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Color control of Japanese soy sauce (*shoyu*) using membrane technology

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ABSTRACT

The present study systemically decolorized soy sauce using a membrane process to analyze the separation mechanism. An ultrafiltration (UF) membrane (NTU-2120) exhibited only slight decolorization ability. A nanofiltration (NF) membrane with a lower molecular weight cut-off and produced by sulfonated polysulfone (NTR-7400 series) rather than polyvinyl alcohol/polyamide (NTR-7250) had higher decolorization ability. The NF membranes rejected total nitrogen by 17–24%, unsalted soluble solid content by 24–32%, reducing sugar by 25–43%, and amino acids by 10–25%. The NTR-7400 series membrane rejected lactic acid by 6–9%, and pyroglutamic acid by 11–21%; other quality indexes were maintained. In the NF membrane processes, higher rejection of acidic amino acids than neutral and base amino acids was observed. The separation performance was governed by the electrical effect as well as the sieve effect. Soy sauce color could be controlled by blending NF membrane-processed soy sauce with feed soy sauce. Color can be matched to preference in accordance with dishes by suitably blending NF membrane-processed soy sauce with feed soy sauce.

Keywords: Japanese soy sauce (*shoyu*); Membrane process; Color; Separation mechanism; Blending

1. Introduction

Developing food to accommodate consumer preference is very important for the development of gastronomic culture as well as production of vendible commodities. Color is a major factor in our acceptance of food, and many studies have confirmed that when the color of food does not match our expectations, our acceptance of that food is difficult (Frick, 2003).

Japanese soy sauce (*shoyu*) is a traditional seasoning of Japanese food and is currently used in cooking worldwide (Kobayashi, 2010). The main type of soy sauce, *Koikuchi-shoyu* (common soy sauce), is an all-purpose seasoning characterized by a pleasant aroma, a strong flavor, and a deep reddish-brown color. It contains many umami taste components and thus is associated with palatable and pleasurable taste (Lioe et al., 2010).

Soy sauce production involves vigorous lactic and alcohol fermentation. The finished product is pasteurized at a rather high temperature (80 °C). Half of the color is formed during the fermentation and aging of mash, and half is formed during pasteurization. Both are due primarily to heat-dependent browning, known as the Maillard browning reaction, between amino compounds and sugars (Yokotsuka, 1986). At the same time, various constituents that determine the soy sauce quality are formed during the fermentation based on the ingredients including soy bean, wheat and sodium chloride (NaCl). Good-quality soy sauce (*Koikuchi-shoyu*) contains 1.3–1.8% (grams per volume) total nitrogen (TN), 1.5–5% reducing sugar (mainly glucose; RS), 1.5–2.5% ethanol, 1–1.5% polyalcohol (primarily glycerol), 1–2% organic acid, and 16–18% NaCl. In order for a soy sauce to have palatable taste, about half of its nitrogenous compounds must be

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free amino acids; in particular glutamic acid (Glu) is a very important component (Yokotsuka, 1986).

The authors study of general consumers' color preference for *Koikuchi-shoyu* (Miyagi, 2012) indicated that lighter-colored soy sauce was generally preferred over the conventional one, although the color preference varied with the dish. Establishing technology to control the color of soy sauce without changing its other sensory qualities has potential for developing new products for customer acceptance.

Adsorption and/or membrane processes have been proposed as simple decolorization processes of soy sauce. We investigated the systemic adsorption of soy sauce and reported that activated carbon, activated clay, and silica gel exhibited decolorizing ability, but diatomaceous earth and magnesium oxide did not (Miyagi et al., 2013). We also documented that activated carbon indicated the greatest decolorizing ability without changing its sensory qualities (Miyagi et al., 2013). However, disadvantages associated with adsorbent systems include leaching of free-form foreign powders into the feed, leaching of metals into the feed, treatment of used adsorbents, and cost.

