Thermal Deterioration of Diacylglycerol and Triacylglycerol Oils During Deep-Frying

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ABSTRACT: The thermal deterioration of cooking oil during deep-frying with a diacylglycerol (DAG)-rich oil (DAG-OIL) was compared with that for a cooking oil composed of a blend of commercial cooking oils (TAG-OIL) with a comparable FA composition and tocopherol content. Analyses of several indices of deterioration indicated no substantial difference in *p*-anisidine values, iodine values, and petroleum ether-insoluble oxidized FA between DAG-OIL and TAG-OIL. The polymerized glyceride (PG) content was lower for DAG-OIL than TAG-OIL. However, the PG value did not reflect the degree of polymerization of the FA chains directly, since both DAG-OIL and TAG-OIL generated polymeric products but of different types. An analysis of the polymerized FA content revealed no significant difference in the degree of polymerization of either of the oils. The total polar compounds included nonaltered DAG as an altered compound, and, as a result, this index was not appropriate for DAG-OIL. DAG-OIL underwent hydrolysis more rapidly than TAG-OIL. This difference was mainly correlated with moisture contained by the oil during frying and with the total molarity of the glycerides. Even though DAG-OIL was used until it became a waste oil, the extent of thermal oxidation was the same as that for TAG-OIL, although some indices showed a different trend from TAG-OIL. Molecular structure had no influence on the thermal deterioration of the frying oil. We conclude that the choice of indices is an important factor when the deterioration of DAG-OIL is evaluated.

Paper no. J10755 in *JAOCS 81,* 571–576 (June 2004).

KEY WORDS: Deep-frying, hydrolysis, molecular structure, oxidation, thermal deterioration.

Diacylglycerol (DAG)-rich cooking oil (DAG-OIL) has the unique property of repressing the accumulation of body fat (1–4), and is marketed as an edible oil for home use in the United States and Japan (5). When heated, cooking oil deteriorates, and numerous studies have appeared on the deterioration of general cooking oil, triacylglycerol (TAG)-rich oil (TAG-OIL), during deep-frying (6,7). The deterioration of TAG-OIL during deep-frying is caused by the thermal oxidation of FA chains, and a variety of undesirable compounds are generated, such as polar compounds, polymerized glycerides (PG), and oxidized FA (OxFA). When frying oil is used appropriately, these undesirable compounds are generated in only small quantities, and safety is maintained (8). However, under severe conditions, frying oil may develop toxicity owing to the accumulation of these undesirable compounds (9,10). To use frying oil safely, guidelines for good frying have been established in many countries (11,12). Oil that does not meet the guidelines specified should not be used. However, little information is available concerning the deterioration of DAG-OIL. The autoxidation of DAG-OIL at ordinary temperatures (13), deterioration indices following high-temperature heating (13), sensory evaluations for various cuisines (14), and deterioration indices during deep-frying (14,15) have been compared with TAG-OIL. These papers reported no differences in most of the deterioration indices and sensory evaluations and that the acid value (AV) of DAG-OIL was higher than that of TAG-OIL. However, no detailed reports have been prepared on the deterioration of DAG compared with TAG based on differences in their molecular structures. The molecular structure of DAG is different from that of TAG in that DAG contains a free hydroxyl group. This structure results in differences in the physical and chemical properties of DAG compared with TAG. Therefore, DAG-OIL may differ from TAG-OIL not only with respect to thermal deterioration during cooking but also in the meaning of the deterioration indices. Hence, an investigation of the deterioration of DAG-OIL during frying and a subsequent evaluation of the findings are important to use DAG-OIL safely as a frying oil. The influence of molecular structures on thermal oxidation was investigated by means of a detailed evaluation of DAG-OIL during deep-frying tests of sliced potatoes. To confirm the influence of molecular structure, DAG-OIL and TAG-OIL with the same FA and antioxidant compositions were used.

