

# Addition of Ferulic Acid, Ethyl Ferulate, and Feruloylated Monoacyl- and Diacylglycerols to Salad Oils and Frying Oils

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**ABSTRACT:** To determine antioxidative effects of ferulic acid and esterified ferulic acids, these compounds were added to soybean oils (SBO), which were evaluated for oxidative stability and frying stability. Additives included feruloylated MAG and DAG (FMG/FDG), ferulic acid, ethyl ferulate, and TBHQ. After frying tests with potato chips, oils were analyzed for retention of additives and polar compounds. Chips were evaluated for hexanal and rancid odor. After 15 h frying, 71% of FMG/FDG was retained, whereas 55% of ethyl ferulate was retained. TBHQ and ferulic acid levels were 6% and <1%, respectively. Frying oils with ethyl ferulate or TBHQ produced significantly less polar compounds than SBO with no additives. Chips fried in SBO with TBHQ or ferulic acid had significantly lower amounts of hexanal and significantly less rancid odor after 8 d at 60°C than other samples. Oils were also aged at 60°C, and stability was analyzed by PV, hexanal, and rancid odor. Oils with TBHQ or FMG/FDG had significantly less peroxides and hexanal, and a lower rancid odor intensity than the control. FMG/FDG inhibited deterioration at 60°C, whereas ethyl ferulate inhibited the formation of polar compounds in frying oil. Ferulic acid acted as an antioxidant in aged fried food. TBHQ inhibited oil degradation at both temperatures.

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**KEY WORDS:** Antioxidants, ethyl ferulate, ferulic acid, fried food, oil, odor, potato chips, soybean oil.

Phenolic compounds such as caffeic acid and ferulic acid are well-known antioxidants in foods (1–4). Ferulic acid also exists in other forms; for example, in rice bran oil this phenolic compound is esterified to phytosterols. Oryzanol, which is considered one of the components of rice bran oil that helps to stabilize this oil, contains several ferulic acid esters (5). Ferulic acid esters have been shown to inhibit deterioration in frying (6). In nonfood applications, ferulic and caffeic acids have been reported to protect skin against damage from UV radiation (7). Compton *et al.* (8) reported that when ethyl ferulate was transesterified with triolein, it produced ferulyl-substituted acylglycerols that were potential UV absorbers. Flavor deterioration in oils and oil-containing foods can be inhibited by antioxidants, but the effects of additives vary widely depending on factors such as oxidation temperature, oil composition, and form of the

oil (liquid, emulsion) (9–12). For example, in a study using lard, the antioxidant activity order for tocopherols was  $\delta > \gamma > \beta > \alpha$  above 60°C; however, results were  $\alpha > \gamma > \beta > \delta$  between 20 and 40°C (10). The objective of this study was to determine the antioxidant effectiveness of ferulic acid and various forms of ferulic acid (ethyl ferulate and feruloylated MAG and DAG (FMG/FDG)) at two temperatures, 60 and 180°C, in soybean oil. Controls were soybean oil with no additives and soybean oil with TBHQ. Effectiveness of the additives in the oils was measured by PV and hexanal in the 60°C oxidations and by total polar compounds after frying tests at 180°C. Potato chips fried in the soybean oil at 180°C were analyzed for oxidative stability and rancidity after storage. In addition, the retention of the additives in the frying oil was measured.

## EXPERIMENTAL PROCEDURES

**Materials and reagents.** Refined, bleached, and deodorized soybean oil with no additives except citric acid added during processing was obtained commercially. FMG and FDG were prepared by transesterification of ethyl ferulate with soybean oil according to procedures described by Compton *et al.* (8) and Laszlo *et al.* (13). Ethyl ferulate dissolved in FA ethyl esters was obtained as a by-product of the transesterification process by molecular distillation and subsequent filtration to remove ferulic acid (13). Ferulic acid was obtained from Aldrich (Milwaukee, WI), and TBHQ was obtained from Eastman (Kingsport, TN). Idaho Russet potatoes were purchased locally.

**Additives.** The additives were mixed into the soybean oil at the following concentrations: 0.9 mM (200 ppm) TBHQ, 1.3 mM ethyl ferulate, 1.2 mM FMG/FDG, and 0.6 mM ferulic acid. A control of soybean oil with no additives other than citric acid was also included in the study.

