Membrane and Additional Adsorption Processes for Quality Improvement of Used Frying Oils

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ABSTRACT: The main focus of our study was to improve the quality of used frying oils for recycling as edible oils. Experiments were conducted with hexane-diluted, used frying oils in a batch membrane cell using a nonporous polymeric composite membrane for improving permeate (oil) flux. The oil flux increased by 9- to 14-fold as compared to the permeate flux obtained with undiluted oil. Furthermore, the quality of processed oils was comparable with membrane-processed, undiluted oil. The maximal oil flux was obtained when the weight ratio of oil to oil-hexane retentate (feed) was 0.47 in the process stream. Examination of successive fractions of permeate revealed that membrane performance was not affected by variation in the feed quality. Although the membrane process improved the overall quality of used frying oil, it was not effective in reducing FFA and oxidation products. By combining membrane and adsorption processes, the quality of the used frying oil could be improved to the level of fresh frying oil. Silica gel along with a small amount of magnesium oxide gave the best results in the adsorption process. Combined membrane and adsorption processes seem to be the appropriate approach for the complete regeneration of used frying oils.

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KEY WORDS: Adsorption processing, free fatty acids, hexane, magnesium oxide, membrane processing, nonporous polymeric composite membrane, permeate flux improvement, polar compounds, silica gel, used frying oil.

Waste disposal problems continue to increase and endanger the preservation of the environment. The worldwide production of edible vegetable oils is about 60 million tons a year (1). About 40% of that oil is used for frying oils (2), and a significant quantity of used frying oil is discarded. Color, foaming, and viscosity are the main criteria for discontinuing the use of frying oil in many restaurants and homes, and frying oils generally are discarded after a few uses (3). Kajimoto *et al.* (3) examined used frying oils from Japanese households and reported that PV were in the range of 1–14 meq/kg and anisidine values (AV) were in the range of 0.5–2 mg/g, which indicated that the degree of degradation of used frying oil was not so severe. Therefore, recycling of used frying oils may require only a simple treatment. The recycling of used frying oils not only could reduce disposal problems but also, more importantly, would increase the effective use of food and agricultural resources.

The degradation reactions of oil occur because of heat, air, and moisture, which lead to polymerization, oxidation, and hydrolysis (4). Additional factors contributing to oil breakdown are foreign materials (4). To maintain consistency of frying oil quality, both insoluble particulates and soluble components have to be removed periodically, thereby decreasing the overall rate of degradation.

The membrane process for improving oil quality is remarkably simple and offers many advantages over other processes, namely, low energy consumption, ambient temperature operation, no addition of chemicals, and retention of nutrients as well as other desirable components. Our earlier studies on the processing of used frying oils with hydrophobic nonporous polymeric composite membranes showed that the quality of used frying oils could be improved, as reflected in the reduction of total polar materials (TPM) including polymers, oxidation products, and color compounds (5,6). However, improvement in permeate flux would be necessary for commercial application (5,6). Furthermore, the nonporous membranes used in the study did not reject FFA, resulting in their enrichment in the processed oils (7).

Filtration using passive and active filters can extend the optimal frying region of the oil quality curve. Active filters (i.e., those based on an adsorption process) not only remove the particulates but also alter the chemistry of the frying oil by removing specific oil-soluble chemical compounds such as surfactants and precursors of polymers in the heated oil, thereby decreasing the overall rate of degradation (8). Hoover (9) patented an adsorption process that used magnesium oxide to reduce the FFA content of used frying oil. We also attempted to improve the quality of the used frying oils using an adsorption process and concluded that silica gel offered the most effective overall improvement in the quality of used frying oil as compared to other adsorbents (10).

In the present study, attempts were made to improve the permeate flux for the commercial application of a membrane process by diluting the feed with an organic solvent (hexane). In addition, a combination study of membrane and adsorption processes was carried out to improve the overall quality of used frying oils.