MATERIALS AND METHODS

Materials. DAG-OIL was prepared from soybean and rapeseed FA. After adjusting the saturated FA content by fractionation, an esterification reaction with glycerol was carried out under reduced pressure using Lipozyme RM IM (Novozymes, Chiba, Japan), which is a 1,3-position-selective immobilized enzyme. FA and MAG were removed by short-path distillation. The residue was then steam-deodorized. To match the FA composition of TAG-OIL with that of DAG-OIL, TAG-OIL was prepared by mixing three types of oil. Nine parts rapeseed oil and one part perilla seed oil were mixed with 10 parts of safflower oil. The deodorized DAG-OIL contained relatively high levels of δ- and γ-tocopherols, the origins of which were soybean and rapeseed FA. On the other hand, the

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origin of the mixed TAG-OIL was mainly safflower oil, and, as a result, it contained α-tocopherol instead of the other tocopherols. Hence, to match the antioxidant composition, 500 ppm of α-tocopherol (E Oil 805; Riken Vitamin Co. Ltd., Tokyo, Japan) was added to the deodorized DAG-OIL, and 0.1% mixed tocopherols (E Oil 600; Riken Vitamin Co. Ltd.) was added to the mixed oil. Table 1 shows the composition of the prepared test oil. The ratio of 1,2-DAG to 1,3-DAG in the deodorized DAG-OIL suggests that isomerization reached equilibrium (1 to 2) as a result of the heat used in the deodorization process.

Frying performance. Deep-frying tests were performed under two sets of conditions with a domestic electric fryer (oil pan 150 × 200 mm). The first did not involve fresh oil replenishment (FR), a procedure that can cause oil to deteriorate in a short time; the other included FR, a process that simulates commercial applications. One kilogram of test oil was placed in each fryer. Because some raw foodstuffs can contribute additional oil, non-prefried frozen sliced potatoes (5 mm thick, Royal Chefs sliced potatoes; Foods Supply International Co. Ltd., Tokyo, Japan) were used. The temperature was set to 180°C, and deep-frying tests were performed over an 8-h period. The frying time per batch was 3 min, and eight batches were processed in 1 h. The oil temperature varied in the range of 150 to 185°C during the frying, and the average temperature of both oils was 170°C. In the test performed without FR, the amount of oil decreased to about half at the end of the test period owing to absorption by the potatoes. To adjust for this decrease in oil, the amount of potatoes in each batch was reduced by about 10%. Based on the result of a preliminary experiment, in each batch, 100 g of potatoes was fried for 2 h from the beginning, 80 g for the next 2 h (2 to 4 h), 70 g for the next 2 h (4 to 6 h), and 60 g for the next 2 h (6 to 8 h). In the test performed with FR, the amount of potatoes in each batch was maintained at 100 g throughout the test period. Some of the oil was discarded from the fryer every hour, and 100 g of fresh oil was added (turnover 10 h). In both tests, about 10 g of oil was withdrawn every 2 h and stored at –20°C until used for analysis.

TABLE 1
Glyceride Composition (%), FA Composition (%), and Tocopherol Content (%) of Test Oils[a]

	DAG-OIL	TAG-OIL
Glyceride composition (%)		
MAG	1.2	0.0
DAG	81.4	1.7
1,2-DAG	28.0	0.7
1,3-DAG	53.4	1.0
TAG	17.3	98.3
FA composition (%)		
16:0	3.1	5.7
18:0	1.2	2.1
18:1	39.8	35.2
18:2	47.1	48.2
18:3	8.4	9.1
20:0	0.3	0.5
20:1	0.2	0.3
Tocopherol content (%)		
α-	0.031	0.032
β-	0.003	0.003
γ-	0.050	0.044
δ-	0.011	0.015
Total	0.096	0.094

[a]DAG-OIL, diacylglycerol (DAG)-rich oil; TAG-OIL, triacylglycerol (TAG)-rich oil; MAG, monoacylglycerol.