**Methods. (i) Initial oil analyses.** FA composition of the initial soybean oil was determined by capillary GC analysis with a Hewlett-Packard 5890 gas chromatograph (Wilmington, DE) equipped with an SP2330 column (30 m, 0.20 mm i.d., 0.20  $\mu$ m film thickness) (Supelco, Bellefonte, PA). Column temperature was first held at 190°C for 5 min and then was programmed to 230°C at 20°C/min. The injector was held at 250°C and the detector at 260°C. Tocopherol content of the soybean oil was determined in triplicate by HPLC with a polar phase column coupled with a fluorescence detector. The high-performance liquid chromatograph was fitted with a 3-mm particle size ultra silica column (25  $\times$  0.49 cm) (Phenomenex, Torrance, CA). The solvent

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system was 2% 2-propanol in hexane and was pumped at 0.5 mL/min. The sample size was 10  $\mu$ L of 50 mg solute/mL mobile solvent. The fluorescence detector was a programmable unit model 1046 A (Hewlett-Packard, Palo Alto, CA). The excitation wavelength was set at 298 nm and the emission wavelength at 345 nm, with the gain at 6. The soybean oil was evaluated for initial PV (AOCS method Cd 8-53) (14) and amount of total polar compounds (AOCS method Cd 20-91 (97) (14) prior to use.

(ii) *Oil shelf life storage.* Oils were aged at 0, 4, and 8 d at 60°C according to AOCS oven storage method Cg 5-99 (13). Oil was evaluated for oxidative stability by PV (AOCS method Cd 8-53) (14) and for hexanal content. A 14-member trained, experienced panel evaluated the oils for rancid odor intensity (AOCS method Cg 2-83) (14).

(iii) *Frying stability.* Intermittent frying at 180°C was conducted with a total frying/heating time of 15 h. The oil (800 g) was heated in 3-L capacity fryers (Model 2540; Presto Industries, Eau Claire, WI) for 5 h each day for 3 d. Fresh Idaho Russet potatoes were peeled then sliced 1-mm thick, rinsed twice in cold water, and fried in 100-g batches for 4 min per batch every 15 min. Fresh oil (80 g) was added to each fryer after 5 and 10 h of frying to maintain the original amount of oil in the fryer. Potato chips were sampled for analysis after 5, 10, and 15 h of frying, then blanketed with argon and frozen until analyzed. Initial oil samples were taken and after 5, 10, and 15 h of frying, and then blanketed with argon and frozen. Potato chips were placed in 1-L wide-mouth glass jars with air in the headspace, and each jar was closed with a screw cap. Potato chips were aged for 0, 4, and 8 d at 60°C. The frying experiment was duplicated.

(iv) *Total polar compound analysis of frying oils.* Amounts of polar compounds formed in the oils were analyzed in duplicate by the AOCS column chromatography method Cd 20-91(97) (14) as a measure of oil deterioration.

(v) *Retention of additives in oils.* Amounts of FMG/FDG, TBHQ, ferulic acid, and ethyl ferulate in the frying oils were measured by HPLC as described previously (8,13).

*Volatile compound analysis of potato chips and salad oils by GC-MS.* Oils and potato chips were analyzed for hexanal content in triplicate using a purge-and-trap apparatus equipped with a test tube adapter (Tekmar model 3000; Tekmar-Dohrmann Co., Cincinnati, OH) coupled with a model 3400 gas chromatograph (GC) and a Saturn 2000 ion trap mass spectrometer (Varian, Inc., Walnut Creek, CA). A 50-mg sample was placed in the 1.9  $\times$  7.6 cm test tube and heated at 100°C for 9 min preheat time. Volatile compounds were trapped on a 30.5-cm Tenax #1 trap, with 10 min sample purge time, 170°C for 6 min desorbing, 180°C desorb temperature, 160°C GC transfer line and valve temperature. Volatile compounds were introduced onto a GC column, DB-1701, 1  $\mu$ m film thickness, 30 m  $\times$  0.32 mm (J&W Scientific, Folsom, CA). The column was programmed at -20°C for 2 min then heated from -20 to 233°C at 3°C/min. Helium flow rate through the column was 2 mL/min with 28 mL/min injector split vent flow. The GC injector was set at 240°C, and the line to the mass spectrometer was set at 230°C. The ion trap mass spectrometer operated in

the EI mode with mass scan range 23 to 400  $m/z$  over 0.8 s. Filament emission current was 25  $\mu$ amp, axial modulation was 2.1 V, the manifold heater was set at 160°C, and the filament/multiplier delay was 2.5 min. Compound structural identifications were made both from spectral comparisons with the NIST 92 MS library (Varian, Inc., Walnut Creek, CA) and from retention time comparisons with standard compounds.

