Regeneration of Used Frying Oils Using Adsorption Processing

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ABSTRACT: Our study was carried out to improve the quality of used frying oils and to assess the feasibility of recycling by using an adsorption process. Experiments with used frying oils were conducted with a column method using four adsorbents, silica gel, magnesium oxide, activated clay, and aluminum hydroxide gel. Silica gel offered the most effective overall improvement in the quality of used frying oil of the four adsorbents. Although the limiting adsorption capacity of silica gel, as obtained from the Langmuir model, was 219 mg/g for total polar materials (TPM), the experimental maximal adsorption capacity of silica gel for TPM was 337 mg/g. For commercial application, silica gel used in the process must periodically be reactivated. Reactivated silica gel also was effective to some extent in the adsorption process, although the adsorption capacity decreased by 20–50%.

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Waste disposal problems continue to increase and to endanger environmental preservation. Recycling used frying oils not only could reduce disposal problems, but, more importantly, would increase the effective usage of food and agricultural resources.

The worldwide production of edible vegetable oil is about 60 million t/yr. The majority of oil produced is used for frying foods, and a significant quantity of used frying oil is discarded. Surveys have indicated that color, foaming, and viscosity are the main criteria in many restaurants and homes for discontinuing the use of frying oil (1). However, the degree of degradation in used frying oil is often not severe (1), and therefore recycling of used frying oils may require only a simple treatment. Oil degradation is due to three factors—heat, air, and moisture, which lead to polymerization, oxidation, and hydrolysis (2). Such degradation affects the usability of frying oil negatively. To maintain consistency of frying oil quality, these degradation effects must be eliminated.

Our earlier studies on the processing of used frying oils using 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), polymers, oxidation products, and color compounds (3,4). However, improvement in permeate flux would be necessary for commercial application, and there was deterioration in the oxidative stabilities of the processed oils. Furthermore, the nonporous membranes used in the study showed higher selectivities for FFA over TG, resulting in their enrichment in the processed oils (5).

Filtration using passive and active filters can extend the optimal frying region (6). Passive filters remove only insoluble particulates of certain sizes and have little effect on the actual chemistry of a frying oil. On the other hand, active filters not only remove 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.

Many active filtration processes, that is, adsorption processes, have been investigated to improve frying oil quality. Activated clay is well known as an oil-bleaching agent. The effect of the magnesium oxide adsorption process to remove FFA from frying oils has been patented (7). Yuki and Hirose (8) examined a number of adsorbents and reported that aluminum hydroxide gel was the most effective for removing FFA. Usuki (9) also attempted to improve the colors and stabilities of used frying oils by using adsorbents such as zeolite, activated carbon, activated clay, aluminum hydroxide gel, oyster shell, ceramic plate, and calcium oxide. In these reports, researchers attempted to remove part of the degradation products of oils. However, reductions in polymers and oxidation products as well as an improvement of overall quality of used frying oil in terms of TPM as a result of using these adsorbents were not reported.

In the present study, we attempted to improve the overall quality of used frying oil for recycling by using adsorption processing with silica gel, magnesium oxide, aluminum hydroxide gel, and activated clay. The physicochemical characteristics of the processed oils, including TPM, oxidation products, polymerization, FFA, color, and oil stability, were studied in detail to enable the selection of the most effective adsorbent for improving the overall quality of oil. The performance of the selected adsorbent was assessed under various operating conditions. In addition, we investigated reactivation of the adsorbent, important for commercial application.

EXPERIMENTAL PROCEDURES

Materials. The departmental canteen of the National Food Research Institute (Kannondai, Tsukuba, Japan) kindly provided used and fresh soybean frying oil with minimal addition of silicone oil, which we labeled UO-I and FO-I, respectively. Used and fresh frying oils were also provided from a private Japanese home (labeled UO-II and FO-II). In addition, we made four subtypes of the UO-I and FO-I oils for a batch adsorption experiment. The properties of the subtype oils were 24.2, 13.2, 8.6, and 4.4% TPM, and 0.91, 0.78, 0.46 and 0.12% FFA, respectively. Samples (60–100 g) were taken and bottled as individual experimental samples, charged with nitrogen gas, and stored in a freezer at temperatures below −30°C. The frozen samples were melted as required for use.