Analytical procedures. AOCS methods (16) were used to determine the AV (Cd 3d-63), *p*-anisidine value (AnV) (Cd 18-90), PG (Cd 22-91), and total polar compounds (TPC) (Cd 20-91). The polymerized FA content (PFA) was determined by analyzing the FAME prepared by the boron trifluoride method (AOCS Ce 2-66) with high-performance size exclusion chromatography (HPSEC) under the same conditions as the PG analysis (Cd 22-91). Glycerol was measured with a Hewlett-Packard 6890 gas–liquid chromatograph (GLC) with an FID by using the water phase of the by-product of prepared FAME. The instrument was fitted with a split/splitless injector (a split ratio of 50:1 was used) and equipped with a nonpolar capillary column with a liquid phase of dimethylpolysiloxane (0.25 mm i.d. × 10 m length, film thickness 0.1 μm). The oven temperature was programmed to increase from 80 to 340°C at 10°C per min, and then held at 340°C for 15 min. Helium was the carrier gas (flow rate 1 mL per min). After neutralization of the water phase with an aqueous solution of potassium hydroxide, the solution was evaporated under a nitrogen stream; the dried matter, containing 5–10 mg of glycerol, was silylated with 0.5 mL of silylating agent (Kanto Chemical Co. Ltd., Tokyo, Japan); and the derivatives were extracted into 1.5 mL of hexane. One microliter of extract was injected into the GLC. The glyceride composition was analyzed by GLC under the same conditions as were used for the glycerol analysis. An aliquot (15 mg) of the sample was silylated in the same manner. Derivatives were extracted with 1.5 mL of hexane, after which 1 μL of extract was injected into the GLC. The theoretical iodine value (IV) was calculated from the FA composition using AOCS Method Ce 1f-96. Petroleum ether-insoluble OxFA were determined according to DAGF Method C-III 3a (17). In addition, the oxidation stability index (OSI) (AOCS Method Cd 12b-92) of the fresh oils at 120°C was measured using a Rancimat test (Model 679; Metrohm Shibata Co. Ltd., Tokyo, Japan). Moisture in the oil during frying was measured using a Karl Fischer coulometric titration moisture meter (Model 831; Metrohm Shibata Co. Ltd., Tokyo, Japan). The oils were placed in screw-capped glass vials, plugged immediately with a Teflon seal to avoid the evaporation of water, cooled to room temperature, and then used in a moisture analysis.

Statistical analysis. Data from the different oils were compared by two-way ANOVA.

Sensory evaluation. A single-blind test was conducted involving five people, aged 20–40 yr, who were experienced in sensory evaluation.

RESULTS AND DISCCUSION

The OSI at 120°C for DAG-OIL and TAG-OIL was 3.2 and 2.9 h, respectively. No difference was found in autoxidation stability. In addition, no differences in moisture evaporation from food or oil absorption by food were found between DAG-OIL and TAG-OIL throughout the test period. Furthermore, although the taste of the fried potatoes gradually deteriorated with frying time, no difference was found between DAG-OIL and TAG-OIL. Thus, DAG-OIL can be used for deep-frying in the same manner as TAG-OIL.