*Sensory evaluation of potato chips.* Potato chips were rated in duplicate for rancid odor by a 12-member trained, analytical, descriptive panel experienced in evaluating fried foods for odor and flavor (15,16). Panelists were presented with 5 g crushed potato chips in 59.2 mL (2 oz) plastic soufflé cups with snap-on lids (Solo Cup Company, Urbana, IL). Panelists rated the potato chips for intensity of rancid odor on a 0–10 intensity scale with 0 = none and 10 = strong. All sensory evaluations were conducted in a panel room with individual booths, temperature control, and with red lighting to mask color differences between samples (15).

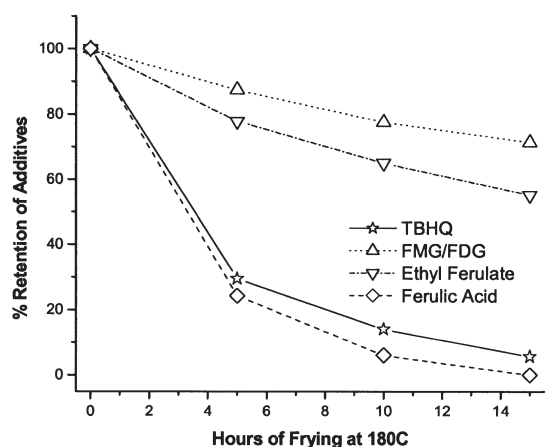
*Statistical analysis.* Data were evaluated by ANOVA (17). Statistical significance was expressed at the  $P < 0.05$  level unless otherwise indicated.

## RESULTS AND DISCUSSION

*Composition of soybean oil.* The unhydrogenated soybean oil was analyzed for FA composition and tocopherol content. Results showed a typical pattern of FA for soybean oils with 10.8% 16:0, 4.8% 18:0, 24.2% 18:1, 52.6% 18:2, and 6.8% 18:3. Total tocopherol content of the oil was 982 ppm (131 ppm  $\alpha$ , 38 ppm  $\beta$ , 648 ppm  $\gamma$ , and 165 ppm  $\delta$ ).

*Loss of additives in fryer oils.* The levels of TBHQ and ferulic acid in the fryer oil decreased rapidly during the first 5 h of frying compared with the decreases of FMG/FDG and ethyl ferulate (Fig. 1). After 5 h of frying potato chips, oil with ferulic acid had lost 77% of the original additive level, whereas there was a 71% loss of TBHQ. In contrast, only 22% of the ethyl ferulate and 13% of the FMG/FDG were lost by 5 h of frying. Over the next 10 h of frying, the levels of the additives decreased at approximately the same rate for ferulic acid and TBHQ, but at a slightly faster rate for FMG/FDG and ethyl ferulate. After 15 h of frying, levels of additives remaining in the oils were <1% for ferulic acid, 8% for TBHQ, 55% for ethyl ferulate, and 71% for FMG/FDG. The within-treatment decreases for each additive were significantly different from one sampling time to the next ( $P \leq 0.05$ ). It is well known that TBHQ dissipates over time at the high temperatures used for frying. Esterification apparently helped to inhibit loss of ethyl ferulate and FMG/FDG compared with ferulic acid.

*Fryer oil degradation.* Total polar compound level is one index of the amount of deterioration in frying oil. Initially, levels of polar compounds were low at 1.9% for all samples (Table 1). At each of the sampling times, the fryer oils with FMG/FDG had higher total polar compound percentages than did the control, with the difference being significant at the 15-h sampling time. The oils with either ethyl ferulate or TBHQ had significantly lower percentages of total polar compounds than



**FIG. 1.** Percentage of additives remaining in soybean oil used for frying potato chips for 0, 5, 10, or 15 h at 180°C. FMG/FDG = feruloylated MAG and DAG.