EXPERIMENTAL PROCEDURES

Materials. The departmental canteen of the National Food Research Institute (Kannondai, Tsukuba, Japan) kindly

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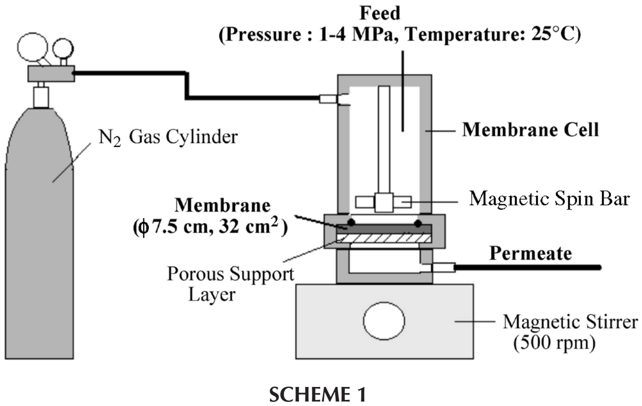
provided samples of fresh soybean frying oil with minimal addition of silicone oil and used frying oils. These samples were divided (60 to 100 g) into individual experimental samples and flushed with nitrogen gas before sealing the bottles and storing in a freezer below −30°C. The frozen samples were melted when required for use. Silica gel (grade 60; particle size, 0.063–0.200 mm) and magnesium oxide (light type) were supplied by Merck Co. (Darmstadt, Germany) and Wako Pure Chemical Industries (Osaka, Japan), respectively.

Membrane. Polymeric composite membrane NTGS-2200, with silicon as the active layer and polyimide as the support layer, was purchased from Nitto Denko Co. Ltd. (Shiga, Japan). This is a nonporous hydrophobic membrane, and separation is mainly affected by a dissolution–diffusion mechanism. Originally developed for gas-separation applications, these membranes are commercially used for hexane vapor recovery in the petroleum industry. The membrane was cut into circular discs (diameter 7.5 cm, effective area 32 cm^2) and fitted in the membrane cell in such a way that the active surface came into contact with the feed material.

Membrane apparatus. Experiments were conducted using a flat membrane test cell (model C-70B; Nitto Denko Corporation) under a nitrogen atmosphere. The membrane apparatus and operating conditions are shown in Scheme 1. The cell was placed on a magnetic stirrer; the magnetic spin bar fitted into the cell provided the agitation. The cell and magnetic stirrer were placed in a thermostatically controlled incubator at 25°C. The required pressure was applied by adjusting the pressure regulator of the nitrogen cylinder. The speed of the spin bar was maintained at 500 rpm.

Membrane batch experiments. The effects of amount of hexane dilution to oil and operating pressure on membrane performance were investigated. Used frying oil (undiluted) was labeled as feed-I. Hexane-diluted feed samples were prepared at four different dilutions by dissolving 50 g of oil in 25, 50, 75, and 100 g of hexane and labeling as feed-II, feed-III, feed-IV, and feed-V, respectively. The operating pressure was maintained at 4 MPa. Feed-I was also processed under otherwise similar conditions for comparison of performance. For studying the effect of pressure, experiments were conducted at 1, 2, 3, and 4 MPa using 100 g of feed-III. Both experiments were stopped when the permeate collections were about 24–30 g as oil.



SCHEME 1

Two batch experiments were conducted for studying the quality of successive fractions of permeates collected over a period of time by charging 100 g of feed-III and 150 g of feed-V, respectively, and maintaining an operating pressure of 4 MPa. Eight successive fractions of permeates were collected during the experimental period (10 g of each permeate for feed-III and 15 g of each permeate for feed-V).

In these experiments, hexane in permeates was removed in a rotary flash evaporator, with trace residuals removed by holding the samples in an evacuated desiccator at 60°C under vacuum (<5 mm Hg of absolute pressure). Solvent-free samples were used for analyses.

Stability of the membrane. These experiments were conducted with feed-III at 4 MPa. The cell was initially charged with 100 g of feed sample and the membrane process was stopped when the processed oil reached 26–27 g (total permeate quantities were 64 g). The cell was then depressurized, and the used membrane and cell were washed with hexane and dried before refitting. Fresh feed (100 g) was charged in the cell before the unit was restarted. These operations were repeated 10 times. Permeates of the first, fourth, seventh, and tenth operations were sampled and analyzed after evaporation of hexane.