Indices for DAG-OIL showing thermal oxidation were compared with those of TAG-OIL. Figure 1A shows time-course changes in AnV values, which increased with frying time. The AnV for DAG-OIL was slightly lower than that of TAG-OIL. The FA content per unit weight was about 5% lower in DAG-OIL than TAG-OIL because of its molecular structure. The degree of oxidation of the FA chains of the two oils was considered to be the same by taking account of the FA content. FR suppressed the changes in AnV for both oils to the same extent. Figure 1B shows time-course changes in IV. Although IV gradually decreased with frying time, there was little change, and the rate of decrease for DAG-OIL was equal to that for TAG-OIL. Figure 2A shows time-course changes in PG. The fraction with a higher M.W. than TAG was included as PG. PG increased with frying time. In the absence of FR, the PG in both oils at the end of the test period exceeded 10%. These oils correspond to the guidelines for waste oil (Belgium, <10%; the Czech Republic, <10%; The Netherlands, <16%; European Federation for the Science and Technology of Lipids, <12%). The PG in DAG-OIL was lower than that in TAG-OIL. However, PG values did not directly reflect the degree of polymerization of the FA chains that were oxidized, because both DAG and TAG generate polymers, and the resulting polymers are different. DAG-OIL produces mainly a DAG polymer, whereas TAG-OIL generates mainly a TAG polymer. If the FA content and degree of polymerization of DAG-OIL had been the same as TAG-OIL, the PG in DAG-OIL would have been lower than that for TAG-OIL. To understand the actual degree of polymerization of FA, it is necessary to measure PFA after taking the influence of molecular structure into account—in other words, after saponification. Figure 2B shows time-course changes in PFA. There was no significant difference ($P > 0.05$) in the formation of PFA in either of the oils. The origin of the difference in PG was the molecular structure. Thus, there was clearly no difference in the degree of polymerization of FA between DAG-OIL and TAG-OIL. TPC in fresh and waste oils were measured in the absence of FR. As shown in Figure 3A, the TPC in DAG-OIL were extremely high in the case of fresh oil. The reason for this is that nonaltered DAG was measured as TPC, and this result did not take the deterioration of DAG-OIL into account. The analysis of TPC in both fresh and waste DAG-OIL with HPSEC revealed that a large part of the TPC in the DAG-OIL were DAG (fresh oil, 95%; waste oil, 85%). Although TPC are widely used as an index of quality control for frying oil, it is not appropriate for the evaluation of DAG-OIL. OxFA were measured in fresh and waste oil in the absence of FR, as shown in Figure 3B. Because a correlation was previously reported between OxFA and TPC (18), OxFA can be used as an alternative index of TPC. Molecular structure does not influence OxFA, because they are measured as the high-polarity fraction of saponified oils. Therefore, OxFA can be used as a suitable index for evaluating the degradation of DAG-OIL for which TPC cannot be applied. The amount of OxFA for DAG-OIL (0.87 wt%) was slightly lower than that for TAG-OIL (0.99 wt%). However, the difference in the oxidation level of FA was much smaller when the difference in FA content of DAG-OIL

