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Green Tea Extract is an Effective Antioxidant for Retarding Rancidity of *Yukwa* (Rice Snacks) Fried in Soybean and Rice Bran Oils

Jae Hee Park and Chang Soon Kim†

Department of Food and Nutrition, Changwon National University, Kyungnam 641-773, Korea

Abstract

***Yukwa* is a popular Korean traditional fried rice snack. The high fat content and porous structure of *Yukwa* cause it to rapidly become rancid, presenting difficult challenges for commercial distribution. In this study, antioxidant activities of green tea extracts (GTE) were evaluated in *Yukwa* fried in soybean oil (SBO), rice bran oil (RBO) and winterized rice bran oil (WRBO) during storage at 40°C for 12 weeks. Lipid oxidation of *Yukwa* was determined by acid value (AV), peroxide value (POV), p-anisidine value (AnV), totox value and sensory evaluation. The addition of GTE to the oils reduced the increases in AV, POV, AnV, and totox. Totox increased most rapidly in *Yukwa* fried in SBO, followed by RBO > WRBO > SBO + 200 ppm GTE > RBO + 200 ppm GTE > WRBO + 200 ppm GTE ($p<0.05$). Sensory evaluation revealed that the addition of 200 ppm GTE delays rancidity in *Yukwa* by 7~8 weeks; providing compelling evidence that GTE is an effective antioxidant for *Yukwa*.**

Key words: lipid oxidation of *Yukwa*, green tea extract, antioxidant activity, frying vegetable oils, storage

INTRODUCTION

Lipid rancidity is often the limiting factor for shelf-life of high-fat foods such as deep-fried snack products. Rancid oils produce undesirable organoleptic qualities including rancid flavors, smells, and discolorations that are unacceptable to consumers, but also decrease nutritional quality and produce harmful lipid byproducts (1,2).

Improvements in food preparation, refrigeration, and packaging, all help to retard oxidation. These types of improvements, however, may not be sufficient to prevent oxidation, as a result, antioxidants are frequently incorporated into frying oils in an attempt to stabilize the deep-fat fried foods against oxidation, and thus prolong the shelf life of the product. Consumers prefer that natural rather than synthetic antioxidants be used in foods because of possible toxicity issues (3). Consequently, there has been an increased interest in developing natural antioxidants, especially those derived from herbs, spices and other plant materials (3-7).

Recently, many researchers have been interested in green tea because of its positive antioxidant effects in oils and fats; as well as various health benefits including antitumor, antiaging and antiplatelet aggregation properties (8,9). It has been reported that the antioxidant activity of green tea catechin is stronger than that of δ-tocopherol (10) and rosemary extract (11), which have been two of the most effective natural antioxidants in oils and fats.

Yukwa is a popular Korean fried snack made from glutinous rice. Korean manufacturers face many problems in distributing *Yukwa* due to its large amount of fat (15~20%) and porous structure, which exposes the fat to oxygen, leading to rapid lipid oxidation (12). The shelf-life of *Yukwa* depends upon the rate at which oxidation of lipids results in rancidity of the product. Previous studies of methods for extending the shelf life of *Yukwa* have used nitrogen flushing in the packaging (13,14). However, the fragile, porus structure of *Yukwa* led to incomplete degassing before nitrogen gas was flushed into the packages which, in turn, resulted in an increase in oxygen content during the storage period thereby defeating the effectiveness of nitrogen flushing and increasing oxidation and rancidity. Therefore, Park et al. (14) suggested that nitrogen flushing combined with an antioxidant additive (such as γ-orizanol) could be effective. There are lack of studies on the use of natural antioxidants for extending the shelf-life of *Yukwa*.

In this study, the effectiveness of GTE as an antioxidant in prolonging the shelf-life of *Yukwa* fried in soybean or rice bran oils was evaluated during storage at 40°C. The degree of oxidation was determined by chemical measurements including acid (AV), peroxide (POV), p-anisidine (AnV), and oxidation values (OV) and by sensory evaluation.

