Fig. 3(b).

Figure 7 shows the estimated value for ethanol in the artificial alcohol solution listed in Table 3. The difference is smaller than 0.5% when KCl is added, whereas it reaches about 2% without KCl addition. It becomes smallest when 100 mM KCl is added, irrespective of

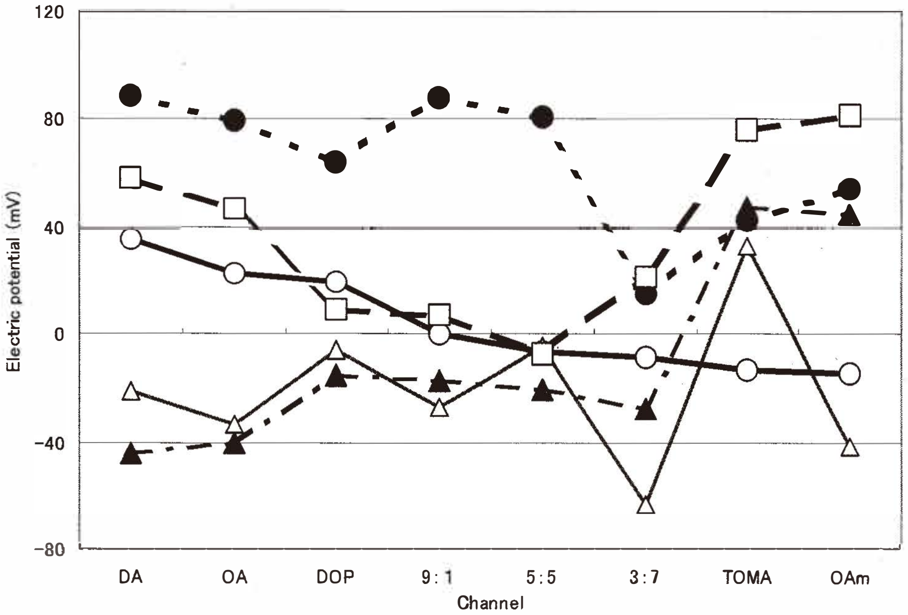


Fig. 6. Response potential of the taste sensor for five basic taste substances.
○ 100 mM NaCl, ● 10 mM tric acid, △ 10 mM IMP, ▲ 30 mM L-alanine, □ 0.1 mM quinine.

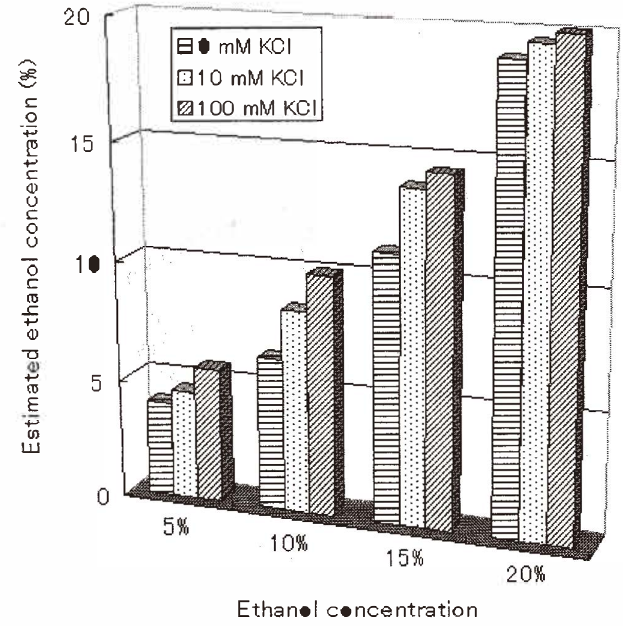


Fig. 7. Estimated ethanol concentration of sample solution.

the ethanol concentration.

Figure 8 shows the difference between the ethanol concentration and the estimated value of sake. The errors lie between 0.05% and 0.6%.

A flow-type measurement system was built by combining solenoid valves, a peristaltic pump, some connectors and a computer. Measurement conditions such as stabilization time and the amount of samples were examined. The flow-type sensing system showed electric responses to taste substances in a similar manner to the conventional batch-type sensor; i.e., discrimination of taste was successful. The ethanol concentration was measured with an error of less than about 0.5% for several samples of sake with different ethanol concentrations.