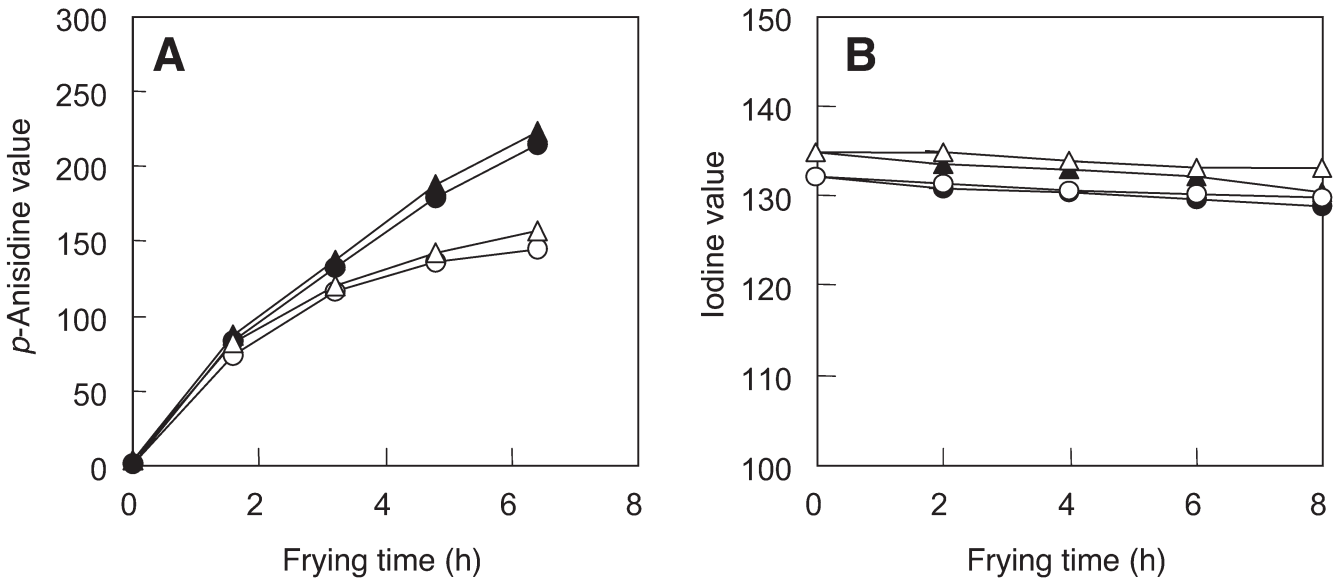

FIG. 1. Time-course changes in (A) the *p*-anisidine value and (B) the iodine value of (●) diacylglycerol (DAG)-rich oil (DAG-OIL) without fresh oil replenishment (FR); (▲) triacylglycerol (TAG)-rich oil (TAG-OIL) without of FR; (○) DAG-OIL with FR; and (△) TAG-OIL with FR.

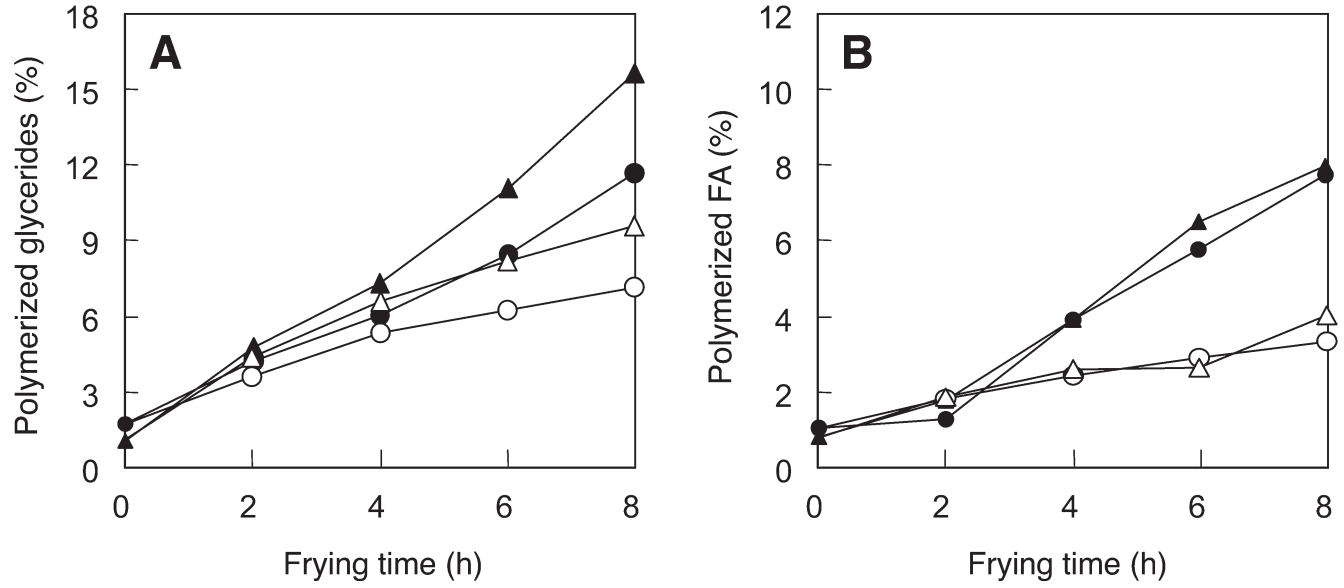

FIG. 2. Time-course changes in (A) polymerized glycerides (%) and (B) polymerized FA (%) in (●) DAG-OIL without FR; (▲) TAG-OIL without FR; (○) DAG-OIL with FR; and (△) TAG-OIL with FR. For abbreviations see Figure 1.