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Silica gel (silica gel 60; particle size, 0.063–0.200 mm) was supplied by Merck Co. Ltd. (Darmstadt, Germany). Magnesium oxide (light type) and activated clay were procured from Wako Co. Ltd. (Osaka, Japan). Aluminum hydroxide gel (Kyoward 200) was supplied by Kyowa Chemical Co. Ltd. (Kagawa, Japan).

Experiments. Experiments were conducted using column and batch adsorption processes. The column process was used for the adsorbent selection experiment, for the investigation of the effect of adsorbent quantity on oil properties, and for the successive adsorption and adsorbent reactivation experiments. Batch processing was used to investigate the adsorption isotherms of TPM and FFA with silica gel. In all the experiments, the temperature was maintained at 25°C.

Column adsorption process. Five- to 15-g adsorption layers were formed in a funnel (diameter, 40 or 60 mm; Kiriyama Corporation, Tokyo, Japan) with filter paper (No. 5A; Advantec-Toyo Corp., Tokyo, Japan) as the support layer. A known quantity of used frying oil was poured into the filter and allowed to pass through the adsorption layer by gravity; 10- to 35-g portions of the filtrate were then collected.

Adsorbent selection experiment. Column adsorption processing was conducted using 10 g of the four adsorbents (silica gel, magnesium oxide, activated clay, and aluminum hydroxide gel) and a funnel (diameter 60 mm); 25 g of filtrates of UO-I were obtained. The processed oils were evaluated in terms of TPM, TOTOX, FFA, viscosity, and color (absorbance at 460 nm). Oil stabilities in terms of induction times (IT) were also estimated.

Effect of adsorbent quantity on oil properties. The effect of the adsorbent quantity was investigated with silica gel, which exhibited good performance in the adsorbent selection experiment. Column adsorption processes were conducted using 5- to 15-g amounts of silica gel and a funnel (diameter 40 mm). About 35 g of UO-I and -II filtrates were obtained.

Successive adsorption experiment. A 10-g silica gel layer was formed in the funnel (diameter 40 mm), and column adsorption processes were conducted as follows. For UO-I, seven successive filtrates were collected in turn. The successive filtrate quantities of the first to seventh times were 10, 10, 10, 10, 10, 25, and 25 g, respectively. For UO-II, five successive filtrates were collected, and the successive filtrate quantities of the first to fifth times were 25, 25, 25, 25, and 32 g, respectively.

Adsorbent reactivation experiment. After the column adsorption process using 10 g of silica gel, a funnel (diameter 40 mm), and UO-I, the adsorbent was reactivated as follows. A preliminary combustion of the used silica gel was conducted to burn almost all of the adsorbates on the silica gel, and the pretreated silica gel was maintained in an electric furnace at 550°C for 30 min. The reactivated silica gel was then reused in a column adsorption process. This was repeated 30 times. Twenty-five-gram filtrates of UO-I were collected and their characteristics were determined.

Batch adsorption experiment. Fifty grams of used frying oil and 2.5–15 g of silica gel were mixed in flasks. A preliminary study showed that the adsorption equilibrium of TPM could be reached after 10 min of mixing at 25°C. Therefore, to ensure the equilibrium state of the system, the mixtures were stirred for 30 min at 25°C and poured into the Kirishima funnel with filter paper (No. 5A) to obtain 20-g filtrates.

Analyses. The physicochemical characteristics of the adsorbent-processed oils were determined by the following analyses. The PV, anisidine value (An.V), FFA, iodine value (IV), TPM, and tocopherols (vitamin E) were determined using AOCS methods Cd 8b-90, Cd 18-90, Ca 5a-40, Cd 1-25, Cd 20-91, and Ce 8-89, respectively (10). The TOTOX value was obtained by the following equation (10):

$$\text{TOTOX} = 2\text{PV} + \text{An.V} \qquad [1]$$

PV represents peroxide and other similar oxidation products. An.V primarily measures the secondary oxidation products, namely α- and β-unsaturated aldehydes. PV in conjunction with An.V can be used to map past and future degradation profiles. Totox is useful for quantifying oxygen-directed oil degradation (6).