In contrast, the membrane process is remarkably simple, offering many advantages over other separation and purification processes without the disadvantages of adsorbent systems. Hashiba (1973) and Sasaki et al. (1993) found it was possible to decolorize soy sauce using an ultrafiltration (UF) membrane. Ikeda et al. (1988) and Suzuki and Mori (1995) attempted to decolorize soy sauce using a nanofiltration (NF) membrane and reported that the NF membrane process had remarkable decolorization ability. Furukawa et al. (2006) developed method of concentrating soy sauce constituents using an NF membrane. Although membrane technology has been practically used for the soy sauce industry, the separation principle has not been sufficiently established. Advances in this technology will require clarifying the membrane separation mechanism and food development in line with consumer preferences using decolorized soy sauce.

The present study systematically investigated membrane separation for decolorization and changes in soy sauce (*Koikuchi-shoyu*) quality (e.g. general nutritional components, amino acids, and organic acids) using various types of membranes and discussed its separation mechanism. In addition, control of soy sauce color was also attempted by blending feed and membrane-processed soy sauces.

2. Experimental

2.1. Materials

Soy sauce (*Koikuchi-shoyu*, *Jyokyu* (superior) grade) was purchased from a certain corporation (Chiba, Japan). Five membrane types (NTU-2120, NTR-7250, NTR-7410, NTR-7430, and NTR-7450) were purchased from Nitto Denko, Ltd. (Shiga, Japan). The NTU-2120 membrane is classified as a UF membrane, and other membranes are classified as NF membranes. The materials and the approximate molecular weight cut-off (MWCO) values are listed in Table 1.

2.2. Membrane experiment

An apparatus with a magnetically stirred membrane cell (Model C-70B; Nitto Denko, Ltd.) used in the experiment was the same as that used in the previous report (Miyagi et al., 2011). In all the experiment runs, the temperature was 25 °C, the operating pressure was 2 MPa, and the speed of the spin bar was 200 rpm. The initial charge of the feed sample was 100 g. The experiment run was continued until the permeate reached 50 g. Color was evaluated immediately after the samples were obtained. For other evaluations, the samples were poured into shield bottles, purged with nitrogen gas, and stored at 5 °C until measurement.

2.3. Blend experiment

The NTR-7450 membrane had the highest decolorization ability in the membrane experiment, therefore we used the permeate in the blend experiment. Various colors of soy sauce were obtained by blending the NTR-7450 processed soy sauce with the feed. The feed/permeate blend ratios prepared were 4:1, 3:2, 2:3, 1:4, 1:9, and 1:19 (v/v). The color was determined immediately after blending.

2.4. Analyses

To estimate color, we determined the absorbance at 420 nm and the $L^*a^*b^*$ (CIELAB) color space in a 10 mm cuvette. The absorbance was recorded using a spectrophotometer (Model U-1500; Hitachi, Ltd., Tokyo, Japan), and the CIELAB color space was determined using a colorimeter (Model CT-210; Minolta, Ltd., Osaka, Japan). When the absorbance exceeded 0.8, we measured the value after suitable dilution with pure water to be within 0.2–0.8, and recalculated according to the *Beer–Lambert* law.

As quality analyses, TN was determined by the *Kjeldahl* method, and NaCl was determined by the *Mohr* method. The unsalted soluble solid content (USSC) was obtained by subtracting the NaCl value from the reading of a sugar refractometer. Ethanol content was determined by gas chromatography with FID. RS was determined using the *Fehling–Lehmann–School* method. The pH was determined using the glass-electrode method. Amino acids were determined using an automatic amino acid analyzer (HPLC). Organic acids were determined using a bromothymol blue (BTB) post-column HPLC with a UV–vis detector. All analyses were the same as in our previous report (Miyagi et al., 2013).

2.5. Calculation and data analysis

The performance of the membrane was expressed in terms of rejection (*R*) calculated as

$$R = 1 - \left(\frac{C_P}{C_F}\right) \quad (1)$$

where C_F is the content of each component in the feed soy sauce and C_P is the content of each component in the processed soy sauce. The rejection of absorbance was also determined.

The blend ratio of the NTR-7450 membrane-processed soy sauce (BRP) was calculated as

$$\mathrm{BRP} = \frac{\mathrm{VPS}}{\mathrm{VPS} + \mathrm{VFS}} \quad (2)$$

where VPS is the volume of the membrane-processed soy sauce and VFS is the volume of feed soy sauce.

All analyses except for the absorbance at 420 nm and pH were repeated three times, and the standard deviation was