(91 wt%) and TAG-OIL (96 wt%) was taken into account. This result indicates that there was no difference in the oxidation of FA chains between DAG-OIL and TAG-OIL. In addition, an analysis of glycerol prepared by saponification resulted in no generation of polyglycerol in any of the samples. Thus, the glycerol backbone was stable over the long-term heating period.

The hydrolysis of DAG-OIL was next compared with that of TAG-OIL. Figure 4, showing the time-course changes in AV, illustrates that the AV increased with frying time. Although the AV for DAG-OIL increased more rapidly than that of TAG-OIL, the AV at the end of the test period in the absence of FR was lower than established guidelines (Japan, AV < 2.5; Belgium, FFA < 2.5%; Germany, AV < 2.0) and is not a justification for discarding the oil. FR reduced the rate of formation of FFA, although the hydrolysis reaction led to an increase in AV. The hydrolysis rates for both oils in the absence of FR were calculated by the least squares method from the data in Figure 4 and are shown in Table 2. The rate of hydrolysis of DAG-OIL was about 2.4 times that of TAG-OIL. We conclude that hydrolysis was influenced by the moisture in the oil and by the molarity of the glyceride, analogous to the alcoholysis given by a second-order reaction between alcohol and glyceride (19). The overall hydrolysis reaction is

