Technical paper

Differential Recovery of Terpene Hydrocarbons and Oxygenated Compounds from Condensates Containing Essential Oil Discharged during Concentration of Citrus Juices Using a Ceramic Membrane

Koji Sakamoto,[1] Kazuyoshi Fujii,[1] Atsuhiko Inoue,[1] Hiroshi Kozuka[2] and Hideaki Ohta[3]

[1]*Prefectural Food Technology Research Center, 12-70 Hijiyamahon-machi, Minami-ku, Hiroshima 732-0816, Japan*
[2]*Hiroshima Bunkyo Women's College, 1-2-1 Kabehigashi, Asakita-ku, Hiroshima 731-0295, Japan*
[3]*Department of Food and Nutrition, Nakamura Gakuen University, 5-7-1 Befu, Jonan-ku, Fukuoka 814-0198, Japan*

Essential oils of citrus fruits have many uses in consumer products so that efficient methods for their isolation are needed. Membranes are often used in the separation process but a common problem is that the flux through the membranes decreases with time. In the present study we examined whether ceramic membranes might be better at maintaining the flux at acceptable levels. To test the membranes, we attempted differential recovery of nonpolar terpene hydrocarbons and aqueous oxygenated compounds from condensates produced as a by-product during the concentration of citrus juices. When zirconia membranes (ZrO$_2$-UF, cut-off molecular weight 50,000 and ZrO$_2$-MF, pore size 0.08 μm) were used, little decrease in the flux was observed during filtration. The nonpolar terpene hydrocarbons were retained while more polar oxygenated flavor compounds passed through the membrane. The percentages of alcohols, esters, and aldehydes in the permeate increased markedly compared with their percentages before filtration. When the condensate from the concentration process of citrus juice sampled from a juice factory was filtered using ZrO$_2$-UF, the hydrocarbons was concentrated, and a water-soluble essence consisting primarily of oxygenated compounds was obtained in the permeate.

Essential oils of citrus fruits are used widely as flavoring ingredients of soft drinks, and in cosmetics and raw materials of terpene resins. When they are used for flavoring, they are made into terpeneless oils by eliminating terpene group hydrocarbons such as *d*-limonene, because hydrocarbons, which are the predominant components of essential oils, do not contribute much to the flavor and are poorly soluble in water, and because their flavors are liable to deteriorate (Kirchner & Miller, 1952; Owusu-yaw *et al*., 1986; Alexander, 1991). Also, purified limonene is used as a raw material of terpene resins. Essential oils of citrus fruits, which have many uses, are manufactured primarily from by-products of citrus juices (Matthews & Braddock, 1987; Braddock, 1995).

Several by-products produced during the manufacturing process of citrus juices can be used as sources of essential oils. These include cold-pressed oil, which is obtained by squeezing the fruit peels, emulsions of peel oil, obtained by spraying the peels with water, and aqueous- and oil-phase essences, obtained from juice evaporation (Johnson & Vora, 1983; Flores & Segredo, 1996). Oil-soluble and water-soluble components of essential oils contained in these fluids can be recovered by a combination of centrifugation and vacuum distillation (Matthews & Braddock, 1987; Veldhuis *et al*., 1972), by ultrafiltration or reverse osmosis (Matsuura *et al*., 1975; Braddock, 1982, Braddock &

Adams, 1984; Braddock *et al*., 1991) and by the use of an adsorptive resin (Ericson *et al*., 1992). However, with ultrafiltration or reverse osmosis using polymeric membranes such as those made with cellulose acetate, the permeation flux is reduced as filtration progresses. This decrease in the flux of an emulsion of essential oils in water is rapid and prevents continuation of filtration although it can be controlled to an extent by the presence of sugars or pectin (Sakamoto *et al*., 1995). We found that essential oil components could be concentrated without a decrease in the flux by using a ceramic membrane. Of the essential oil components, hydrophobic terpene hydrocarbons such as limonene are present as oil droplets in an oil-water suspension so that the hydrophobic components are more concentrated in the retentate while oxygenated compounds such as linalool are more concentrated in the permeate. Utilizing this phenomenon, we performed continuous differential recovery of terpene hydrocarbons primarily consisting of limonene and oxygenated compounds primarily consisting of linalool from an essential oil-water suspension using a ceramic membrane and an adsorption resin (Porapack Q).

