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RESEARCH ARTICLE

Effects of Catechin and α-Tocopherol Addition on the Autoxidative Stability of Diacylglycerol Oil Derived from an Olive Oil and Perilla Oil Mixture

Leejin Jung, Edwald Lee, and Eunok Choe

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Abstract The effects of tea catechin and α-tocopherol addition (1 mM) on the oxidative stability of oleic and linolenic acid-rich diacylglycerol (DAG) oil derived from an extra virgin olive oil and perilla oil mixture (6:4, w/w) were evaluated. Oil was oxidized at 50°C for 10 days, after which oxidation was evaluated based on headspace oxygen consumption and peroxide values (POV). The polyphenol and tocopherol contents were also monitored. Addition of catechin did not affect the oxygen consumption or POV of DAG oil, whereas α-tocopherol acted as a prooxidant. Addition of antioxidants had no significant (p>0.05) effect on the fatty acid composition of the oil. Degradation of γ-tocopherol during oil oxidation was inhibited by addition of α-tocopherol, and addition of antioxidants inhibited polyphenol degradation. The autoxidative stability of DAG oil can be improved using polar rather than non-polar antioxidants.

Keywords: oxidation, diacylglycerol oil, α-tocopherol, catechin

Introduction

Diacylglycerol (DAG) is an acylglycerol consisting of 2 fatty acid chains covalently bonded to a glycerol molecule through ester linkages, and is present in edible oil as a minor component. Olive oil has a higher DAG content (up to 6%) than other plant oils (1). DAG is known to increase bulk oil oxidation by reducing the surface tension of oil and accelerating oxygen diffusion (2). However, ingestion of DAG has been shown to lower the concentration of triacylglycerol (TAG) in the chylomicron fraction, compared to ingestion of TAG, due to faster oxidation (3). In addition, hydrolysis and reassembly of DAG to TAG are not efficient, which reduces the amount of lipids stored in adipose tissue (4-6). Thus, DAG oil in which DAG is a major component is promising as a functional oil.

The health functionality of DAG oil can be improved by modifying the fatty acid composition to include health-useful unsaturated fatty acids and an improved oxidative stability. DAG oil containing high contents of oleic and linolenic acids (>50%) has been used, but the photo-oxidative stability is not as good as the stability of TAG oil due to loss of antioxidants during manufacture (7). Antioxidants are commonly used to improve the oxidative stability of oil, and the efficiency of inhibiting oil oxidation is dependent on the characteristics of antioxidants (8) and the food matrix (9,10). Since DAG, with a small polar head group, tends to form inverted micellar structures, thereby exposing small areas of non-polar regions of neighboring molecules (11), DAG forms a matrix that is different from TAG. Thus, DAG oil containing a high content of DAG may interact differently with antioxidants of different polarities.

The oxidative stability of DAG oil has been reported to be lower than the stability of TAG oil during autoxidation (12) and photo-oxidation (7) due to a high temperature manufacturing process and low levels of antioxidants. On the other hand, Nakatsugawa *et al.* (13) and Ohno (14) reported a higher oxidative stability of DAG oil in autoxidation and thermal oxidation. Although improvement of the auto or photo-oxidative stability of DAG oil has been suggested for application to the food industry, the effects of antioxidant addition on DAG oil have not been

Leejin Jung, Edwald Lee, Eunok Choe (✉)
Department of Food and Nutrition, Inha University, Incheon 402-751, Korea
Tel: +82-32-860-8125; Fax: +82-32-862-8120
E-mail: eochoe@inha.ac.kr

extensively reported. This study evaluated the effects of α-tocopherol and polar catechin addition on the autoxidative stability of oleic and linolenic acid-rich DAG oil, and the stability of antioxidants was also determined during DAG oil oxidation.