MATERIALS AND METHODS

Materials

Glutinous rice harvested in year 2000 was purchased from

†Corresponding author. E-mail: cskim@changwon.ac.kr
Phone: +82-55-279-7482, Fax: +82-55-281-7480

Hamyang Agricultural Cooperative. The three antioxidant free oils used in frying were: soybean oil (SBO) provided by Cheil Jedang, Co. (Korea), rice bran oil (RBO), and winterized rice bran oil (WRBO) provided by Sinyang Rice & Oil Co. (Korea). Corn syrup, brix 75%, was provided by Daesang Co. Green tea extract (GTE), standardized to 85% total phenols, was obtained from Healthland Supplies Ltd. (China). Total phenol content of the GTE was measured by Sugiura method (15). General reagents were purchased from Duksan Co. (Korea).

Fatty acid composition

Fatty acid contents of the fresh oils were determined by gas chromatography (Model 5890 series, Hewlett Packard, USA). The injector and flame ionization detector temperatures were 230°C and 240°C, respectively; and the column temperature was 250°C (SP 2330, SUPELCO, USA). Fatty acid concentrations were calculated from their relative percentages by the area of each peak.

Preparation and storage of *Yukwa*

Yukwa was prepared by the method of Lee et al. (16). The glutinous rice was washed and then steeped for 7 days at room temperature. Water was drained for 4 hrs and the rice ground with a roller mill (far-infrared ceramic roller, Samjung Machine Industry Co., Korea) and stored in a freezer until use. A mixture of 1 kg of glutinous rice powder and 3 kg water was steamed for 1 hr and then mixed with the dough wing of an electronic automatic rice cake machine (AFC-166, Matsushita Electric Trading Co., Ltd., Japan) for 30 min. After sprinkling wheat flour on the rice dough to reduce stickiness, the dough was cut into $0.5\times3\times1$ cm blocks and put into a far-infrared drying oven preheated to 50°C and dried to 17% moisture content. The dried *Yukwa* pellets were placed in a desiccator for aging (24 hrs) and fried in SBO, RBO or WRBO. The oils were in an emulsion state containing a mixture of GTE and ethanol, prepared according to the methods of Jung et al. (7) and Chen et al. (11). Two frying times and temperatures were used; the 1st frying time was 45 sec at 120°C, and the 2nd was 30 sec at 180°C. Twenty *Yukwa* pellets were fried in 1200 mL of oil at one time. The same frying oil was not used more than 20 times. The excess oil on the fried *Yukwa* base was drained for 1 hr, after which the *Yukwa* bases were dipped in 70°C corn syrup for 5 min and then coated with ground puffed rice. The finished *Yukwa* were wrapped in aluminum laminated film and stored in an incubator at 40°C for 12 weeks.

Extraction of oil and analysis of lipid oxidation of *Yukwa* during storage

Tests for lipid oxidation were carried out on the oil extracted from ground *Yukwa*. The oils in the *Yukwa* were extracted with ethylether, filtered, and the ethylether removed using a vacuum rotary evaporator (Eyela, N-N series, Tokyo Rikakikai Co). The AOCS official method (17) was used for the determination of fat content, acid (AV), peroxide (POV) and p-anisidine values (AnV). The OV or totox value was calculated as the AnV plus twice the POV (18). The formation of primary oxidation products was monitored by measuring the POV of oils (19). AnV measures aldehydes (secondary oxidation products) produced during lipid oxidation (20).

Sensory evaluation

A nine-member panel was selected from among departmental graduate students with experience in sensory evaluation to evaluate the rancidity of *Yukwa* during the storage period. The control *Yukwa* was stored in a freezer (-20 °C). Prior to evaluation, the *Yukwa* was ground. Panelists were provided with tepid water to cleanse the palate between samples. The test *Yukwa* samples were compared with controls for degree of rancid flavor using a 7 point scale (0, equal to control; 1, slight difference; 2, more distinct difference but still acceptable; 3, beginning to lose acceptability; 4, more distinct loss of acceptability; 5, very distinct loss of acceptability; 6, unacceptable) (21).