Materials and Methods

Sample A model fluid of the condensate containing the essential oils was prepared by mixing distilled oil obtained by vacuum distillation of a commercial orange oil (KATAYAMA Chemical, Osaka) with distilled water at 0.1% w/w and homoge-

E-mail: sakamoto@shokuhin-kg.pref.hiroshima.jp

nizing the mixture at 20,500 rpm for 1 min using an IKA-Labortechnik (JANKEL & KUNKEL GMBH Co., Germany). The content of oxygenated compounds in the distilled oil was 1.13% by gas chromatographic analysis.

Differential recovery of essential oil by ultrafiltration and microfiltration The model fluid containing 0.1% (w/w) essential oil (18.5 *l*) was filtered until the volume of the concentrate decreased to 0.5 *l*, corresponding to a concentration factor of 37 times. The concentrate and the permeate containing essential oils were continuously applied to a glass column packed with 50 ml of Porapack Q, and the essential oils adsorbed in the column were recovered using diethyl ether. The recovered essential oil was dehydrated with anhydrous sodium sulfate, subjected to a nitrogen flow at 40°C or below to remove diethyl ether, and weighed. From the recovered essential oil, the hydrocarbon fraction was eluted with 200 ml of *n*-pentane, and the oxygenated compound fraction was eluted with 500 ml of diethyl ether using a silica gel column (Kieselgel 60, Merck, 20 g) equilibrated with *n*-pentane. These fractions were dehydrated with anhydrous sodium sulfate, concentrated by removing the solvents at 40°C or below, and their compositions were determined by gas chromatography.

Ceramic membranes and conditions of filtration A zirconia membrane (ZrO$_2$-UF, cut-off molecular weight 50,000 Da, membrane area 73 cm^2, Carbosep®, TECH-SEP) was used for ultrafiltration, and a zirconia membrane (ZrO$_2$-MF, pore size 0.08 μm, membrane area 73 cm^2, Carbosep®, TECH-SEP) and an alumina membrane (Al$_2$O$_3$-MF, pore size 0.20 μm, membrane area 73 cm^2, Membralox®, TOSHIBA CERAMIC) were used for microfiltration. The zirconia membranes were consisted of an inorganic composite membranes with a zirconia-active layer deposited on a carbon support. Cross-flow filtrations were carried out to compare the three types of filtration membranes under the same operating conditions (membrane surface linear velocity, 4.0 m/s, 18°C, and pressures of 0.44, 0.26, and 0.10 Mpa). The unstable suspension was continuously stirred.

The membranes were washed with P3-ultrasil 75 (0.5%, Henkel-Hakusui) for 30 min. and with P3-ultrasil 10 (0.5%) at 55°C for 30 min. The membranes were found to nearly recover their permeability to pure water after washing by this method.

Gas chromatography/mass spectrometry (GC-MS) The recovered essential oil concentrate was analyzed by GC/MS under the following conditions. Apparatus: MAGNUM (Finnigan MAT), Column: Chemical bond type silica capillary column (DB-WAX, 60 m×0.25 mm i.d.×0.25 μm; J&W Scientific), Column temperature: 40°C–230°C (3°C/min), Port temperature: 230°C, Carrier gas: Helium, Linear velocity: 33.1 cm/s, Split ratio: 1 : 30, Ionization mode: EI and CI (Isobutane), Scanning: Full scan mode (26–300 aum/s), Multiplier voltage: 1250 eV, Ion trap manifold temperature: 230°C. The components were identified by using a library search system (MAGNUM Library-Search System: NIST MASS SPECTRA DATABASE 62,235 compounds) and the Kovats Index (KI) and by comparing their mass spectra and KI values with those of standards (Wako Pure Chemical Industries, Tokyo Kasei Kogyo, Aldrich Chemical Co.). The peak area percentage was calculated from the total ion count. The percentages of hydrocarbons and oxygenated compounds were determined by identifying and quantifying them in the concentrate before fractionation by GC-MS.