Materials and Methods

Materials and chemicals DAG oil was derived from extra virgin olive oil (CJ Co., Seoul, Korea) and perilla oil which was obtained by expressing perilla seeds roasted at 180°C for 20 min. The weight ratio of extra virgin olive oil to perilla oil was 6:4, and DAG oil was manufactured according to the method of Kim and Choe (7). Briefly, the oil mixture was mixed with the same weight of water, followed by hydrolysis using Lipozyme TLIM (10%; Novozymes Corp., Bagsvaerd, Denmark) at 55°C and centrifugation at 1,480×*g* (H-500R; Kokusan, Tokyo, Japan). Lipozyme RMIM (10%; Novozymes Corp.) was added for re-esterification at 55°C, and DAG oil was obtained by subsequent centrifugation (Avanti J; Beckman, Fullerton, CA, USA) at 12,100×*g* and molecular distillation at 200°C and 0.005 mbar with a short path distillation mini pilot plant (KD-6; UIC GmbH, Alzenau, Germany). The lipid composition of the obtained DAG oil was determined using TLC with Kieselgel 60 F254 plates (Merck, Darmstadt, Germany) and a mixture of hexane, diethyl ether, and acetic acid (50:50:1, v/v/v) as a developing solvent (7). Quantification was performed using densitometry with an imaging densitometer (model GS-700; Bio-Rad, Hercules, CA, USA) after visualization using iodine vapor, then 5% sulfuric acid, followed by heating at 200°C for 2 min.

HPLC grade n-hexane, isopropanol, ethylacetate, methanol, and water were purchased from J.T. Baker (Philipsburg, NJ, USA). Caffeic acid, α, γ, and δ-tocopherols, and Folin-Ciocalteau reagent were purchased from Sigma-Aldrich Corp. (St. Louis, MO, USA). Tea catechin (82% polyphenols based on HPLC) from green tea extracts was donated by Nongshim Co. (Seoul, Korea). All other chemicals were of analytical grade.

Sample preparation and oxidation Either tea catechin or α-tocopherol (1 mM) was added to DAG oil, which corresponded to the antioxidant content of the extra virgin olive and perilla oil mixture. The oil was distributed into 20 mL glass vials at 6 g each. The vials were tightly sealed using aluminum caps and rubber stoppers, and then stored in a 60°C incubator (model LBI-250; Daihan Labtech Co., Seoul, Korea) in the dark for 10 days. All samples were prepared in duplicate. Control samples did not receive tea catechin or α-tocopherol.

Analysis of the oil composition The fatty acid composition was analyzed using GC (7) with a Younglin M600D gas chromatograph (Younglin Co., Ltd., Anyang, Korea). An HP-Innowax column (30 m×0.53 mm, 1.0-μm thick; Agilent, Böblingen, Germany) and a flame ionization detector were used. Temperatures of the oven, injector, and detector were 200, 270, and 280°C, respectively. The helium flow rate was 10 mL/min, and the split ratio was 10:1.

The tocopherol contents of the oil were determined using HPLC (7) with a Younglin YL9100 HPLC equipped with an autosampler, a μ-porasil column (330×3.9 mm, 10 μm i.d.; Waters, Milford, MA, USA), and a fluorescence detector with an excitation wavelength of 290 nm and an emission wavelength of 330 nm. The mobile phase was 0.2% isopropanol in n-hexane (v/v) at 2.0 mL/min. Polyphenols in the oil, expressed as caffeic acid equivalents, were determined using a UV-visible spectrophotometer (HP 8453; Hewlett Packard, Wilmington, DE, USA) at 725 nm with Folin-Ciocalteau reagent (7). Other minor compounds, such as chlorophylls and carotenoids, which might originate from the extra virgin olive oil or the perilla oil, were determined using HPLC (7) with a Younglin SP 930D HPLC equipped with a symmetry C18 column (5.0 μm, 4.6×300 mm; Waters) and UV-Vis detector (ACME 930; Younglin Co., Ltd.) set at 438 nm, or a μ-porasil column (300×3.9 mm, 10 μm i.d.; Waters) and a UV-Vis detector set at 436 nm. The elution solvents for chlorophylls and carotenoids were a mixture of ethylacetate, methanol, and water (50:37.5:12.5, v/v/v), and a mixture of n-hexane and isopropanol (97:3, v/v), respectively.

Evaluation of oil oxidation Oxidation of DAG oil was evaluated based on peroxide values (POV) according to the AOCS method Cd 8-53 (15) and headspace oxygen consumption using a Younglin YL 6100 GC equipped with an autosampler, a stainless steel column packed with 80/100 mesh molecular sieve 13X (1.83 m×0.32 cm; Agilent Co.), and a thermal conductivity detector (7). Helium was used at 20 mL/min. Temperatures of the oven, injector, and detector were 35, 100, and 140°C, respectively. The conversion factor for the oxygen peak area in 1 mL of headspace gas to μmol of oxygen was 9.35, based on the oxygen content (20.95%) of the air (7).