the control after 15 h. Polar compound levels of the oils with these additives increased as frying time increased linearly, which is typical of most frying oils (15). The amounts of additives remaining in the oils (Fig. 1) did not seem to have an effect on the levels of polar compounds. For example, although the oils with TBHQ or ferulic acid had low amounts of the additives remaining after 15 h, they had low levels of polar compounds. Therefore, inhibitory effects due to products formed from the decomposition of TBHQ and ferulic acid may have occurred. On the other hand, oils treated with FMG/FDG, with 71% of the original amount remaining, had the highest level of total polar compound formation even though they retained the highest levels of the additive, indicating that this compound did not have an inhibitory effect on polar compound formation. A high correlation was seen between percent additive retention and polar compound level for the oil with ethyl ferulate.

**Oxidative stability of potato chips.** Volatile decomposition products in aged fried food are an important index to the stability of potato chips. Hexanal, which is a direct decomposition product of linoleic acid hydroperoxides (18,19) was determined by purge-and-trap GC-MS as a measure of oxidative stability. In potato chips sampled at the 5-h oil use time, samples fried in oils with either ferulic acid or TBHQ had significantly lower hexanal levels than the potato chips fried in oil with ethyl ferulate, FMG/FDG, or the control (Fig. 2A). Potato chips fried in oil with ferulic acid were more oxidatively stable than the control; however, the FMG/FDG additive had a pro-oxidant effect compared with the control. Even though TBHQ

and ferulic acid were retained at only 25–30% after 5 h of frying, the levels were apparently high enough to protect the potato chips fried in these oils from developing high levels of hexanal as was observed in the control. After 15 h of frying time, the hexanal contents of the potato chips fried in the control and in oil with ferulic acid or ethyl ferulate were not significantly different (Fig. 2B). The potato chips fried in soybean oil with TBHQ had significantly less hexanal than the control, whereas the potato chips fried in the oil with FMG/FDG had significantly more hexanal than the control. These patterns remained the same after 8 d of storage, except that the potato chips fried in soybean oil with either TBHQ or ferulic acid had significantly less hexanal than the other samples. The addition of FMG/FDG had a pro-oxidant effect on potato chips fried in 5-h and in 15-h oil (Figs. 2A, 2B). The amounts of hexanal in these samples were significantly higher than in the control.

**Odor analysis of potato chips.** Volatile degradation compounds are important markers for oil and fried food deterioration, but sensory analysis of food is the best method for determining the effects of antioxidants on food products. Sensory panelists rated rancid odor intensity of the potato chips from the 15-h fry time as a measure of fried food stability. As expected, the results of the rancid odor evaluations (Fig. 3) agreed with the results of the hexanal measurements for the 15-h samples (Fig. 2B). Only results for the 15-h potato chips are shown because of the low scores (<1) for rancid odor intensity for the 5-h samples. After 4 d of storage at 60°C, no significant differences were seen between the intensity of rancid odor in the control and the other potato chip samples except for the TBHQ sample. Potato chips fried in oil with TBHQ had significantly less rancid odor intensity than the other potato chips. After 8 d of storage at 60°C, potato chips fried in oil with TBHQ were not significantly different from the potato chips fried in oil with ferulic acid. All other potato chips had significantly more rancid odor than the potato chips fried in the TBHQ-treated oil. As in the hexanal analysis, the potato chips fried in the oil with FMG/FDG had significantly more oxidation than the control. Under frying conditions, the additions of either TBHQ or ferulic acid acted as antioxidants in the potato chips whereas FMG/FDG was a pro-oxidant compared with the control. The measurement of the additives remaining in the frying oil may not be a good predictor of the oxidative stability of the oil or fried food. For example, the potato chips with the best stability were fried in oil with either TBHQ or ferulic acid. Degradation products such as quinones from carnosic acid in rosemary can act as antioxidants (20–22); therefore, quinones and other an-

**TABLE 1**  
Percent Total Polar Compounds<sup>a</sup> in Soybean Oil Used for Frying Potato Chips for 0, 5, or 15 h at 180°C

Hours of frying	Additives				
	Control	TBHQ	FMG/FDG	Ethyl ferulate	Ferulic acid
0	1.9a	1.9a	1.9a	1.9a	1.9a
5	4.3a	4.2a	5.0b	4.0a	4.8b
15	8.5a	7.3b	9.4c	7.2b	8.2a

<sup>a</sup>Values in rows with letters in common are not significantly different ( $P \leq 0.05$ ). FMG/FDG = feruloylated MAG and DAG.