Results and Discussion

Changes in the permeation flux of a model fluid of the condensate containing essential oil Citrus essential oils consist of a mixture of the nonpolar hydrocarbons, terpenes and sesquiterpenes, and the polar oxygenated compounds, aldehydes, alcohols, ketones, acids and esters, with low molecular weights in the range of 40–300 Da. The water solubility of hydrocarbons is negligible. However, the solubility of oxygenated compounds often reaches levels of 0.05–1.0%. The essential oil droplet size in mandarin juice is in the range 1.7–40 μm, as measured by a Coulter counter (Ifuku & Maeda, 1976). In this experiment, the suspension, immediately after it was prepared, consisted of only water and essential oil, with the droplet size in the range 2.0–60 μm, as measured by microscopic observation. The suspension was unstable.

Figure 1 shows changes in the flux when a model fluid containing 0.1% (w/w) essential oil was filtered through the membranes. When the model fluid was filtered with the ZrO$_2$-UF (cut-off molecular weight 50,000 Da) and ZrO$_2$-MF (pore size 0.08 μm) membranes, there was little decrease in flux as the concentration increased to a factor of 37. When the Al$_2$O$_3$-MF (pore size 0.20 μm) was used, the flux was initially high because of the large pore size, but it gradually decreased. Since the ZrO$_2$ and Al$_2$O$_3$ membranes are hydrophilic, water and many water-soluble molecules permeated through the pores. The water-insoluble oil droplets, consisting mainly of hydrocarbons, acted as particles. They were rejected at the membrane pore and were swept out the end of the module in a concentrated stream. The rapid decline in flux with time is explained by the build-up of a polarized layer of oil emulsion at the membrane surface and also by the blocking of pores by oil droplets (Lee *et al.*, 1984; Lipp *et al.*, 1988). Of the three ceramic membranes examined in this study, both ZrO$_2$-UF and ZrO$_2$-MF allowed filtration to advance efficiently with little formation of the fouling layer by oil droplets or pore blocking. In contrast, in the Al$_2$O$_3$-MF membrane, which has the largest pore size, the flux decreased with time, presumably because oil droplets introduced into the pores contributed to an increase in the filtration resistance and prevented the filtration of other oil droplets, which were primarily hydrocarbons. The details are described
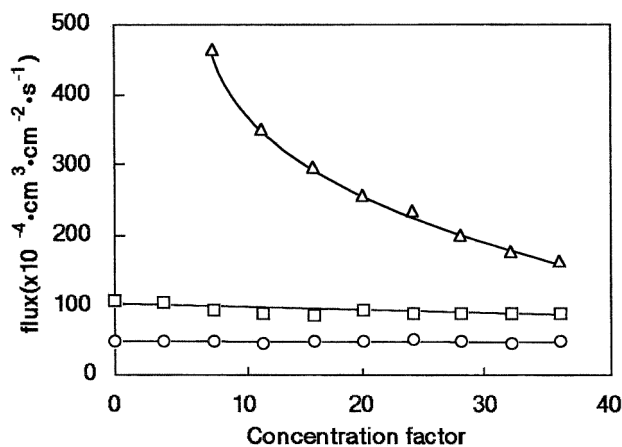

Fig. 1. Permeation flux of ultrafiltration and microfiltration using membranes. Concentration of essential oil: 0.1%. ○, Zr-O$_2$ (molecular weight cut-off: 50,000 daltons); □, Zr-O$_2$ (pore size: 0.08 μm); △, Al-O$_2$ (pore size: 0.2 μm)

later.