Statistical analysis All measurements were replicated for all samples, which were prepared in duplicate, resulting in 4 values for each sample. Statistical analysis of data included Duncan's multiple range test using the SAS/PC (SAS 9.1; SAS Institute, Inc., Cary, NC, USA) and SPSS (SPSS 19.0; SPSS Korea, Seoul, Korea) software packages, with a significance level of 5%.

Results and Discussion

Characteristics of DAG oil DAG oil derived from a mixture of olive oil and perilla seed oil (6:4, w/w) consisted of DAG (70.61%) and, more specifically, of 1,3-DAG (44.79%) and 1,2-DAG (25.82%). The TAG content was 19.64% (Table 1). The free fatty acid and monoacylglycerol contents of DAG oil were 5.30 and 4.45%, respectively. Among fatty acids, oleic acid was the most abundant (53.44%), followed by linolenic acid (27.64%). The contents of linoleic, palmitic, and stearic acids were 9.85, 6.69, and 2.38%, respectively. DAG oil contained a small amount of antioxidants. Concentrations of tocopherols and polyphenols were 6.02 and 62.05 mg/kg, respectively. There were no carotenoids or chlorophylls detected in DAG oil using HPLC. Unrefined oils, such as extra virgin olive oil and perilla oil contain appreciable amounts of antioxidants, such as tocopherols and polyphenols, and pigments, such as chlorophylls and carotenoids (16). Thus, most of the natural antioxidants and pigments in the source oil were lost during DAG oil manufacturing, mostly during distillation at 200°C, suggesting that the oxidative stability of DAG oil should be artificially controlled, for example, via extraneous antioxidant addition.

Table 1. Chemical characteristics of diacylglycerol oil derived from a mixture of olive oil and perilla oil (6:4, w/w)

Characteristics		DAG oil
Lipid subclass composition (relative %)	Triacylglycerol	19.64±0.15
	Free fatty acid	5.30±0.73
	1.3-Diacylglycerol	44.79±1.25
	1.2-Diacylglycerol	25.82±0.77
	Monoacylglycerol	4.45±0.08
Fatty acid composition (relative %)	16:0	6.69±0.57
	18:0	2.38±0.16
	18:1	53.44±0.60
	18:2	9.85±0.01
	18:3	27.64±0.20
	U/S[1)]	10.03±0.49
	PUFA/S[2)]	4.13±0.16
Tocopherol (mg/kg)	α-	ND[3)]
	γ-	6.02±0.03
	δ-	ND
	Total	6.02±0.03
Polyphenols (mg/kg)		62.05±12.97

[1)]Content ratio of unsaturated fatty acids to saturated fatty acids
[2)]Content ratio of polyunsaturated fatty acids to saturated fatty acids
[3)]ND, not detected

Oxidative stability of DAG oil and effects of α-tocopherol and tea catechin addition The headspace oxygen contents and POV of DAG oil with and without added α-tocopherol and tea catechin are shown in Table 2. The headspace oxygen contents of oil decreased with time due to oxygen consumption during oil oxidation, whereas the POV increased due to production of hydroperoxides in the oil. There was no significant (p>0.05) difference in the headspace oxygen contents between DAG oil with added tea catechin or added α-tocopherol, and a control oil without added antioxidants. There was also no significant (p>0.05) difference in POV between oil with added tea catechin and a control oil. DAG oil with added α-tocopherol showed significantly (p<0.05) higher POV after 6 days of oxidation (p<0.05), compared with controls. Thus, tea catechin did not affect the oxygen consumption or peroxide production of DAG oil over 10 days, although α-tocopherol addition increased peroxide production, suggesting that 1 mM tea

Table 2. Effects of 1 mM antioxidants on headspace oxygen contents and peroxide values of diacylglycerol oil during oxidation in the dark at 25°C