Statistical analysis

All data were analyzed by analysis of variance (ANOVA) and Duncan's Multiple Range Test using SPSS (Statistical Package, SPSS Inc.). Differences were considered significant at $p<0.05$, unless otherwise indicated (22).

RESULTS AND DISCUSSION

Antioxidant effect of different levels of GTE in *Yukwa* fried in SBO

During the 12 week storage period, antioxidant protection of GTE against lipid oxidation of *Yukwa* fried in SBO was evaluated. Synthetic antioxidants, such as butylated hydroxytoluene (BHT), butylated hydroxyanisole (BHA) and tertiary butyl hydroquinone (TBHQ) are permitted in foods at levels not to exceed FDA-and USDA-permitted levels, resulting in a maximum total antioxidant content of 200 ppm of the total fat or oil content; tocopherols are permitted in rendered animal fat or combinations of animal and vegetable fats at concentrations of 300 ppm (23). Therefore, we set the minimum level of antioxidants to be used in this experiment at 200 ppm. The concentrations of GTE evaluated were 200 ppm, 400 ppm and 600 ppm. Changes in AV are shown in Fig. 1. As the concentrations of GTE increased from 200 ppm to 600 ppm, the increase of AV was reduced in a dose dependent manner. After three weeks of storage, there was a sharp increase of AV in the control *Yukwa* fried in SBO, but containing no GTE; but AV was

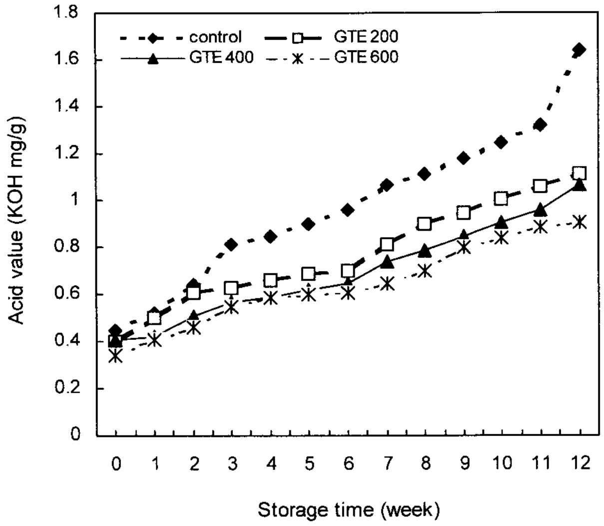

Fig. 1. Changes in acid value of *Yukwa* fried in soybean oil containing green tea extract (GTE) at various concentrations (0, 200, 400, 600 ppm) during storage at 40°C.

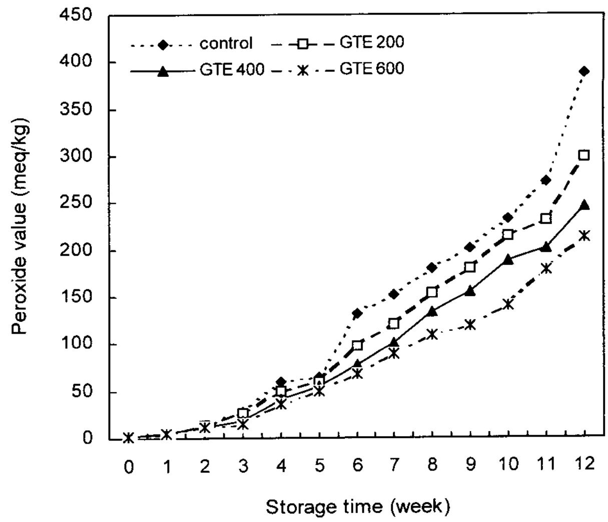

Fig. 2. Changes in peroxide value of *Yukwa* fried in soybean oil containing green tea extract (GTE) at various concentrations (0, 200, 400, 600 ppm) during storage at 40°C.

only slightly elevated in *Yukwa* with GTE. These results agree with those of Park et al. (24) who obtained lower AVs for soybean oil preserved with aqueous green tea than for control oil containing no aqueous green tea during storage at 60°C. Increases in AV are primarily due to hydrolysis of glyceride linkages by water present in *Yukwa* coated with corn syrup. Although the AV is an index of hydrolytic rancidity, it was nevertheless measured, as acids contribute to the development of off-flavor in the product. However, the lower AV of *Yukwa* with GTE, compared to those without GTE, cannot be ascribed to direct antioxidant action because phenolic compounds of GTE act by inhibiting oxidation reactions and have no direct effect on hydrolytic reactions.