Combined membrane and adsorption process. The membrane process was used as a pretreatment step, which was followed by an adsorption process. Membrane experiments were conducted with feed-III at 2 MPa in the batch cell. Oil content in the permeate obtained was 48.5%. The adsorption experiments were conducted as follows. Permeate (80 g) was mixed in flasks along with 0–15 g of silica gel and 0–3 g of magnesium oxide. The mixtures were stirred for 60 min at 25°C and filtered through filter paper (No. 5C; Advantec Toyo Corporation, Tokyo, Japan). Filtrates were analyzed after evaporation of hexane as described earlier.

Analyses. The physicochemical characteristics of various oil samples were determined by the following analyses. The PV, AV, FFA, and TPM were determined using AOCS methods Cd 8b-90, Cd 18-90, Ca 5a-40, and Cd 20-91, respectively (11). The TOTOX value was obtained by the following equation (11).

$$\text{TOTOX} = 2\,\text{PV} + \text{AV} \qquad [1]$$

Viscosity measurements were carried out using a falling-ball viscometer (microviscometer; Haake Corporation, Karlsruhe, Germany) at 25°C.

The sample color was estimated from the absorption spectra in the visible range (400 to 700 nm), and a representative absorbance at 420 nm was also measured. The spectroscopic data were recorded using a spectrophotometer (U-1500; Hitachi Co. Ltd., Tokyo, Japan) in a 10-mm quartz cuvette with hexane as blank. If the absorbance at 420 nm of the sample was over 0.8, the sample was diluted several times with hexane to be measured within the linear range (0.1–0.8).

Performance parameters. The performance of the membrane/adsorption process was expressed in terms of percentage improvement (PI), which is an index used for comparing the quality of the processed oil with that of fresh oil. PI for each component and physical properties of the oil were calculated using Equation 2:

$$PI = 100(C_u - C_p)/(C_u - C_f) \qquad [2]$$

where C_u, C_f, and C_p are the contents of each component or property in the used frying oil, fresh frying oil, and the processed oil.

The total and oil permeate fluxes, J and J_o (kg/m^2·h), were calculated as follows:

$$J = W/(A \cdot t) \qquad [3]$$

$$J_o = W_o/(A \cdot t) \qquad [4]$$

where W and W_o are the weights of the total permeate and oil content in the permeate (kg), A is the effective membrane area (0.0032 m^2), and t is the time taken for collecting the permeate (h).

RESULTS AND DISCUSSION

Batch membrane experiments. The properties of the used and fresh frying oils are given in Table 1. The total and oil permeate fluxes and the properties of the membrane-processed oils at different hexane dilutions and operating pressures are also presented in Table 1. The oil flux increased by 9- to 14-fold with hexane dilution of used frying oil, and the total flux as well as the oil flux increased with an increase in hexane dilution. At a fixed hexane dilution, however, the oil flux remained almost constant regardless of the operating pressure.

The TPM is the "chemical index" used to determine the degree of cumulative degradation of oil (8). TPM is essentially the non-TG fraction of the oil, which is broadly grouped into polymerized and decomposed products based on M.W. and polarity. Frying oil quality is assessed mainly in terms of TPM, and so is the quality of the regenerated used frying oil. PV indicates the peroxide and other similar oxidation products. AV primarily measures the secondary oxidation products, namely, α- and β-unsaturated aldehydes. PV in conjunction with AV can be used to map the past and future degradation profiles. TOTOX is a useful value for quantifying oxygen-directed oil degradation (8). Oxidation products such as peroxides, aldehydes, ketones, hydroperoxides, polymers, and oxidized monomers can cause harmful effects (12). Peroxide formation is a major concern from a toxicological point of view. In the case of undiluted feed (feed-I), the PI of TPM and TOTOX were 61 and 39%, respectively. Processing of hexane-diluted oils under otherwise similar conditions resulted in PI of TPM and TOTOX in the range of 52–61 and 25–41%, respectively. The rejection of TPM and oxidation products by the membrane slightly decreased with the increase in hexane dilution in the feed and with the decrease in operating pressure.