Many studies have used hydrophilic polymeric membranes to examine the recovery or concentration of flavors from products, such as apple juice water (Matsuura *et al.*, 1975), orange essence (Braddock *et al.*, 1991), lemon (Kane *et al.*, 1995), apple juice (Chou *et al.*, 1991), mango puree (Olle *et al.*, 1997) and citrus peel oil (Braddock & Adams, 1984). For example, Braddock (1982) recovered limonene, which is a major component of citrus essential oil, using a polysulfone membrane and a cellulose acetate reverse osmosis membrane. With these membranes, the flux decreased with time. Koltuniewicz *et al.* (1995) separated of dodecane from an oil-in-water emulsion using three types of ceramic microfiltration membrane: a Millipore 0.45 μm hydrophilic PVDF membrane, a Gelman 0.1 μm hydrophilic polysulfone membrane, and a Ceramesh 0.1 μm hydrophilic zirconia-coated nickel alloy mesh composite membrane. Microfiltration of the oil-in-water emulsion occurred at a higher flux with the hydrophilic zirconia-coated membrane than with the other membranes. The flux through the hydrophilic PVDF and hydrophilic polysulfone membranes was greatly reduced. Thus, these polymeric membranes are not efficient for the separation of oil-in-water emulsions because of their tendency to easily foul. Braddock *et al.* (1991) and Koltuniewicz *et al.* (1995) showed that the dramatic decline in the flux of polymeric hydrophilic membranes was due to changes in membrane structure. Using ceramic membranes for the separation of essential oils can markedly reduce the effort needed to wash the membrane. The ceramic membrane is clearly superior in this respect.

Percentages of hydrocarbons and oxygenated compounds and their compositions in the permeate The model fluid containing 0.1% (w/w) essential oil was concentrated 37 times using a zirconia membrane and an alumina membrane, and essential oil was recovered continuously by applying both the concentrate and the permeate to a column packed with Porapack Q. Table 1 shows the permeability and ratio of hydrocarbons and oxygenated compounds in the permeate.

The Al_2O_3-MF membrane had the highest permeability to the total essential oil (2.3%), followed by the ZrO_2-MF membrane (1.6%) and ZrO_2-UF membrane (1.1%). However, the ZrO_2-MF membrane had the highest permeability to oxygenated compounds (74.4%), and the permeability of ZrO_2-UF membrane to hydrocarbons was extremely low (0.3%). The results shown in Table 1 suggest that the oil phase, which is composed primarily of nonpolar terpene hydrocarbons, is retained in the retentate and that aqueous oxygenated compounds present primarily in the water phase are able to pass through the membrane. The percentage of hydrocarbons in the permeate was highest with Al_2O_3-MF and lowest with ZrO_2-UF, and more hydrocarbons passed through the membrane when the pore size was larger. The ZrO_2-
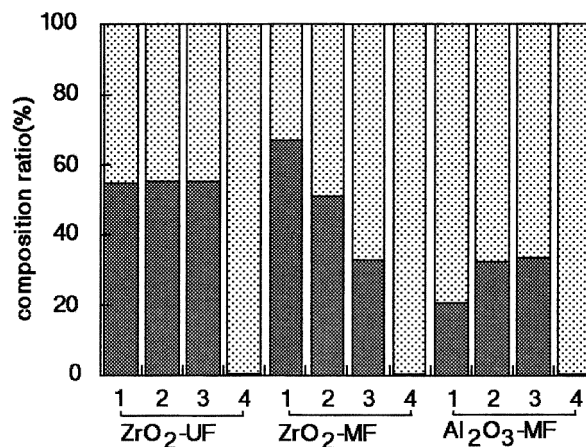

Fig. 2. Quantitative distribution of oxygenated-compounds and hydrocarbons in permeate and concentrate. 1, Permeate (0~6 *l*), concentration factor (×1~13); 2, Permeate (6~12 *l*), concentration factor (×13~25); 3, Permeate (12~18 *l*), concentration factor (×25~37); 4, Concentrate. ▦, Hydrocarbons; ▧, Oxgenated-compounds.

Table 2. Percentages of functional groups of essential oil in permeates.

Functional group	Before filtration (%)	ZrO_2-UF (%)	ZrO_2-MF (%)	Al_2O_3-MF (%)
Aldehydes	0.42	9.93	6.83	3.35
Alcohols	0.39	38.8	29.9	18.8
Esters	0.03	1.41	1.16	1.05
Others	0.05	2.81	2.99	3.69
Hydrocarbons	99.1	44.8	57.5	72.0
Unknown	0.04	2.17	1.63	1.17

MF membrane had the highest permeability to oxygenated compounds, but of the three permeates, the one whose essential oil had the highest percentage of oxygenated compounds (55.2%) was that obtained with the ZrO_2-UF membrane. This percentage is high compared with the initial percentage of oxygenated compounds in essential oil (0.9%). These results indicate that selective differential recovery of essential oil composed primarily of hydrocarbons and essence consisting of oxygenated compounds can be accomplished by filtration through a ceramic membrane.