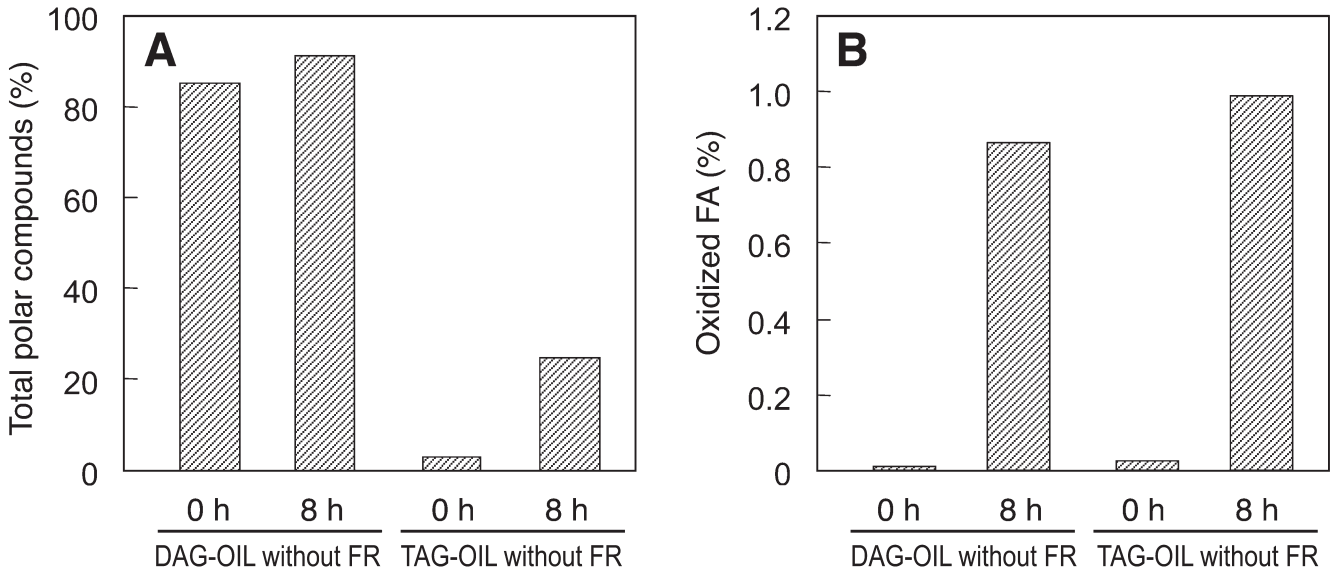

FIG. 3. (A) Total polar compounds (%) in DAG-OIL and TAG-OIL in the absence of FR. (B) Petroleum ether-insoluble oxidized FA (%) of DAG-OIL and TAG-OIL in the absence of FR. For abbreviations see Figure 1.

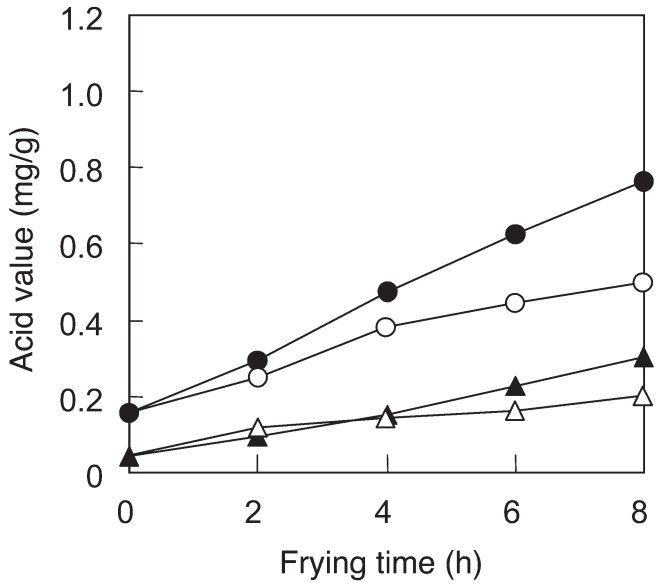

FIG. 4. Time-course changes in the acid value (mg/g) of (●) DAG-OIL without FR; (▲) TAG-OIL without FR; (○) DAG-OIL with FR; and (△) TAG-OIL with FR. For abbreviations see Figure 1.

TABLE 2
Rate of Hydrolysis, Moisture Content, and Total Glyceride Level of Each Oil[a]

	Rate of hydrolysis (mol/kg/s)	Moisture content (mol/kg)	Total glyceride level (mol/kg)
DAG-OIL	0.382	0.153	1.55
TAG-OIL	0.162	0.0869	1.15
Ratio (DAG/TAG)	2.36	1.76	1.35

[a]For abbreviations see Table 1.

shown in Equation 1. Assuming that the reaction rate was not dependent on the molecular structure, that rate is given by Equation 2.

$$\text{glycerides} + H_2O \rightarrow \text{FFA} + \text{glycerides or glycerol} \quad [1]$$

$$d[\text{FFA}]/dt = k[\text{glycerides}][H_2O] \quad [2]$$

where $d[\text{FFA}]/dt$ is the increasing rate of FFA, k the rate constant of hydrolysis, [glycerides] is the total molarity of all glycerides (sum of MAG, DAG, and TAG), and $[H_2O]$ is the moisture content of the oil. Moisture in the oil during frying and the total molarity of all glycerides calculated from the glyceride composition are shown in Table 2. Since DAG contains a hydroxyl group, its affinity for water molecules is higher than that of TAG. The moisture in DAG-OIL during frying was 1.8 times that of TAG-OIL. The total glyceride molarity of DAG-OIL was 1.4 times that of TAG-OIL because of the lower M.W. of DAG compared with TAG. However, the ratio of these two factors, the moisture content and the glyceride molarity, of DAG-OIL to TAG-OIL, which accelerate the reaction, was smaller than the ratio of the hydrolysis rates; the product of the ratio of each factor was equal to the ratio of the hydrolysis rate. This suggests that, based on the above supposition, the rapid increase in the AV of DAG-OIL can be attributed to a higher moisture content and the total glyceride molarity of the oil. The hydrolysis of DAG-OIL was not related to thermal deterioration.