The IT for oxidation was determined using an oil stability test system (679 Rancimat; Metrohm Corporation, Herisau, Switzerland) (AOCS, Cd 12b-92) (10). The sample weight was 3.0 g, the temperature was 95°C, and the air flow rate was maintained at 20 L/h.

Viscosity measurements were performed using a falling-ball viscometer (Microviscometer; Haake Corp., Karlsruhe, Germany) at 25°C.

Sample color was determined by the absorption spectra in the visible range (400 to 700 nm). Spectroscopic data were recorded using a spectrophotometer (V-570; Jasco Corp., Tokyo, Japan) in a 10-mm quartz cuvette with methylene chloride as the blank. A representative absorbance at 460 nm was determined.

Most of the analysis data, except for the viscosity, were obtained in duplicate and are presented as the mean values when data points did not differ more than 3–5%. When difference was more than 3–5%, a few more data points were collected and mean values were determined. For the viscosity measurements, 5–10 replicates were collected, and the mean values were obtained. All SD of the viscosity values were within ±0.4 mPa s.

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

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

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

RESULTS AND DISCUSSION

Properties of UO-I, FO-I, UO-II, and FO-II. The properties of the UO-I, FO-I, UO-II, and FO-II oils are shown in Table 1.

The deterioration in quality of UO-I from the departmental canteen was greater than the deterioration in quality of UO-II from the private home.

The IT measurements of UO-I and FO-I were 19.2 and 10.5 h, respectively. A decrease in IT indicates deterioration of oxidative stability. The degree of unsaturation of FA and the presence of antioxidants and prooxidant metals in the oil contribute significantly to oxidative stability. The IV values of UO-I and FO-I were approximately the same. This suggests that the degree of unsaturation of FA had not changed and hence may not be responsible for the decreased oil stability of UO-I. Antioxidants are reduced in the oil during frying; this appears to be the main reason for the decreased oil stability of UO-I compared to FO-I.

Vitamin E is an important antioxidant. Vitamin E values of FO-I and UO-I were 507 and 396 mg/kg, respectively, which is a difference of 20%. Hence, vitamin E would have some influence on the oxidative stabilities of frying oils.

Adsorbent selection experiments. The properties of the oils that had been processed by the individual adsorbents are shown in Table 2. Included in TPM are polar substances, such as MG, DG, and FFA, which occur in unused oils, as well as polar compounds that are formed during the frying of foodstuffs and/or during heating (10). Nonpolar materials are mostly unaltered TG (10). 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. Of the four adsorbents studied, silica gel exhibited the highest removal of TPM with a PI value of 70.1%. Magnesium oxide had the lowest PI value, 4.7%, whereas the activated clay PI value was 23.3% and aluminum hydroxide gel achieved 31.8%. TPM is the chemical index used to determine the degree of cumulative degradation of oil and is an excellent predictor of food quality for many operations (6). Fritsch (11) reported that determination of TPM in frying fat provides the most reliable measure of the extent of deterioration in the majority of cases. In this context, the higher reduction of TPM is significant and indicates the feasibility of recycling used frying oil as well as retaining fried-food quality over a longer duration of fryer operation.

Oxidation products, such as peroxides, aldehydes, ketones, hydroperoxides, polymers, and oxidized monomers, can cause clinically harmful effects (12). Peroxide formation is a major concern from a toxicological point of view. Activated clay showed a greater reduction of peroxides in the processed oil, producing a PI value of 95.0%, whereas silica gel showed the highest reduction of An.V. (PI 81.6%) as well as TOTOX values (PI 68.9%) in the processed oil. Magnesium oxide and aluminum hydroxide gel processing reduced the TOTOX value only slightly. Silica gel was found to be the most effective of the adsorbents for removing oxidation products.

The FFA value is an indication of the degree of hydrolysis of oils. If FFA are formed in considerable quantities, it is important to quantify FFA separately, even though they are included in the decomposition product fraction (12). FFA in the oil on the surface of a fried food offer a "sink" for oxygen in a food-containing package and can be used to predict the shelf life of the food with respect to the development of rancidity (6). Magnesium oxide and aluminum hydroxide gel decreased the FFA content, with a PI value in the range of 87 to 89%. The PI value in terms of reduction of FFA with silica gel was 68%, whereas activated clay did not decrease the FFA content of used frying oil.