To determine the percentages of hydrocarbons and oxygenated compounds recovered from each fraction, the permeate was divided into three fractions (initial, middle, late) (Fig. 2). While the percentage of hydrocarbons was stable from the beginning to the end of filtration (44.7–44.9%) through the ZrO_2-UF membrane, it increased from 32.8% to 67.1% during filtration through the ZrO_2-MF membrane. It increased from the initial period to the middle period of filtration through the Al_2O_3-MF membrane but remained fixed thereafter. Oil droplets consisting of hydrocarbons are believed to behave as particles in water. Therefore, the difference in the percentage between hydrocarbons and oxygenated compounds is considered to be due to the differences in the size of oil droplets and the pore size of the membrane. We assume that the ZrO_2-UF membrane scarcely allows the entry of oil droplets into the pores, thus selectively permitting only the components of essential oil present in the water phase to pass, while the ZrO_2-MF membrane with a larger pore size progressively allows the entry of oil droplets into its pores as the fluid is concentrated, permitting part of them to move to the permeate.

Table 1. Permeability and ratio of hydrocarbons and oxygenated compounds in permeates.

Membrane	Permeability(%)		Ratio(%)	
	Hydrocarbons	Oxygenated-compounds	Hydrocarbons	Oxygenated-compounds
ZrO_2-UF	0.3	51.0	44.8	55.2
ZrO_2-MF	0.9	74.4	57.5	42.5
Al_2O_3-MF	1.6	68.6	72.0	28.0

Table 3. Percentages of essential oil compounds in permeates.

Compounds	Before filtration	ZrO$_2$-UF	ZrO$_2$-MF	Al$_2$O$_3$-MF
Aldehydes				
Hexanal	—	0.04	0.02	0.02
Heptanal	—	0.12	0.04	—
Octanal	0.12	3.49	1.92	0.91
Nonanal	0.02	0.22	0.14	0.07
Citronellal	0.03	0.56	0.35	0.11
Decanal	0.15	—	0.26	0.31
(*E*)-2-Decenal	—	0.08	0.02	0.05
Neral	0.03	1.85	1.54	0.69
Dodecanal	0.02	0.06	0.02	0.08
Geranial	0.04	2.48	1.67	0.73
Perill aldehyde	0.02	1.03	0.74	0.36
(*E,E*),2,4-Decadienal	—	—	0.06	—
α-Sinensal	—	—	0.05	—
Alcohols				
2-Butanol	—	0.21	0.16	0.17
3-Pentanol	—	0.09	0.14	0.17
2-Pentanol	—	0.22	0.32	0.39
1-Hexanol	—	0.05	0.04	0.05
(*Z*)-3-Hexenol	—	0.02	0.04	0.04
2-Ethyl-1-hexanol	—	—	0.02	—
Linalool	0.28	24.1	18.8	11.3
1-Octanol	0.03	2.37	1.66	1.39
Terpinen-4-ol	—	0.46	0.35	0.20
p-Mentha-*cis*-2,8-dien-1-ol (T)	0.01	2.52	1.17	0.78
p-Mentha-*trans*-2,8-dien-1-ol (T)	0.01	1.41	0.81	0.65
α-Terpineol	0.03	2.28	1.76	0.90
1-Decanol	0.01	0.58	1.31	0.35
Citronellol	0.01	0.50	0.38	0.20
Nerol	—	0.88	0.52	0.26
trans-3-(10)-Caren-2-ol	0.01	1.74	1.19	1.12
p-Cymen-8-ol	—	0.37	0.30	0.30
1,8-*p*-Menthdien-9-ol	—	0.44	0.33	0.21
Perill alcohol	—	0.09	0.08	0.09
β-Elemol (T)	—	0.48	0.55	0.24
Esters				
Ethyl butanoate	—	0.04	0.02	0.04
Ethyl-2-methyl-butanoate	—	0.04	0.02	0.03
3-Methyl-butyl butanoate	—	0.06	0.05	0.03
Hexyl acetate	—	0.34	0.22	0.42
Hepthyl acetate	—	—	0.02	—
Octyl aceate	0.02	0.09	0.07	0.06
Linalyl acetate	0.01	0.28	0.26	0.23
Nonyl acetate	—	0.06	0.05	—
Citronellyl acetate	0.01	0.24	0.21	0.13
Neryl acetate	—	—	0.03	—
Geranyl acetate	—	0.26	0.20	0.10
Others				
1,8-Cineole	—	0.07	0.13	0.16
cis-Limonene oxide	0.02	0.98	1.23	2.11
trans-Limonene oxide	0.03	1.58	1.51	1.34
trans-Sabinene hydrate	—	0.18	0.13	0.08
d-Carvone	0.04	3.26	2.56	1.81
Hydrocarbons				
α-Pinene	0.48	0.08	0.23	0.24
β-Pinene	0.23	—	—	—
Sabinene	0.42	—	—	—
δ-3-Carene	0.08	—	0.04	0.05
Myrcene	1.74	0.41	0.76	1.11
d-Limonene	95.4	40.5	53.4	68.6
β-Phellandrene	0.22	0.2	0.16	0.04
γ-Terpinene	0.25	—	0.13	0.06
trans-β-Ocimene	0.04	—	0.06	0.05
p-Cymene	0.08	—	—	—
Terpinolene	0.03	—	0.06	—
α-Copaene	0.01	—	—	—
β-Cubebene	0.01	—	—	—
β-Elemene	0.01	—	—	—
β-Caryophyllene	0.02	—	—	—
Germacrene D	0.01	0.33	0.07	0.03
Valencene	0.04	—	—	—
Unknowns	0.04	2.17	1.63	1.17