The time-course changes in DAG content for DAG-OIL are shown in Figure 5. The DAG content dropped slightly with deep-frying over a long period of time. However, the ratio of 1,2-DAG and 1,3-DAG (1 to 2) did not change during the test period. Hydrolysis and randomization of the ester distribution by transesterification between glyceride molecules represent the most likely cause of this result. However, only a slight change in DAG was observed during the test period. Thus, one can conclude that its nutritive properties did not change during the test period of deep-frying.

Nakatsugawa and coworkers (13) reported that DAG-OIL was more stable to autoxidation than TAG-OIL. They proposed that the hydroxyl group of the DAG-OIL acts as an antioxidant, similar to a sugar alcohol, or as a donor for the chelation of trace metals. However, these antioxidant effects were not confirmed under deep-frying conditions. The above results suggest that even if DAG-OIL were used for an extended time until it became a waste oil, no differences in thermal oxidation would be evident between DAG-OIL and TAG-OIL. Finally, the findings herein confirm that the thermal degradation of oil was caused by the thermal oxidation of FA chains and was not influenced by the molecular structure. The increase in AV was faster for DAG-OIL than for TAG-OIL, but a tendency for FFA to promote oxidation (20) was not confirmed. In addition, although AV is an indirect but useful measure of the deterioration of oil for TAG-OIL (21), the thermal degradation of DAG-OIL

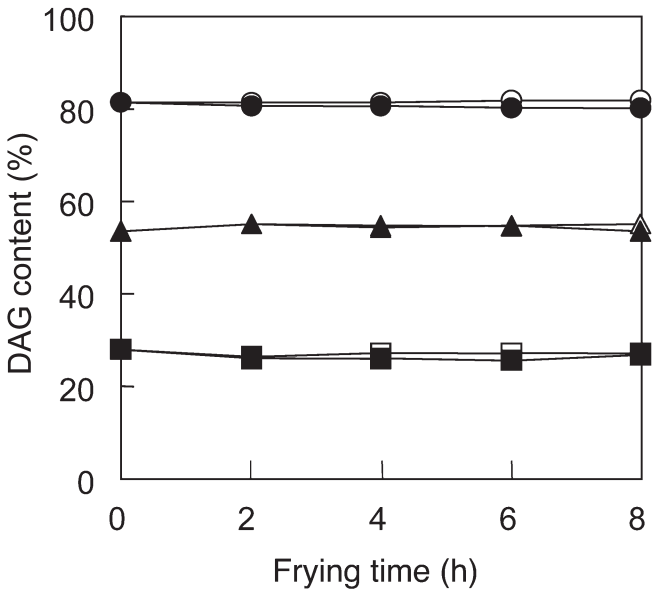

FIG. 5. Time-course changes in the total DAG content (%) of (●) DAG-OIL without FR; (○) DAG-OIL with FR; 1,3-DAG content (%) of (▲) DAG-OIL without FR; (△) DAG-OIL with FR; 1,2-DAG content (%) of (■) DAG-OIL without FR; and (□) DAG-OIL with FR.

should not be evaluated using AV, because the AV of DAG-OIL was higher than that of TAG-OIL, even when the oxidation levels of both oils were the same. Similarly, PG cannot be used as a degradation index in the case of DAG-OIL, because even with the same degree of polymerization of FA, the PG value was lower for DAG-OIL than for TAG-OIL. Furthermore, TPC is not appropriate for evaluating DAG-OIL, as this measurement included nonaltered DAG as an altered component. Although we confirmed that there was no difference in thermal deterioration between DAG-OIL and TAG-OIL during deep-frying, the choice of indices is important when evaluating DAG-OIL.

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[Received November 20, 2003; accepted March 30, 2004]