Oxidation parameter	Oxidation time (day)	Antioxidant added		
		None (control)	Catechin	α-Tocopherol
Headspace oxygen content (μmol/mL)	0	8.65±0.05$^{a1)}$	8.65±0.05^{a}	8.65±0.05^{a}
	2	6.16±0.08^{bc}	6.48±0.02^{b}	5.90±0.66^{c}
	4	4.53±0.06^{e}	5.04±0.03^{d}	4.52±0.17^{e}
	6	3.61±0.01^{f}	3.75±0.02^{f}	3.57±0.06^{f}
	8	2.77±0.03^{fg}	3.01±0.21^{g}	2.96±0.06^{fg}
	10	2.58±0.02^{i}	2.55±0.02^{i}	2.61±0.06^{hi}
Peroxide values (meq/kg)	0	2.22±0.11^{i}	2.59±0.20^{i}	3.81±0.63^{gh}
	2	3.96±0.45^{g}	2.97±0.25^{hi}	4.13±0.11^{g}
	4	5.05±0.61^{ef}	4.43±0.07^{fg}	3.98±0.12^{g}
	6	5.30±0.62^{ef}	5.48±0.53^{e}	5.15±0.45^{ef}
	8	6.52±0.19^{d}	6.52±0.07^{d}	7.68±0.98^{c}
	10	9.80±0.01^{b}	9.74±0.38^{b}	16.50±0.02^{a}

[1)]Different superscripts indicate significantly different values within the same oxidation parameters based on Duncan's multiple range test (p<0.05).

Table 3. Fatty acid composition of diacylglycerol oil with and without added antioxidants during oxidation at 60°C in the dark

Antioxidant added	Oxidation time (day)	Fatty acid contents (relative % based on total fatty acid contents)				
		16:0	18:0	18:1	18:2	18:3
No (control)	0	8.240±0.876[a1)]	3.463±0.383[a]	54.120±5.543[ab]	9.499±0.958[a]	24.678±2.464[ab]
	2	8.082±0.0544[a]	3.363±0.0141[a]	54.101±1.2291[ab]	9.484±0.1894[a]	24.970±0.58143[ab]
	4	8.123±0.103[a]	3.399±0.039[a]	54.107±0.983[ab]	9.553±0.181[a]	24.818±0.543[ab]
	6	8.217±0.028[a]	3.440±0.028[a]	54.102±1.292[ab]	9.485±0.069[a]	24.756±0.667[ab]
	8	8.251±0.193[a]	3.521±0.142[a]	54.088±1.893[ab]	9.522±0.348[a]	24.616±0.734[ab]
	10	8.194±0.439[a]	3.468±0.232[a]	53.857±3.341[ab]	9.568±0.567[a]	24.912±1.471[ab]
Tea catechin	0	8.240±0.876[a]	3.463±0.383[a]	54.120±5.543[ab]	9.499±0.958[a]	24.678±2.464[ab]
	2	8.222±0.026[a]	3.474±0.026[a]	53.986±0.428[ab]	9.553±0.078[a]	24.765±0.169[ab]
	4	8.162±0.592[a]	3.435±0.283[a]	54.019±3.823[ab]	9.546±0.746[a]	24.838±1.982[ab]
	6	8.305±0.957[a]	3.496±0.493[a]	53.750±6.360[ab]	9.595±1.055[a]	24.854±2.618[ab]
	8	8.253±0.087[a]	3.482±0.037[a]	53.903±1.054[ab]	9.571±0.186[a]	24.790±0.471[ab]
	10	8.272±0.088[a]	3.542±0.063[a]	53.841±0.492[ab]	9.535±0.038[a]	24.811±0.076[ab]
α-Tocopherol	0	8.240±0.876[a]	3.463±0.383[a]	54.120±5.543[ab]	9.499±0.958[a]	24.678±2.464[ab]
	2	8.212±0.990[a]	3.469±0.432[a]	53.630±6.704[ab]	9.637±1.227[a]	25.052±3.024[ab]
	4	8.164±0.075[a]	3.470±0.000[a]	53.920±0.584[ab]	9.630±0.112[a]	24.816±0.236[ab]
	6	8.204±0.230[a]	3.492±0.109[a]	53.852±1.585[ab]	9.608±0.230[a]	24.843±0.617[ab]
	8	8.234±1.476[a]	3.521±0.623[a]	53.956±10.053[b]	9.579±1.693[a]	24.708±4.152[b]
	10	8.226±0.698[a]	3.486±0.308[a]	54.102±4.735[a]	9.442±0.544[a]	24.743±1.823[a]

[1)]Different superscripts indicate significantly different values within the same fatty acid based on Duncan's multiple range test ($p<0.05$).

catechin had no significant ($p>0.05$) effect on DAG oil oxidation, compared with controls, whereas 1 mM α-tocopherol acted as a prooxidant.