The formation of polymers during frying is mainly responsible for the changes in the viscosity of the oil (12). The viscosity of the frying oil is an important factor determining the total volume of oil sticking in the large cavities in the crust of the food product. Higher viscosity results in a larger volume of oil in the fried food (12). Hence, reduced viscosity will result in less oil adsorption in fried products. Reduction in viscosity is a direct indication that the content of polymers, which are the high-M.W. fraction of the degradation products, has been reduced in the oil. Silica gel and aluminum hydroxide substantially reduce the viscosities of used frying oils, with PI values of 46.0 and 50.0%, whereas activated clay is not very effective in reducing the viscosity as indicated by the lower PI value of 28.4%. Magnesium oxide did not reduce the viscosity of used frying oil.

Surveys (1) have indicated that color is the main criterion for discontinuing usage of frying oil in many restaurants and homes. Regulations in a number of countries stipulate that color must be used as a criterion for discarding frying oils (13). The absorbance spectra for the fresh, used, and processed oils were determined between 400 and 700 nm. The absorbance of each oil decreased with the increase of wavelength, without any peaks. Absorbance at 460 nm was employed as the color index for the present study. The color of oils, with a higher absorbance at 460 nm, was brownish. Of the four adsorbents evaluated, activated clay was the most effective for improving the color of used frying oil. Used frying oils were also effectively decolorized by silica gel and aluminum hydroxide gel.

TABLE 1
Properties of Used and Fresh Frying Oils I and II[a]

	TPM (%)	PV (meq/kg)	An.V (meq/kg)	TOTOX (meq/kg)	FFA (%)	Viscosity (mPa s)	ABS (460 nm) (—)	IV (—)	I.T. (h)
UO-I	13.2	28.4	79.4	136	0.78	55.8	1.36	129	10.5
FO-I	2.5	2.4	2.4	7.2	0.04	48.4	0.146	125	19.2
UO-II	4.4	6.7	21.5	34.9	0.12	50.6	0.595	—	—
FO-II	3.2	2.4	1.2	6.0	0.02	47.8	0.018	—	—

[a] UO-I, used frying oil-I; FO-I, fresh frying oil-I; UO-II, used frying oil-II; FO-II, fresh frying oil-II; TPM, total polar materials; An.V, anisidine value; TOTOX, oxidation products; ABS, absorbance; IV, iodine value, I.T., induction time (index for oil stability).

TABLE 2
Properties and Percent Improvement (PI) of Silica Gel, Magnesium Oxide, Activated Clay, and Aluminum Hydroxide Gel Adsorption-Processed Oil[a]

	TPM		PV		An.V		TOTOX		FFA		Viscosity		ABS (460 nm)		
	Content (%)	PI (%)	Content (meq/kg)	PI (%)	Content (meq/kg)	PI (%)	Content (meq/kg)	PI (%)	Content (%)	PI (%)	Property (mPa s)	PI (%)	Property (—)	PI (%)	I.T. (h)
Silica gel	5.7	70.1	15.4	50.0	16.6	81.6	47.4	68.9	0.28	67.6	52.4	45.9	0.585	63.8	10.0
Magnesium oxide	12.7	4.7	25.7	10.4	71.6	10.1	123	10.2	0.14	86.5	55.3	6.8	0.895	38.3	7.7
Activated clay	10.7	23.3	3.7	95.0	63.2	21.0	70.6	50.9	0.77	1.4	53.7	28.4	0.428	76.8	5.3
Aluminum hydroxide gel	9.8	31.8	17.2	43.1	66.4	16.9	101	27.4	0.12	89.2	52.1	50.0	0.562	65.7	8.9

[a] For abbreviations see Table 1.

The IT was 10.5 h for used frying oil, 10.0 h for oil processed by silica gel, 7.7 h for oil processed by magnesium oxide, 5.3 h for oil processed by activated clay, and 8.9 h for oil processed by aluminum hydroxide gel. Oil stabilities deteriorated in the magnesium oxide, activated clay, and aluminum hydroxide gel processes. IT values of used frying oil and silica-gel-processed oil were almost the same, indicating that the silica gel process did not cause deterioration in oil stability.