The formation of polymers during frying is mainly responsible for the changes in the viscosity of the oil (12). The reduction in viscosity is a direct indication that polymers, which are the high M.W. fraction of the degradation products, are reduced in the oil. The color increases dramatically during frying and is highly influenced by frying temperature (13). Regulations in a number of countries stipulate that color must be used as a criterion for discarding frying oils (14). Absorbance spectra between 400 and 700 nm were obtained for the fresh, used, and processed oils. The absorbance spectra did not give any peak in this range and showed a pattern of decreasing absorbance with increase in wavelength. Absorbance at 420 nm was used as the color index in the present study. A visual inspection revealed that the color of the oils was brownish, which indicated a higher absorbance at 420 nm. In the case of membrane processing of undiluted oil (feed-I), PI of viscosity and color were 99 and 98%, respectively. Membrane processing of hexane-diluted oils resulted in similar performance in terms of a reduction in viscosity and color with PI in the ranges of 99–104 and 93–97%, respectively.

The FFA value denotes the degree of hydrolysis of the oils. In many countries, FFA value is used as the index for regulation of frying oil (14). It is important to quantify FFA if they are formed in considerable quantities, although they are included in the decomposition product fraction (12). The mem-

TABLE 1
Permeate Flux and Percent Improvement (PI) of Various Properties of Membrane-Processed Used Frying Oils at Different Hexane Dilutions and Operating Pressures[a]

Feed						TPM		TOTOX		FFA		Viscosity		ABS (420 nm)	
Type	Oil-hexane (g)–(g)	Pressure (MPa)	Permeate oil-hexane (g)–(g)	Total flux [kg/(m²·h)]	Oil flux [kg/(m²·h)]	Content (%)	PI (%)	Content (—)	PI (%)	Content (%)	PI (%)	Property (mPa·s)	PI (%)	Property (—)	PI (%)
Used frying oil						13.9		139		0.81		59.0		2.95	
Fresh frying oil						2.5		7		0.04		48.4		0.13	
Feed-I	51–0	4.0	25–0	0.09	0.09	6.9	61	88	39	0.91	−13	48.5	99	0.19	98
Feed-II	50–25	4.0	27–14	1.22	0.80	6.9	61	87	39	0.85	−5	48.5	99	0.22	97
Feed-III	51–52	4.0	26–38	2.52	1.03	7.3	58	90	37	0.86	−7	48.1	103	0.32	93
Feed-IV	50–75	4.0	27–59	3.58	1.12	7.4	57	94	34	0.87	−8	48.2	102	0.27	95
Feed-V	51–103	4.0	24–82	5.48	1.23	7.5	56	101	29	0.91	−13	47.7	107	0.29	94
Feed-III	51–52	3.0	27–37	2.22	0.93	6.9	61	85	41	0.91	−13	48.2	102	0.26	95
Feed-III	50–52	2.0	28–36	2.24	0.98	7.3	58	100	30	0.86	−7	48.0	104	0.29	94
Feed-III	50–52	1.0	30–35	2.15	0.99	8.0	52	106	25	0.81	0	48.0	104	0.32	93

[a] TPM, total polar materials; TOTOX, oxidation products; ABS, absorbance.

TABLE 2
Permeate Flux and PI of Various Properties of Membrane-Processed Used Frying Oils During Membrane Stability Assessment Experiment[a]

Run no.	Feed oil-hexane (g)–(g)	Permeate oil-hexane (g)–(g)	Total flux $[\mathrm{kg/(m^2 \cdot h)}]$	Oil flux $[\mathrm{kg/(m^2 \cdot h}]$	TPM		TOTOX		FFA		Viscosity		ABS (420 nm)	
					Content (%)	PI (%)	Content (—)	PI (%)	Content (%)	PI (%)	Property (mPa·s)	PI (%)	Property (—)	PI (%)
Used frying oil					13.9		139		0.81		59.0		2.95	
1st Operation	51–52	26–38	2.52	1.03	7.3	58	90	37	0.86	−7	48.1	103	0.32	93
4th Operation	50–51	27–37	2.63	1.10	7.4	57	90	37	0.88	−9	48.7	97	0.34	93
7th Operation	50–51	27–37	2.61	1.10	8.0	52	103	27	0.90	−12	49.5	90	0.42	90
10th Operation	51–52	27–37	2.61	1.09	8.7	46	105	26	0.91	−13	49.5	90	0.45	89

[a] For abbreviations see Table 1.

brane process did not reduce FFA content of used frying oil irrespective of hexane dilution and operating pressure.