POV, an indicator of the levels of primary oxidation products, was found to increase with storage regardless of treatments (Fig. 2), however, as the concentration of GTE increased from 200 to 600 ppm, the rate of increase in POVs was reduced. POV for *Yukwa* fried in SBO containing no GTE showed an overwhelming increase from the 6 week storage period onward. Peroxides have no flavor or odor, but break down rapidly to form aldehydes, which have a rancid flavor, thereby reducing the shelf-life of a food product. Hence, they are a commonly used indicator of rancidity.

AnVs, a measurement of secondary products of lipid oxidation, also increased with storage time in all samples (Fig. 3). AnV for *Yukwa* fried in SBO containing no GTE increased notably after 3 weeks. However, incorporation of GTE into SBO reduced AnV significantly.

Changes in OV are shown in Fig. 4. OV or totox value, which is defined as the sum of the AnV and two times the POV, is a measure of the total oxidation occurring in

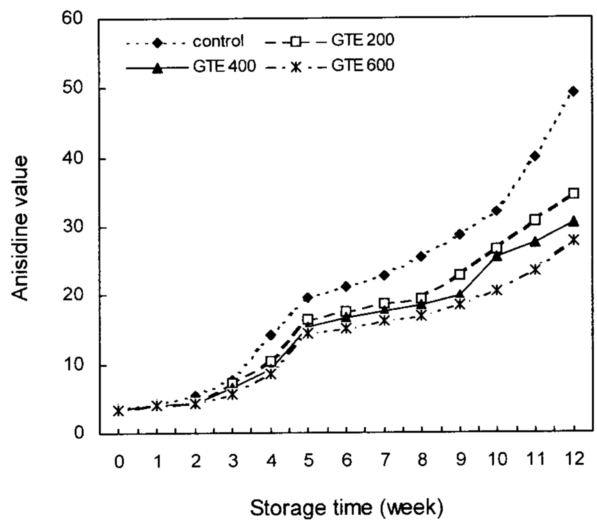

Fig. 3. Changes in p-anisidine value of *Yukwa* fried in soybean oil containing green tea extract (GTE) at various concentrations (0, 200, 400, 600 ppm) during storage at 40°C.

the oil (18). GTE at 600 ppm resulted in smallest increase of totox during storage, demonstrating a strong antioxidant effect. When *Yukwa* was fried in SBO treated with various concentrations of GTE, obvious differences of totox were obtained from the 6 week period onward. The incorporation of GTE into SBO reduced totox for *Yukwa* in a time dependent manner, demonstrating the effectiveness of GTE in retarding the lipid oxidation of *Yukwa*.

In the sensory evaluation of *Yukwa*, a score higher than 3 points was considered to indicate rancidity. Rancid flavors in control *Yukwa* was already detected after 3 weeks, but not detectable in *Yukwa* fried in SBO treated with 200 and 400 ppm GTE detected until after 10 weeks (Fig. 5). SBO treated with 600 ppm GTE never became rancid (not

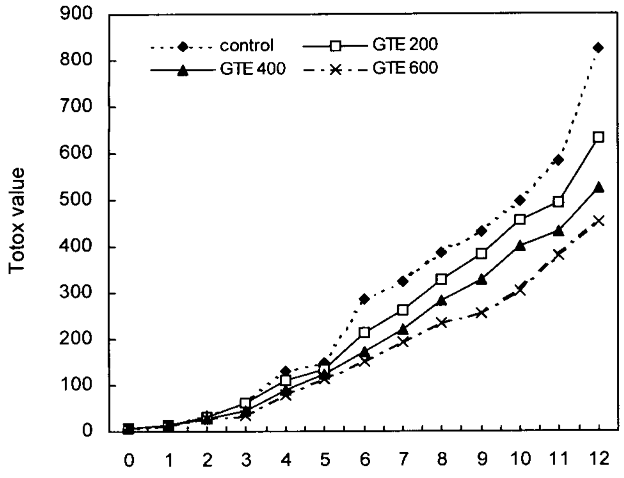

Fig. 4. Changes in totox value of *Yukwa* fried in soybean oil containing green tea extract (GTE) at various concentrations (0, 200, 400, 600 ppm) during storage at 40°C.