The conventional taste sensor can measure after-taste using a CPA method.(8,12,13) The term CPA means "Change in membrane Potential due to Adsorption of chemical substances onto the membrane." This CPA measurement makes it possible to quantify a bitter taste or astringent taste, because these tastes originate in the strong binding of chemical substances with artificial lipid membranes of the taste sensor. A similar phenomenon occurs in the human tongue to result in after-taste. This CPA method can also be adopted in the flow-type taste sensor. However, the rinsing process is different between the flow-type and the conventional ones. This might lead to a slight difference of CPA value obtained by this measurement. Construction of a calibration curve enables us to get the after-taste, which is the sense felt by humans.

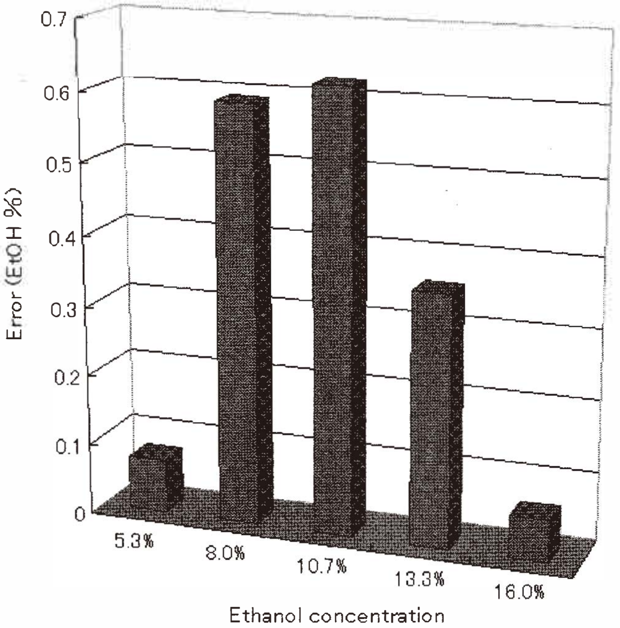


Fig. 8. Measurement error of the ethanol concentration of sake.

At present, the ethanol concentration is determined using a buoy with an accuracy of 0.1%. However, this measurement equipment is not compact or handy, and it takes about one hour to obtain the result. Therefore, the method proposed here is desirable from the viewpoints of its portability and rapidness; however, the accuracy must be improved for real use, for which the error below 0.1% must be attained, although this depends on the purpose and target. This point is left as a future task. Of course, the flow-type sensing system can be used for other targets including a wide range of foodstuffs.

References

1 T. Yamanaka, R. Matsumoto and T. Nakamoto: Sensors and Actuators B **89** (2003) 112.
2 C-F. Mandenius, A. Hagman, F. Dunås, H. Sundgren and I. Lundström: Biosens. Bioelectron. **13** (1998) 193.
3 H. Nanto, Y. Hamaguchi, M. Komura, Y. Takayama, T. Kobayashi, Y. Sekikawa, T. Miyatake, E. Kusano, T. Oyabu and A. Kinbara: Sensors and Materials **14** (2002) 1.
4 T. Katsube, Y. Sasaki and H. Uchida: Tech. Digest 13th Sensor Symposium (1995) 49.
5 K. Hayashi, M. Yamanaka, K. Toko and K. Yamafuji: Sensors and Actuators B **2** (1990) 205.
6 K. Toko: Biominetic Sensor Technology (Cambridge University Press, Cambridge, 2000) p. 113.
7 K. Toko: Sensors and Actuators B **64** (2000) 205.
8 Y. Miyanaga, Y. Kobayashi, H. Ikezaki, A. Taniguchi and T.Uchida: Sensors and Materials **14** (2002) 455.
9 S. Takagi, K. Toko, K. Wada and T. Ohki: J. Pharm. Sci. **90** (2001) 2042.
10 Y. Arikawa, K. Toko, H. Ikezaki, Y. Shinha,T. Ito and I. Oguri: J. Ferment. Bioeng. **82** (1996) 371.
11 T. Tsukatani and K. Toko: Food Sci. Technol. Res. **5** (1999) 43.
12 H. Ikezaki, A. Taniguchi and K. Toko: Trans. IEE Japan, 117-E. **9** (1997) 465.