Dilution of oil with hexane in the membrane process improved the oil flux significantly, and the properties of the processed oils were similar to those obtained with undiluted processed oil. Although the PI value of TPM was more than 50%, regeneration of the oil is still incomplete until TPM, including TOTOX and FFA, is completely eliminated from the oil. This necessitates an additional processing step that could take care of the final quality even when the degradation is very high in the used oil. An adsorption process was proposed for this purpose.

Stability of the membrane. Table 2 shows total and oil flux, and PI of TPM, TOTOX, FFA, viscosity, and color of membrane-processed oils. Although 10 runs were conducted with the same membrane, only the results obtained with runs 1, 4, 7, and 10 are presented. Total flux and oil flux were maintained between 2.5–2.6 and 1.0–1.1 kg/m^2·h, respectively, during the 10 experimental runs. However, PI values of all properties of the processed, used frying oil decreased marginally with every run. The extent of decrease of PI from the first to tenth run was about 10% for TPM, TOTOX, and viscosity, and 4% for color. The PI value of FFA also showed a slight decrease with every run as indicated by the increase in its negative value. For commercial applications, stability of the membrane is very important. In our previous study, long-term experiments using the same series of membranes (NTGS-2100 and NTGS-1100) with undiluted crude oil were conducted up to 97 d at appropriate operating conditions (15). During the experiments, the permeate flux and processed oil qualities remained almost constant (15). However, present stability experiments with hexane-diluted feed revealed a slight decrease in the performance of the membrane with every experimental run. It is desirable to investigate further the long-term stability of membranes with hexane-diluted oils as well as to standardize the cleaning procedure to be adopted between experimental runs.

Successive fractions of permeates in batch experiments. Changes in the quality of the permeate (TPM) were assessed during the membrane process with eight successive permeate fractions collected from each of the two batch runs using feed-III and feed-V. Mean values of TPM of the first and second, third and fourth, fifth and sixth, and seventh and eighth fractions are presented in Table 3.

In any batch membrane process, certain compounds are rejected during the process and their concentrations increase in the retentate with time. Therefore, the concentration of rejected compounds in the batch cell is higher with every collection of permeate fraction. The increased concentration of these compounds in turn enhances their diffusion through the membrane, resulting in a higher concentration of these compounds in the permeate. However, in the present case, rejection of TPM remained constant for feed-III and increased slightly for feed-V as indicated by the PI values (Table 3). Edible oil is a multicomponent system, and some of the polar compounds are rejected by the membrane, while others, such as FFA and tocopherols, are preferentially permeated through the membrane (7). In addition, permeation rates of hexane were greater than the oil constituents, and the ratio of oil to hexane increased with every fraction collected during the membrane processing, which may also have some influence on the rejection performance of the membrane. This appears to be the reason for the observed trend of rejection that was contrary to the general expectation. The results suggested that membrane performance was not affected significantly with the variation in feed quality.

Permeate flux. Permeate flux is an important factor in the membrane process as it has a direct bearing on the process economics. The data obtained with successive fractions of permeates collected in the batch experiments using feed-III and feed-V were analyzed for optimizing the hexane dilution to obtain maximal permeate flux (oil flux). Total flux compared

TABLE 3
TPM Content of Successive Fractions of the Permeate in a Batch Membrane Experiment[a]

	Permeate oil-hexane (g)	TPM	
		Content (%)	PI (%)
Feed-III	50.1–51.7	13.9	
1st + 2nd	5.9–14.1	7.5	56
3rd + 4th	7.5–12.5	7.6	55
5th + 6th	10.2–9.8	7.5	56
7th + 8th	14.4–5.6	7.5	56
Feed-V	50.6–101.5	13.9	
1st + 2nd	3.9–26.1	7.8	54
3rd + 4th	5.4–24.6	7.5	56
5th + 6th	8.2–21.8	7.3	58
7th + 8th	14.9–15.1	7.2	59

[a] For abbreviations see Table 1.