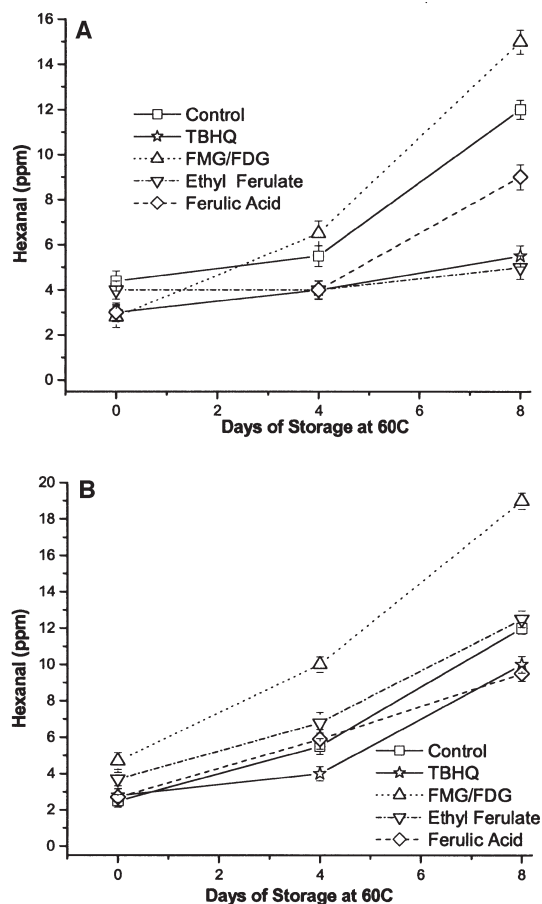


FIG. 2. Hexanal content in potato chips fried in soybean oils used for frying potato chips (A) for 5 h and (B) for 15 h and (A,B) aged 0, 4, or 8 d at 60°C. For abbreviation see Figure 1.

tioxidative degradation products from TBHQ and ferulic acid may possibly help to inhibit oxidation in the samples in this study even though the original additive is reduced to low levels. Quinones are known to act as antioxidants during frying because of the low oxygen pressure at high temperatures (23).

**Peroxide formation in salad oils.** As reported in other studies (9–12), we also found that changing the temperature of oxidation significantly influenced the effects of the additives. No significant differences in peroxide levels were found between any of the additive types at 0, 2, or 4 d storage at 60°C (Fig. 4). Soybean oil with TBHQ had little increase in PV during storage even after 8 d at 60°C. Oil with FMG/FDG also had significantly less peroxide formation than the control after 8 d at 60°C. No difference in PV was found between the control and the oil with ferulic acid after 8 d. The addition of ethyl ferulate acted as a pro-oxidant by 8 d of aging compared to the control.

**Hexanal formation in salad oils.** The results of the hexanal levels in the oils after aging at 60°C showed a similar pattern of stability as did the PV at the 8-d storage time (Fig. 5). TBHQ provided the greatest effect as an antioxidant, followed by FMG/FDG. No differences were noted in hexanal levels between the control and the oil with ferulic acid. Ethyl ferulate acted as a pro-oxidant compared with the control. At the 4-d

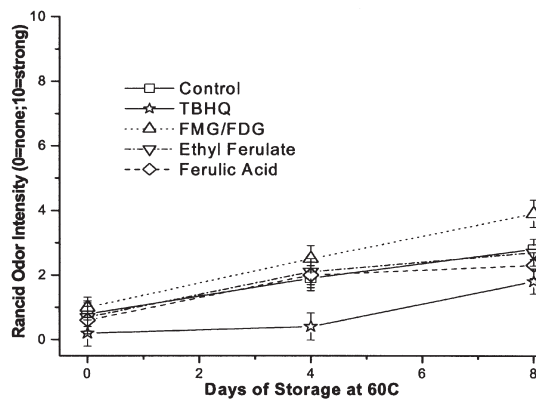


FIG. 3. Rancid odor intensities in potato chips fried in soybean oils used for frying for 15 h and aged 0, 4, or 8 d at 60°C. For abbreviation see Figure 1.

storage time, we found one primary difference between the results from PV and hexanal. In the hexanal analysis, all samples had significantly less hexanal formation than the control; however, no significant differences in peroxide levels were shown. This may be partially explained by the point in the oxidation mechanism where these additives had the best inhibitory effect. For example, these additives may have more effect in inhibiting the decomposition of the hydroperoxides into volatile compounds such as hexanal than in inhibiting the formation of the hydroperoxides.