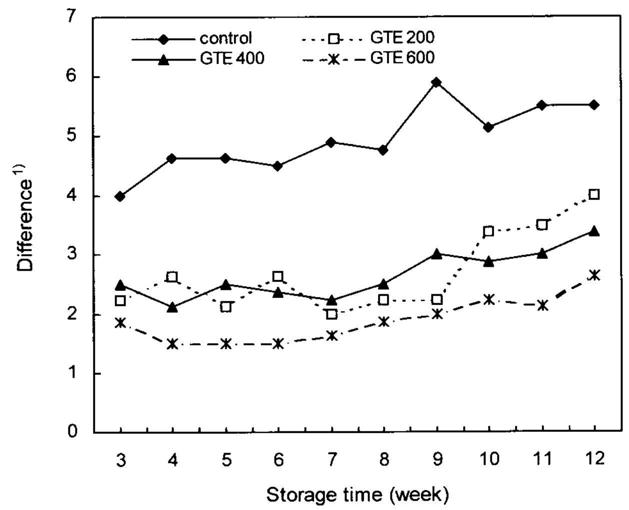

Fig. 5. Result of the difference from control test for rancid flavor of *Yukwa* fried in soybean oil containing GTE at various concentrations during storage at 40°C.

[1)]Means of difference score from control: 0, equal to control; 1, slight difference; 2, more distinct difference but still acceptable; 3, beginning to lose acceptability; 4, more distinct loss of acceptability; 5, very distinct loss of acceptability; 6, unacceptability.

exceeding 3 points) throughout the entire storage period. Therefore, the sensory evaluation data is in concordance with the chemical measurements in demonstrating that GTE is an effective antioxidant and is able to significantly retard the development of rancidity in *Yukwa*.

Antioxidant effect of GTE in *Yukwa* fried in different oils

The fatty acid compositions of fresh oils are shown by Table 1. RBO and WRBO had less unsaturated fatty acids, especially linoleic acid (18 : 2) and linolenic acid (18 : 3) than SBO. The mean fat contents of *Yukwa* fried in SBO, RBO and WRBO were 12.6, 12.3 and 12.6%, respectively.

Table 1. Fatty acid compositions of fresh oils (wt% of total fatty acids)

Fatty acid	SBO	RBO	WRBO
C 14:0	0.07	0.42	0.30
C 16:0	11.06	19.44	18.08
C 16:1	-	0.22	0.21
C 18:0	4.03	2.05	1.88
C 18:1	24.22	42.09	41.78
C 18:2	52.76	33.23	35.13
C 18:3	6.40	1.45	1.64
C 20:0	0.99	1.20	1.03
C 20:1	0.52	-	-
UR	5.20	3.33	3.70

Abbreviation: SBO = soybean oil; RBO = rice bran oil; WRBO = winterized rice bran oil; UR = unsaturated ratio.

AV for all *Yukwa* samples increased with storage time (Table 2). AVs for SBO, RBO and WRBO on the final day of the study (12th week) were 1.64, 1.52 and 1.48 KOH · mg/g, respectively, and were not significantly different. The incorporation of 200 ppm GTE into the three oils reduced AV increases, which did not exceed 2 KOH · mg/g after 12 weeks, an acceptable level for fried snack foods (25).