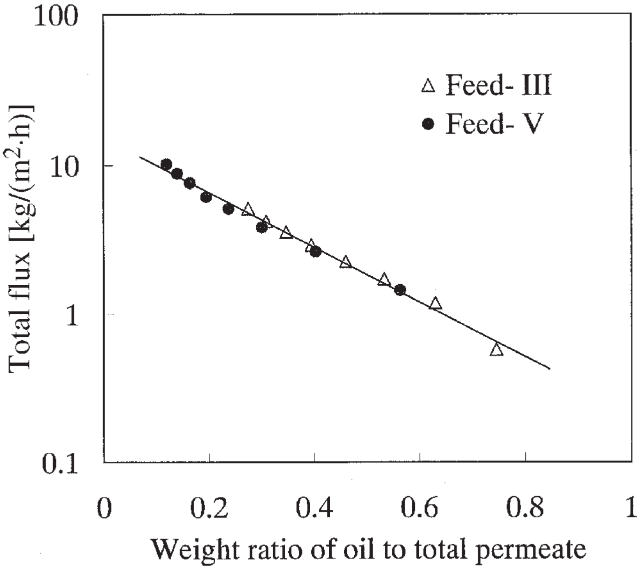


FIG. 1. Relationship between weight ratio of oil to total permeate and total flux during batch experiments [feed-III, 1:1 (w/w) oil/hexane; feed-V, 1:2 (w/w) oil/hexane].

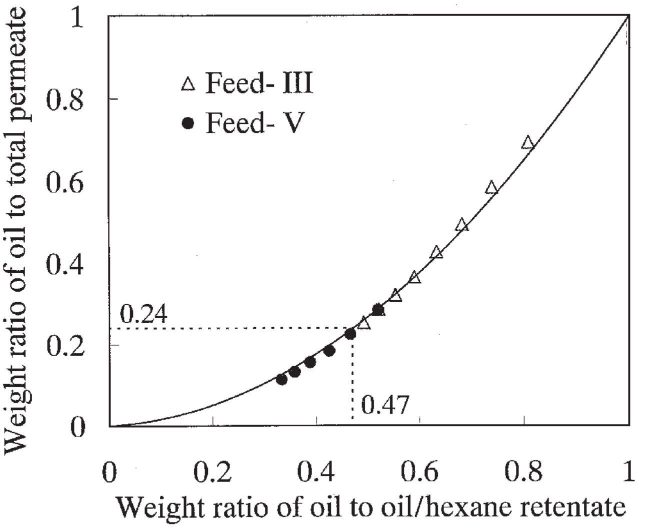


FIG. 2. Relationship between weight ratio of oil to oil/hexane retentate and weight ratio of oil to total permeate during batch experiments [feed-III, 1:1 (w/w) oil/hexane; feed-V, 1:2 (w/w) oil/hexane].

to the weight ratio of processed oil to total permeate for every fraction collected was plotted in Figure 1. The experiment was conducted in a batch mode and, despite the changes in the hexane dilution of the retentate, the total flux, J [kg/(m^2·h)], and the weight ratio of oil to total permeate, Y, had a good correlation and was approximated by Equation 5:

$$J = 15.3 \exp(-4.24Y) \qquad (R^2 = 0.991) \qquad [5]$$

The oil flux, J_o (kg/m^2·h), can be described as follows:

$$J_o = YJ = 15.3Y \exp(-4.24\,Y) \qquad [6]$$

By differentiating Equation 6 with respect to Y and equating the function to zero, the condition for maximal J_o is obtained [$Y = 0.24$; corresponding to $J_o = 1.33$ (kg/m^2·h)].

Weight ratios of oil to oil-hexane retentate, X, against the weight ratio of oil to total permeate, Y, were plotted in Figure 2. The relationship between them is approximated by Equation 7:

$$Y = 0.93\,X^2 + 0.07\,X \qquad (R^2 = 0.996) \qquad [7]$$

From Equation 7, the X value for maximal J_o is obtained as 0.47, corresponding to a Y value of 0.24.