On the other hand, vitamin E values were 396 mg/kg for UO-I, 368 mg/kg for silica-gel-processed oil, 397 mg/kg for magnesium-oxide-processed oil, 376 mg/kg for aluminum-hydroxide-gel-processed oil, and 363 mg/kg for activated-clay-processed oil. Percent reductions of vitamin E contents were in the range of 0–8%. Although adsorption processes, except for the silica gel process, resulted in the deterioration of oxidative stabilities, vitamin E content was not reduced by each adsorption process. Oxidative stability of each processed oil would depend on other antioxidants and prooxidants as well as vitamin E.

The magnesium oxide and aluminum hydroxide gel processes significantly reduced FFA in used frying oil, but the removals of TPM and oxidation products were not satisfactory. In the activated clay process, the used frying oil was decolorized, but the FFA content was not reduced. In contrast, the silica gel process reduced FFA, decolorized the used frying oil, and reduced the TPM and oxidation products. The silica gel process also reduced the viscosity of used frying oil. The stability of the silica-gel-processed oil did not change, whereas the other processes resulted in the deterioration of oil stability. The most effective adsorbent for improving the overall quality of oil was silica gel, and further investigations were carried out with this adsorbent.

Effect of adsorbent quantity on oil properties. The properties of the silica-gel-processed oils are shown in Table 3 as they relate to the quantity of silica gel used. The PI value of each component increased with the increase in the silica gel quantity.

For UO-I, when the silica gel quantity was 5 g (ratio of filtered oil to silica gel, 7:1), PI values of TPM, TOTOX, and FFA were 33–35% and PI values of viscosity and ABS were 53–56%. When the silica gel quantity was 10 g (ratio: 3.5:1), PI values of TPM, TOTOX, and FFA were 60–72%, and PI values of viscosity and ABS were 82 to 94%; thus, each component was considerably improved. When the silica gel quantity was 15 g (ratio: 2.3:1), PI values were 79 to 99%, and the processed oil was improved to the level of fresh oil. IT values were almost constant, in spite of the changing silica gel quantities.

For UO-II, when the silica gel quantity was 5 g (ratio: 7:1), the PI value of TPM was 208%, and the overall quality of UO-II was improved beyond that of fresh oil. When the silica gel quantity was 10 g (ratio: 3.5:1), the PI value of TOTOX, FFA, and viscosity were 94 to 100%, and each property was improved to the level of fresh oil. When the silica gel quantity was 15 g (ratio: 2.3:1), the PI value of TOTOX, FFA, and viscosity were over 100%, as well as TPM. However, the PI value of absorbance was 85.4%, even though the silica gel quantity was 15 g.

Successive adsorption experiment. Properties of the silica-gel-processed oils during the successive adsorption experiments are shown in Table 4. The quantity of silica gel used was 10 g. The PI of each component progressively decreased with the increasing number of adsorption processes.

For UO-I, the processed oil was improved to the level of fresh oil in the first filtration. The PI values for TPM and TOTOX were almost the same at each collection of filtrate, falling off rapidly to 15 to 21% by the fifth filtration. The PI values for FFA at the third and fourth filtrations were 27 and 7%, much less than for TPM and TOTOX. The PI value for viscosity decreased in a similar manner to the cases of TPM and TOTOX. The PI value for absorbance decreased less than the others and reached a constant value of 45% after the fifth filtration.

For UO-II, at the first filtration PI values of all indexes except the absorbance value exceeded 100%. The PI value for TPM remained over 100% until the third filtration. The PI value for TOTOX decreased rapidly to 29% by the third filtration. The PI values for FFA and viscosity gradually decreased. The PI value for absorbance was only 84% at the first filtration but continued at an almost constant value of 70 to 84% until the fifth filtration.