The prooxidative effect of α-tocopherol (1.25×10^{-5} M) on linoleic acid in aqueous media has been reported by Cillard *et al.* (17). α-Tocopherol is oxidized and produces tocopherol peroxy or oxy radicals during oil oxidation, which both increase the transfer of headspace oxygen into oil by reduction of surface tension (18). On the contrary, Ohno (19) reported delayed autoxidation of DAG oil (DAG content of 93.3%) by addition of a high concentration of tocopherols (approximately 1,800 mg/kg). Although addition of tea catechin did not significantly ($p>0.05$) improve the oxidative stability of DAG oil in this study, compared with controls, the oxidation level of oil with added catechin tended to be lower than levels of oil with added α-tocopherol, partly due to different polarities of the antioxidants. α-Tocopherol is soluble in TAG oil, but catechin dissolves more easily than α-tocopherol in more polar DAG oil. Although this indicates that catechin can act as an antioxidant, there were no significant ($p>0.05$) antioxidant effects with catechin addition in this study, compared with controls. Based on the effective concentrations (50-200 mg/kg) of tert-butylhydroquinone (TBHQ) in soybean DAG oil (DAG content of 78.63%) and palm DAG oil (DAG content of 74.49%) (12), along with the low antioxidant activities of green tea extracts (<4.6 h of induction time), compared with TBHQ (39.9 h of induction time) in chicken fat oxidation (20,21), 1 mM tea catechin was insufficient to improve the autoxidative stability of DAG oil.

The fatty acid composition of controls did not significantly ($p>0.05$) change during the 10 day oxidation period, and there was no significant ($p>0.05$) difference observed by addition of tea catechin and α-tocopherol (Table 3). Thus, oxidation for 10 days at 60°C did not affect the fatty acid composition of oleic and linolenic acid-rich DAG oil, indicating no effects for tea catechin or α-tocopherol addition on the oxidation rates of the different fatty acids in DAG oil.

Changes in tocopherol and polyphenol contents during DAG oil oxidation Changes in the contents of tocopherols and polyphenols in DAG oil with and without added antioxidants during 10 days of autoxidation at 60°C are shown in Table 4. The tocopherol content of DAG oil decreased during oxidation, indicating tocopherol degradation. Tocopherols are degraded by free radicals produced during oil oxidation (22). The γ-tocopherol initially present in DAG oil at a low level (approximately 6 mg/kg) was degraded at a rate constant of 0.072, 0.056, and 0.012/day in a control oil without antioxidant addition, and in oils with added tea catechin and α-tocopherol, respectively ($r^2>0.75$). Thus, degradation of γ-tocopherol was slower in DAG oil with added α-tocopherol than in oil with added tea catechin, or a control oil without antioxidants, indicating protection of γ-tocopherol from degradation by α-tocopherol during DAG oil oxidation. The capacity to donate phenolic hydrogen to free radicals is higher in α-tocopherol than in

Table 4. Tocopherol and polyphenol contents (mg/kg) of diacylglycerol oil with and without 1 mM added antioxidants and degradation rates (k) during 10 days of oxidation at 60°C in the dark