A plant with 100 m^2 membrane area could process about 60 tons of used frying oils in a year (7000 h of operation) with undiluted used frying oil as the feed to the process. The same plant would be able to handle about 900 tons of used frying oils in a year if the feed were diluted with hexane, maintaining an oil to oil/hexane ratio of 0.47 during the plant opera-

TABLE 4
PI of Various Properties of Membrane- and Additional Adsorbent-Processed Used Frying Oils[a]

Silica gel–magnesium oxide (g)	TPM Content (%)	TPM PI (%)	TOTOX Content (—)	TOTOX PI (%)	FFA Content (%)	FFA PI (%)	Viscosity Property (mPa·s)	Viscosity PI (%)	ABS (420 nm) Property (—)	ABS (420 nm) PI (%)
Used frying oil	13.9		139		0.81		59.0		2.95	
Membrane-processed oil										
0–0	8.3	49	109	23	0.82	–1	49.3	92	0.44	89
Additional adsorbent-processed oil										
0–3.0	8.5	47	108	24	0.27	71	49.0	94	0.40	90
3.0–0	6.6	64	83	42	0.68	17	48.8	96	0.25	96
3.0–3.1	7.1	60	82	43	0.16	85	48.7	97	0.28	95
6.0–0	5.0	78	60	60	0.58	29	48.5	99	0.21	97
6.0–3.0	4.9	79	60	60	0.12	89	47.9	105	0.23	96
10.0–0	3.6	90	41	75	0.48	43	47.4	109	0.16	99
10.0–1.5	3.3	93	42	74	0.24	74	47.3	110	0.16	99
15.0–0	2.1	104	26	86	0.38	56	47.7	107	0.12	100
15.0–1.5	2.4	101	27	85	0.17	83	48.0	104	0.13	100

[a] For abbreviations see Table 1.

tion. The increase of oil flux by 15-fold would sharply improve the economics of the process.

Combined membrane and adsorption process. In these experiments, membrane-processed oils were further treated by an adsorption process using silica gel and magnesium oxide. The results obtained with different combinations of silica gel and magnesium oxide are presented in Table 4. Although the membrane process improved the quality of the used frying oil, oxidation products were not reduced much and FFA preferentially permeated as indicated by a negative PI value.

TPM and TOTOX of used frying oils showed greater reduction when membrane and silica gel processes were combined. Increasing the amount of silica gel in the adsorption process improved the performance, and PI using 15 g of silica gel (dosage of silica gel on oil, 38%) in the combined process were 104 and 86% for TPM and TOTOX, respectively. However, reduction in FFA was not satisfactory when only silica gel was used in the adsorption process. When magnesium oxide (3 g) only was used in the adsorption process, the PI of FFA was 71%. However, TPM and TOTOX remained unaltered. These results were in agreement with our previous study on screening the adsorbents (10). The amount of TPM per unit weight of silica gel was about 220 mg/g. The amounts of FFA adsorbed per unit weight of silica gel and of magnesium oxide were about 18 and 71 mg/g, respectively. The membrane process was very effective in reducing the polymers and color compounds, which is reflected by greater PI of 92 and 89% for viscosity and color, respectively. Viscosity and color improved further by the subsequent adsorption process. Color improvement was not very sharp with magnesium oxide as compared to silica gel in the adsorption process.

Further adsorption experiments were conducted using both silica gel and magnesium oxide with membrane-processed used frying oils. The adsorption process with 15 g silica gel (dosage of silica gel on oil, 38%) and 1.5 g of magnesium oxide (dosage of magnesium oxide on oil, 3.8%) gave the best results; PI of TPM, TOTOX, FFA, viscosity, and ABS were 101, 85, 83, 104, and 100%, respectively. The final quality of the processed oil obtained from the combined membrane and adsorption process was almost the same as that of the fresh oil. The results showed that silica gel together with a small amount of magnesium oxide was more effective in the adsorption process than individually as adsorbents. A combined membrane and adsorption process seems to be the appropriate approach for the complete regeneration of used frying oils and is suitable even for highly degraded, used frying oil.

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