**Odor analysis of oils.** The results of the rancid odor evaluations of the oils at the 8-d storage (Fig. 6) were not much different from the trends for PV and hexanal contents. The oil with TBHQ had the lowest level of rancid odor, followed by FMG/FDG then ferulic acid, which all had significantly lower odor intensities than the control. The oil with ethyl ferulate had significantly higher rancid odor intensity than the control.

These data showed that FMG/FDG had an antioxidant effect at 60°C but acted as a pro-oxidant in aged fried food and

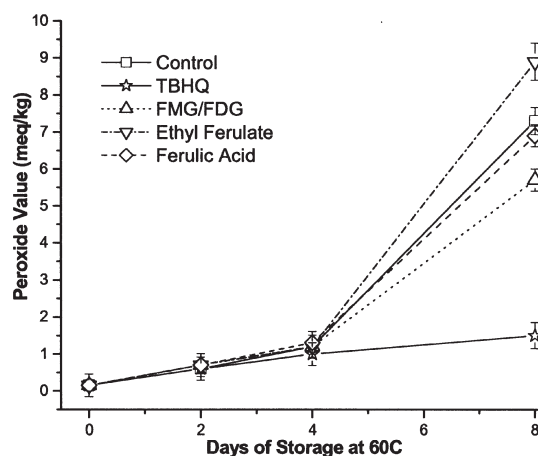


FIG. 4. PV in oils aged 0, 2, 4, or 8 d at 60°C. For abbreviation see Figure 1.

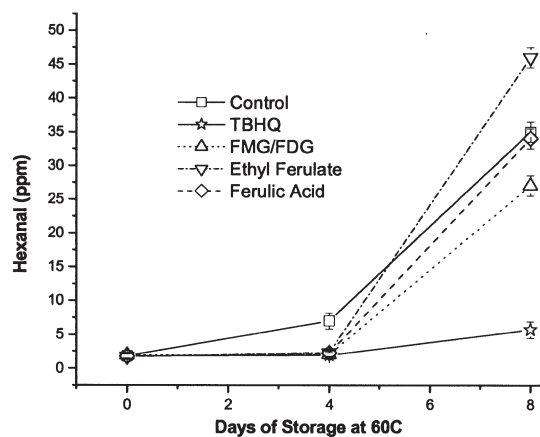


FIG. 5. Hexanal content in oils aged 0, 4, or 8 d at 60°C. For abbreviation see Figure 1.

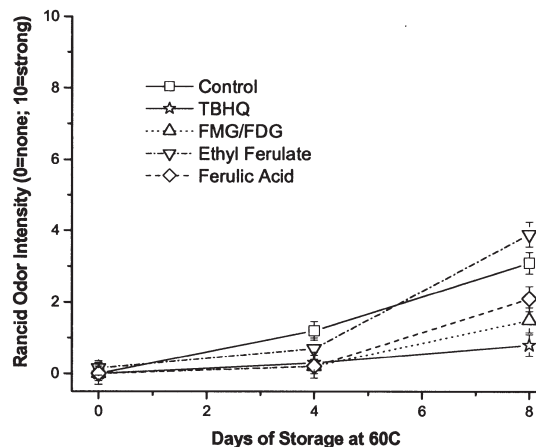


FIG. 6. Rancid odor intensities in oils aged 0, 4, or 8 d at 60°C. For abbreviation see Figure 1.

had the highest level of total polar compounds. On the other hand, ethyl ferulate was effective in frying oils to inhibit formation of total polar compounds, but was not different from the control in the oxidative stability of the fried food. In 60°C storage of the oils, ethyl ferulate was a pro-oxidant. Ferulic acid was better than the control for fried food stability but was similar to the control in 60°C storage tests. Quinones can act as antioxidants better at high temperatures because of low oxygen pressure, which might account for the observation that ferulic acid (and its decomposition products) performed better at 180°C than at 60°C. Some of the differences between the effects of additives may be because of additive concentration, either from the original amount added or from solubility differences in the oil at the two temperatures.

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