The total adsorbed quantity (TQ) of TPM per unit weight of silica gel in the successive adsorption experiment is also shown in Table 4. For UO-I, the TQ of TPM was saturated at 350 mg/g. The saturation point of TPM was reached at the sixth filtration (ratio of oil to silica gel: 7.5:1). For UO-II, the TQ

TABLE 3
Properties and PI of Oils Processed by Different Quantities of Silica Gel Adsorbent[a]

	TPM		TOTOX		FFA		Viscosity		ABS (460 nm)		
Adsorbent[b] (g)	Content (%)	PI (%)	Content (meq/kg)	PI (%)	Content (%)	PI (%)	Property (mPa s)	PI (%)	Property (–)	PI (%)	I.T. (h)
For UO-I											
5 (g)-35 (g)	9.7	32.7	92.1	34.2	0.52	35.1	51.9	52.7	0.676	56.3	10.4
10 (g)-35 (g)	5.5	72.0	59.5	59.5	0.25	71.6	49.7	82.4	0.222	93.7	9.7
15 (g)-35 (g)	4.5	81.3	34.1	79.1	0.15	85.1	48.6	97.3	0.159	98.9	10.2
For UO-II											
5 (g)-35 (g)	1.9	208	16.8	62.6	0.05	70.0	48.8	64.3	0.149	77.3	—
10 (g)-35 (g)	1.0	283	7.7	94.1	0.02	100	47.9	96.4	0.18	82.7	—
15 (g)-35 (g)	0.6	317	2.7	111	0.02	100	46.5	146	0.102	85.4	—

[a] For abbreviations see Tables 1 and 2.
[b] Amount of adsorbent used with 35 g of the specified oil.

increased slowly compared to UO-I because of the low TPM content.

Batch adsorption isotherm experiment. The adsorption isotherm for TPM on silica gel was investigated in a batch adsorption experiment. The adsorption isotherm is expressed as q_e, the amount of TPM adsorbed per unit weight of adsorbent, as a function of C, the TPM content at equilibrium. Figure 1 shows the adsorption isotherm of TPM on silica gel. This is an S-type isotherm. In general, an S-type isotherm is estimated by a BET isotherm corresponding to multilayer adsorption (14). However, the S-type isotherm of TPM was not reflected by the BET isotherm. Hence, in the range of results up to inflection, the Langmuir isotherm model was employed, assuming that a first monomolecular layer is adsorbed on the silica gel.

The linear representation of the Langmuir isotherm is as follows (14):

$$C/q_e = 1/(b \cdot Q^o) + C/Q^o \qquad [3]$$

$$q_e = (C_o - C)/W \qquad [4]$$

where C_o and C are the initial and residual concentrations at equilibrium, Q^o is the limiting adsorption capacity (the limiting amount of solute adsorbed per unit mass of adsorbent in forming a complete monolayer on the surface), b is a constant related to the energy of adsorption, and W is the mass of adsorbent per unit mass of the used frying oil. Q^o was calculated from the linear form of the equation. The linear form by plotting C/q_e as a function of C of TPM is also shown in Figure 1. Q^o for TPM was determined to be 219 mg/g.

The adsorption isotherm of TPM showed break points (180 mg/g) after the Langmuir isotherm model was fitted to the data. The presence of the break point may be due to interactions between the same or different compounds as well as to multilayer adsorption. Thus, in the successive adsorption experiment using UO-I, the TQ of TPM was 1.5 times larger than Q^o (Table 4).

Adsorbent reactivation experiment. For commercial applications, the adsorbed silica gel must be recycled due to the low degree of limiting adsorption capacity for each component. To evaluate the characteristics of reactivated silica gel, the adsorption capacity of the reactivated silica gel was estimated for each component. The reactivation ratio was expressed as follows:

$$\text{reactivation ratio (\%)} = 100 \cdot (C_u - C_n)/(C_u - C_0) \qquad [5]$$

TABLE 4
Properties, PI, and Total Adsorbed Quantities (TQ) of Silica Gel Adsorbent-Processed Oils During Successive Experiments[a]