By the 4th week, the POV for *Yukwa* had already exceeded 40.0 meq/kg, sufficiently high to be unacceptable for fried snack foods (25), except for *Yukwa* fried in GTE incorporated WRBO (38.4 meq/kg). Shin et al. (26) reported that the shelf-life of *Yukwa* puffed in oil is less than 4 weeks at 30°C, which is in close agreement with our results. POV for *Yukwa* fried in all three oils containing no GTE showed an overwhelming increase from the 6 week storage period onward. The rate of increase in POV and AnV was highest for *Yukwa* fried in SBO followed by RBO > WRBO. Incorporation of GTE reduced the increases in both POV and AnV for *Yukwa* fried in the three oils, demonstrating the effectiveness of GTE in the prevention of primary and secondary oxidation products. Consequently, the incorporation of GTE reduced an increase of totox for *Yukwa* fried in all three oils. Totox increased most rapidly in *Yukwa* fried in SBO containing no GTE, followed by RBO > WRBO > SBO + GTE > RBO + GTE > WRBO + GTE ($p < 0.05$). Therefore, *Yukwa* fried in GTE incorporated WRBO had the most oxidative stability of all treatments.

Sensory results for rancid flavor in *Yukwa*

The *Yukwa* samples were periodically examined by sensory evaluation to detect any development of rancid flavors. Rancidity was defined as objectionable flavors resulting from the accumulation of oxidative decomposition products. Sensory evaluation results are shown by Table 3.

Table 2. Acid, peroxide, p-anisidine and totox values in *Yukwa* samples fried in three different oils with 200 ppm green tea extract (GTE) at the begining of experiment and after certain storage time at 40°C

Quality characteristic values	Storage time (week)	Oils					
		RBO		WRBO		SBO	
		-	GTE	-	GTE	-	GTE
Acid value (KOH · mg/g)	0	$0.36^{b1)}$	0.23^{cd}	0.27^{c}	0.21^{d}	0.45^{a}	0.40^{ab}
	12	1.52^{b}	1.17^{c}	1.48^{b}	1.11^{d}	1.64^{a}	1.11^{d}
Peroxide value (meq/kg)	0	2.83^{a}	2.58^{ab}	2.54^{ab}	2.28^{b}	1.76^{c}	1.68^{c}
	4	58.49^{a}	40.76^{c}	59.25^{a}	38.42^{d}	58.97^{a}	49.75^{b}
	6	132.16^{a}	99.32^{c}	120.59^{b}	95.77^{e}	132.50^{a}	98.03^{d}
	12	372.19^{b}	292.53^{e}	354.12^{c}	263.94^{f}	387.07^{a}	297.68^{d}
p-anisidine value	0	4.54^{a}	4.51^{a}	4.46^{a}	4.43^{a}	3.59^{b}	3.37^{b}
	4	12.36^{b}	9.59^{d}	11.91^{b}	9.30^{d}	14.28^{a}	10.28^{c}
	6	21.54^{a}	17.61^{b}	21.50^{a}	16.67^{c}	21.11^{a}	17.61^{b}
	12	47.04^{b}	33.35^{e}	45.15^{c}	32.49^{f}	49.28^{a}	34.51^{d}
Totox value	0	10.18^{a}	9.67^{ab}	9.54^{ab}	8.99^{b}	7.11^{c}	6.73^{c}
	4	129.34^{b}	91.11^{d}	130.41^{ab}	86.14^{e}	132.22^{a}	109.8^{c}
	6	285.86^{a}	216.25^{d}	262.68^{b}	208.21^{e}	286.11^{a}	213.67^{c}
	12	791.42^{b}	618.41^{e}	753.39^{c}	560.37^{f}	823.42^{a}	629.87^{d}

Abbreviation: See footnote of Table 1.

[1)]Means with the same letter in each row among samples are not significantly different ($p<0.05$).