—, Trace or not detected. (T), Tentative compound.

Also, in the Al$_2$O$_3$-MF membrane with an even larger pore size, part of the oil droplets are believed to form a deposit layer and obstruct the pores, causing a reduction in the effective pore size after the middle period of condensation. Koltuniewicz *et al.* (1995) reported that in the microfiltration of an oil-in-water emulsion using a ceramic membrane, after an initial phase of pore blocking, the buildup of a layer of droplets and the resultant switch to cake filtration greatly reduced the performance. López *et al.* (1995) also reported that a zirconia ultrafiltration membrane (Carbosep®) rejected the hydrocarbons contained in a synthetic emulsion made with crude oil, which resulted in a higher flux. The flux performance depends on the droplet size distribution, and the smaller droplets were able to penetrate into the active layer inducing a reduction in the pore size (López *et al.*, 1995). The external pore blocking by cake deposition was caused by the larger droplets and the internal pore plugging was caused by the smaller droplets. Similarly, a suspension of essential oil has a wide droplet size distribution. The reduction in the flux through the Al$_2$O$_3$-MF membrane and the change of the ratio of hydrocarbons and oxygenated compounds during filtration through the Al$_2$O$_3$-MF membrane can be explained by these mechanisms.

Table 2 shows the composition of the essential oil recovered from the permeate according to the functional group and Table 3 shows the percentages of the major components. In the essential oil in the permeate obtained with the 3 ceramic membranes, the percentage of alcohols was highest, followed by esters and aldehydes in this order, and the percentage of alcohols was markedly increased compared with that in the essential oil before filtration (Table 2). Alcohols, which are the most soluble in water, passed through all membranes in the greatest quantities, and the composition of essential oil in the permeates varied widely depending on the membrane type. As mentioned above, the permeate of the ZrO$_2$-UF membrane had the greatest percentage of oxygenated compounds. When the ZrO$_2$-UF membrane was used, the contents of aldehydes, alcohols, and esters in the permeate were increased about 24, 100, and 47 times, respectively, compared with those in the essential oil before filtration. When the suspension of essential oil was filtered through the Al$_2$O$_3$-MF membrane, the percentage of oxygenated compounds was reduced compared with the permeates through the other membranes, because the percentage of hydrocarbons was relatively increased. Also, when the composition of essential oil was compared before and after filtration, the percentage of *d*-limonene, which is the primary component of essential oil, was reduced, and the percentages of the oxygenated compounds, linalool, octanal, and geranial (but not decanal) were markedly increased after filtration. Decanal was found to pass more readily through the Al$_2$O$_3$-MF membrane, which is more permeable to hydrocarbons, than through the ZrO$_2$-UF membrane, which is more permeable to oxygenated compounds, and showed filtration characteristics different from those of other oxygenated compounds. Decanal is estimated to have greater affinity for the hydrocarbon than to water molecules in water suspensions containing essential oil.