Antioxidants added	Oxidation time (day)/ kinetic parameter[1)]	Tocopherols			Polyphenols
		α	γ	Total	
None	0	ND[2)]	6.02±0.21[d3)]	6.02±0.21[e]	62.05±12.97[de]
	2	ND	4.84±0.22[e]	4.84±0.22[e]	51.99±13.05[efg]
	4	ND	3.12±0.07[i]	3.12±0.07[e]	46.28±1.91[efg]
	6	ND	3.02±0.03[i]	3.02±0.03[e]	47.11±1.11[efg]
	8	ND	2.86±0.22[i]	2.86±0.22[e]	ND
	10	ND	3.01±0.04[i]	3.01±0.04[e]	ND
	k (/day)	NA[4)]	0.072	0.072	1.571
	r^2		0.76	0.76	0.70
Tea catechin	0	ND	6.13±0.34[d]	6.13±0.34[e]	220.76±14.99[a]
	2	ND	4.36±0.31[f]	4.36±0.31[e]	114.34±1.70[b]
	4	ND	4.20±0.05[fd]	4.20±0.05[e]	113.62±2.62[b]
	6	ND	3.87±0.04[g]	3.87±0.04[e]	37.46±3.50[g]
	8	ND	3.50±0.13[h]	3.50±0.13[e]	8.28±1.45[h]
	10	ND	3.24±0.14[hi]	3.24±0.14[e]	7.22±0.52[h]
	k (/day)	NA	0.056	0.056	0.373
	r^2		0.88	0.88	0.93
α-Tocopherol	0	430.78±0.15[a]	8.68±0.15[a]	439.46±3.36[a]	74.63±7.97[cd]
	2	239.76±0.03[b]	8.57±0.03[ab]	248.33±0.17[b]	69.92±1.18[cd]
	4	85.17±0.17[c]	8.48±0.17[ab]	93.65±4.29[c]	70.62±5.54[cd]
	6	53.30±0.22[d]	8.23±0.22[b]	61.53±2.04[d]	59.91±9.23[def]
	8	49.76±0.04[d]	7.84±0.04[c]	57.60±0.30[d]	42.90±1.77[g]
	10	84.71±0.11[c]	7.74±0.11[c]	92.46±25.94[c]	45.32±2.39[fg]
	k (/day)	0.190	0.012	0.180	0.118
	r^2	0.67	0.94	0.68	0.84

[1)]Estimated based on regression assuming 1st-order kinetics, ln ($[A]/[A]_0$)= −k×time (day), where [A] and $[A]_0$ are the concentrations of antioxidants (mg/kg) at times t and 0, respectively. r^2=determination coefficient
[2)]ND, not detected
[3)]Different superscripts indicate significantly different values within the same kind of antioxidant based on Duncan's multiple range test (p<0.05).
[4)]NA, not applicable

γ or δ-tocopherol (23), which spared γ-tocopherol during oxidation of oil with added α-tocopherol.

Since DAG oil did not contain α-tocopherol, degradation of α-tocopherol was observed only in oil with added α-tocopherol at a rate constant of 0.190/day (r^2=0.67), which was higher than the rate for the γ-isomer, indicating reduced stability of α-tocopherol, compared to γ-tocopherol during DAG oil oxidation. α-Tocopherol has been reported to be less stable than γ-tocopherol during autoxidation of soybean oil at 40°C (24) and linoleic acid (25). Choe (26) also demonstrated a higher stability of γ-tocopherol than α-tocopherol during oxidation of sunflower oil due to a lower reactivity towards lipid and peroxy radicals. Oxidized α-tocopherol is a well-known prooxidant (18). Thus, DAG oil with added α-tocopherol showed an increased level of oil oxidation.

The polyphenol content of a sample with added tea catechin (220.76 mg/kg) was higher than the content of a control sample without antioxidant addition (62.05 mg/kg), or of a sample with added tocopherol (74.63 mg/kg). The polyphenol contents also decreased with time during oxidation of DAG oil. Degradation rates were 1.571, 0.373, and 0.118/day in DAG oil without antioxidant addition, and in oils with added tea catechin and added α-tocopherol, respectively. Thus, addition of either tea catechin or α-tocopherol slowed polyphenol degradation during DAG oil oxidation, possibly due to higher scavenging levels of free radicals due to the added antioxidants. The phloroglucinol moieties of polyphenol structures easily form hydrogen bonds with radicals produced by oil oxidation (27), resulting in degradation of polyphenols. However, added α-tocopherol and catechin donated phenolic hydrogens to the radicals, resulting in a decrease in reactivity between radicals and polyphenols and, eventually, more polyphenols in the oil. Degradation rates of polyphenols were higher than rates of tocopherols (Table 4), indicating a higher antioxidant activity for polyphenols than for tocopherols in DAG oil and corroborating the result of this study of a reduced

oxidation level in DAG oil with added tea catechin than in oil with added α-tocopherol. The antioxidant activity of polyphenols was reported to be much higher than the activity of α-tocopherol in olive oil (28).

In conclusion, 1 mM tea catechin had no effect on the autoxidative stability of oleic and linolenic acid-rich DAG oil. α-Tocopherol acted as a prooxidant, based on produced degradation products. The oxidative stability of DAG oil can be improved using polar rather than non-polar antioxidants.

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