Table 3. Results[1)] of the difference from control test for rancid flavor of *Yukwa* fried in various oils during storage period at 40°C

Storage week	RBO		WRBO		SBO	
	-	GTE	-	GTE	-	GTE
3	${}^{B}2.75^{b}$	${}^{C}1.5^{c}$	${}^{D}2.13^{bc}$	${}^{B}1.63^{c}$	${}^{C}4^{a}$	${}^{B}2.25^{bc}$
4	${}^{AB}3.38^{b}$	${}^{BC}1.88^{c}$	${}^{BCD}3^{bc}$	${}^{B}1.75^{c}$	${}^{BC}4.63^{a}$	${}^{AB}2.63^{bc}$
5	${}^{AB}3.5^{ab}$	${}^{BC}2.13^{b}$	${}^{B}4.13^{ab}$	${}^{B}2.25^{b}$	${}^{BC}4.63^{a}$	${}^{B}2.13^{b}$
6	${}^{AB}3.63^{ab}$	${}^{BC}2.38^{b}$	${}^{CD}2.5^{b}$	${}^{B}2.38^{b}$	${}^{BC}4.5^{a}$	${}^{AB}2.63^{b}$
7	${}^{A}4.25^{ab}$	${}^{C}1.75^{d}$	${}^{BCD}3.25^{ab}$	${}^{AB}2.88^{cd}$	${}^{ABC}4.88^{a}$	${}^{B}2^{cd}$
8	${}^{AB}3.38^{b}$	${}^{BC}2.5^{b}$	${}^{BC}3.38^{b}$	${}^{B}2.5^{b}$	${}^{BC}4.75^{a}$	${}^{B}2.25^{b}$
9	${}^{AB}4^{b}$	${}^{BC}2.38^{cd}$	${}^{B}4^{b}$	${}^{B}1.75^{d}$	${}^{A}5.88^{a}$	${}^{B}2.25^{cd}$
10	${}^{A}4.5^{ab}$	${}^{ABC}2.75^{c}$	${}^{BC}3.63^{ab}$	${}^{B}2.5^{c}$	${}^{AB}5.13^{a}$	${}^{AB}3.38^{bc}$
11	${}^{AB}4^{bc}$	${}^{AB}3.13^{cd}$	${}^{B}3.75^{bc}$	${}^{B}2.5^{d}$	${}^{AB}5.5^{a}$	${}^{AB}3.5^{cd}$
12	${}^{A}4.5^{bc}$	${}^{A}3.88^{c}$	${}^{A}5.25^{ab}$	${}^{A}4^{bc}$	${}^{AB}5.5^{a}$	${}^{A}4^{bc}$

Abbreviation: See footnote of Table 1.

[1)]Means of difference score from control: 0, equal to control; 1, slight difference; 2, more distinct difference but still acceptable; 3, beginning to lose acceptability; 4, more distinct loss of acceptability; 5, very distinct loss of acceptability; 6, unacceptability.

[a-d]Means with the same letter in each row among samples are not significantly different ($p<0.05$).

[A-C]Means with the same letter in each column among samples are not significantly different ($p<0.05$).

Rancidity was considered to have occurred when a sample was scored 3 points or higher by the panels. Without GTE, rancid flavors in *Yukwa* fried in SBO was already detected after 3 weeks, and after 4 weeks in those fried in RBO and WRBO. With GTE, rancid flavors in *Yukwa* fried in SBO, RBO and WRBO were delayed until weeks 10, 11, and 12, respectively; the addition of 200 ppm GTE delayed rancidity in *Yukwa* by 7 ~ 8 weeks. Therefore, these results demonstrate that GTE can retard the development of rancid flavors in *Yukwa* fried in SBO, RBO and WRBO.

In summary, as the concentrations of GTE in SBO increased from 200 ppm to 600 ppm, increases of the chemical measurements of oxidation (AV, POV, AnV and totox) in *Yukwa* fried in SBO decreased in a dose dependent manner over the 12 week storage period at 40°C. Oxidative stability of *Yukwa* fried in SBO was least; followed by RBO > WRBO. Incorporation of GTE into all three oils reduced the values of chemical measurements of oxidation in *Yukwa* during storage; therefore acting as an antioxidant. In SBO, RBO and WRBO, the effectiveness of GTE was similar. As the chemical measurements of rancidity increased, rancid flavors increased in a similar manner. These results provide convincing evidence that the shelf-life of *Yukwa* can be extended by the incorporation of 200 ppm GTE into the frying oils, thereby reducing the rancidity of *Yukwa*, and demonstrating that GTE is an effective antioxidant for *Yukwa*.

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