Differential recovery of essential oil from the condensates collected from a juice factory using ceramic membranes According to the results of an experiment using the model fluid of the condensates, the percentage of oxygenated compounds in the essential oil recovered from the permeate was highest when the ZrO$_2$-

UF membrane was used for filtration. Therefore, a sample of the condensate from the concentration process of citrus juice produced at a juice factory was concentrated using the ZrO$_2$-UF membrane in the same way that the model fluid was concentrated. The result was that the permeation flux and the changes in the permeation flux were found to be nearly identical to those of the model fluid. Table 4 shows the percentages of major components of essential oil recovered from the permeate.

When the composition of essential oil in the concentrates from the concentration process of citrus juice was analyzed in advance, the percentages of hydrocarbons and oxygenated compounds were 96.4% and 3.6%, respectively. When essential oil was differentially recovered from this condensate using the ZrO$_2$-UF membrane, the percentages of hydrocarbons and oxygenated compounds recovered from the permeate were 30.3% and 69.7%, respectively, and water-soluble essence with a very high content of oxygenated compounds was recovered. Simultaneously, essential oil with a high content of hydrocarbons, which were predominantly *d*-limonene, could be recovered from the retentate. The permeability of essential oil components was 2.4%.

Concerning the composition of the essence recovered from the permeate, the permeability of alcohols was highest, followed by aldehydes. Among hydrocarbons, the percentage of *d*-limonene in essential oil, which was initially 84.3%, decreased to 21.8%, and that of linalool, which is a major oxygenated compound, increased from 0.37% to 14.3%. The permeability of the ZrO$_2$-UF membrane to decanal was as low as the permeability to the model fluid, resulting in only a trace level of decanal in the permeate.

While water-soluble flavors can be recovered from the permeate by various methods including steam distillation and reverse osmosis, the adsorption method using Porapack Q (Sakamoto *et al.*, 1993) employed in this study is a method appropriate for membrane separation in which simple and continuous processing is required. A recovery method for flavors using a styrene-divinylbenzene adsorptive resin has also been reported (Ericson *et*

Table 4. Percentages of major compounds of essential oil in permeate recovered from condensates collected from a juice factory.

	Functional group	Before filtration (%)	ZrO$_2$-UF (%)
Oxygenated-compounds	Aldehydes	0.39	4.06
	Nonanal	0.03	0.34
	Decanal	0.08	0.00
	Alcohols	2.31	51.4
	Linalool	0.37	14.3
	α-Terpineol	0.23	10.6
	1-Octanol	0.01	0.14
	1-Nonanol	0.09	0.73
	Citronellol	0.03	1.49
	Esters	0.38	1.35
	Isobutyl acetate	0.13	0.11
	Linalyl acetate	0.04	0.88
	Others	0.36	9.96
	2-Pentanone	0.05	0.18
	cis-Limonene oxide	0.01	0.41
	Unknown	0.16	2.93
	Total	3.60	69.7
Hydrocarbons	Limonene	84.3	21.8
	γ-Terpinene	5.83	1.99
	p-Cymene	1.17	5.28
	Myrcene	1.19	0.29
	β-Caryophyllene	0.19	0.00
Total		96.4	30.3

al., 1992), so that a recovery method using an absorptive resin is considered sufficiently practical.

These observations suggest that ceramic membranes can be used to differentially recover oil-soluble and water-soluble components of essential oil from condensates obtained from a juice factory as well as from a model fluid. The microfiltration membrane had a higher flux but posed a higher risk of essential oil breakthrough. Therefore, we consider that an ultrafiltration membrane, which has tighter pores, is better suited to ensure a steady permeate quality. Filtration of a suspension of essential oil using a ceramic membrane, in which there is little decrease in flux, is applicable to a wide variety of fields including the manufacturing of terpeneless oil.

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