		TPM			TOTOX		FFA		Viscosity		ABS (460 nm)	
Fraction number	Weight of filtrate (g)	Content (%)	PI (%)	TQ (mg/g)	Content (meq/kg)	PI (%)	Content (%)	PI (%)	Property (mPa s)	PI (%)	Property (–)	PI (%)
For UO-I												
1st	0–10	2.8	97.2	104	16.5	92.8	0.07	95.9	48.1	104	0.127	102
2nd	10–20	4.9	77.6	187	47.5	68.8	0.15	85.1	49.3	87.8	0.317	85.9
3rd	20–30	7.7	51.4	242	66.6	54.0	0.58	27.0	50.9	66.2	0.531	68.3
4th	30–40	9.4	35.5	280	96.7	30.6	0.73	6.8	52.5	44.6	0.712	53.4
5th	40–50	11.0	20.6	302	117	14.7	0.75	4.1	53.4	32.4	0.795	46.5
6th	50–75	1.8	13.1	337	130	5.3	0.74	5.4	54.1	23.0	0.828	43.8
7th	75–100	13.3	-0.1	337	131	3.7	0.75	4.1	54.5	17.6	0.844	42.6
For UO-II												
1st	0–25	0.9	292	88	5.4	102	0.02	100	46.6	143	0.110	84.1
2nd	25–50	1.8	217	153	14.5	70.6	0.04	80.0	48.0	92.9	0.148	77.5
3rd	50–75	3.2	100	183	26.5	29.1	0.04	80.0	48.8	64.3	0.172	73.3
4th	75–100	3.7	58.3	200	33.6	4.5	0.06	60.0	48.7	67.8	0.183	71.4
5th	100–132	3.9	50.0	218	35.7	−2.8	0.06	60.0	49.5	39.3	0.191	70.0

[a] TQ, total adsorbed quantity; for other abbreviations see Tables 1 and 2.

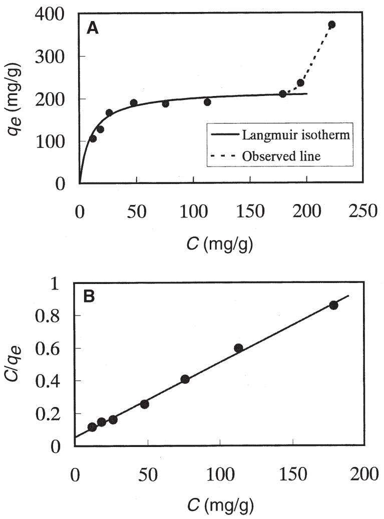

FIG. 1. (A) Adsorption isotherm of total polar materials (TPM) on silica gel. (B) Linearized Langmuir isotherm in TPM on silica gel. Abbreviations: q_e, the amount of TPM adsorbed per unit weight of adsorbent; C, TPM concentration of equilibrium.

where C_u is the content of each component in UO-I, C_n is the content of processed oils using n times reactivated silica gel, and C_0 that in silica gel as intially supplied. The relationship between the number of reactivations and the reactivation ratio is shown in Figure. 2.

The capacity of reactivated adsorbent for each component decreased with repetition of the reactivation in the initial steps up to the tenth reactivation. This phenomenon may be due to fusion-plugging of micropores on the silica gel by the reactivation. It is known that the fusion-plugging of micropores occurs at 320°C on silica or alumina particles (15). After the tenth reactivation, the adsorbent capacities for TPM remained almost constant at 50 to 54%, for TOTOX at 60 to 79%, for viscosity at 62 to 63%, for absorbance at 76 to 83%, and for FFA at 57 to 63%. Therefore, the reactivation system for recycling adsorbed silica gel was useful to some extent, although the adsorption capacity decreased by 20 to 50%.

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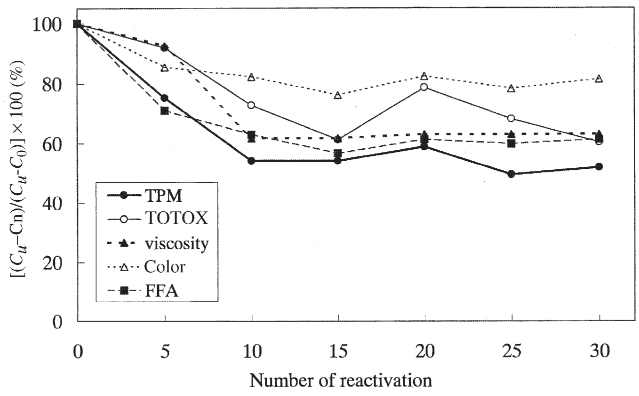

FIG. 2. Adsorption capacity changes vs. number of reactivations of silica gel. C_u, contents of each component; C_n, content of processed oil using n times reactivated silica gel; C_0, initial content of component in silica gel as initially supplied.

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