

## Preparation of Spread Oils Meeting U.S. Food and Drug Administration Labeling Requirements for *Trans* Fatty Acids via Pressure-Controlled Hydrogenation

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On July 11, 2003, the U.S. Food and Drug Administration (FDA) announced final regulations for *trans* fatty acid (TFA) labeling. By January 1, 2006, the TFA content of foods must be labeled as a separate line on the Nutrition Facts label. Products containing <0.5 g of TFA/14 g serving may be declared as zero. This paper describes technologies allowing compliance with TFA labeling requirements. Soybean oil was hydrogenated in a 2-L vessel at temperatures ranging from 120 to 170 °C at a hydrogen pressure of 200 psi. A commercial nickel-supported catalyst (25% Ni) was used at 0.02% Ni by weight of oil. The hydrogenated oils were characterized for fatty acid composition, solid fat content, and melting point. Compared to commercially processed soybean oil basestocks that typically contain ~40% TFA, those obtained at lower temperatures and higher pressures contain >56% less TFA. Basestocks prepared in the laboratory when blended with liquid soybean oil will yield spread oils meeting FDA labeling requirements for zero TFA, that is, <0.5 g of TFA/serving.

**KEYWORDS:** Soybean oil; hydrogenation; *trans* fat; labeling requirements

### INTRODUCTION

In 1990, the U.S. Congress enacted the Nutrition Labeling Education Act (NLEA) requiring food manufacturers to list total fat, mono- and polyunsaturated fat, and saturated fat on food labels along with cholesterol, calories, and sodium. By 1993, initial proposed U.S. Food and Drug Administration (FDA) regulations had been published that did not include *trans* fatty acids (TFA). Several groups petitioned the FDA to include such information. On November 17, 1999, the FDA announced proposed regulations that included TFA to be listed on labels as a separate line along with a footnote to the effect that TFA consumption should be kept as low as possible. During the period 1999–2003, the proposed regulations were opened for public comment on several occasions. On July 11, 2003, the FDA announced final rules for TFA labeling (1). The key provisions include TFA to be listed as a separate line (<0.5 g of TFA/serving may be declared as zero), and the rule is to be effective January 1, 2006.

In view of the TFA regulations, there is much interest in technologies to decrease the *trans* and saturated fatty acids in food products. Historically, preparation of low/zero *trans* fats has been accomplished by interesterification and fractionation of tropical fats. Blending of tropical fats with liquid vegetable oils and genetic/plant breeding to produce oilseeds with altered fatty acid compositions failed to meet specific end-use require-

ments such as improved flavor and oxidative stability for frying or for use in plastic fats including margarine spreads and shortening (2, 3).

Traditionally, hydrogenation has been the technology of choice for fat modification. Hydrogenated vegetable oils are versatile because a wide variety of spreads, shortenings, and frying fats can be formulated from a basestock system consisting of several hydrogenated oils. However, margarine and shortening basestocks tend to be high in TFA (4). Typically, iodine value (IV) 65 margarine stock soybean oil and IV 80 shortening stock contain 40 and 32% TFA, respectively. Although it is generally well-known that TFA suppression during hydrogenation can be achieved through control of temperature, pressure, catalyst concentration, and agitation, the industry has been slow to adapt changes (5). TFA isomers are reduced in favor of *cis* with increasing hydrogen pressure (6). Low temperatures, high pressures, and high catalyst concentrations are apparently not attractive to oil processors because of decreased throughput, higher catalyst costs, and the retooling of converters to withstand higher pressures. This paper reports studies aimed at reducing the TFA content of margarine spread oils via pressure-controlled hydrogenation. Although the 200 psi used in this study is significantly higher than commercial pressures of 20 psi, it is deemed a reasonable alternative (7).

### EXPERIMENTAL PROCEDURES

A single lot of refined, bleached, deodorized (RBD) soybean salad oil was used for the hydrogenation studies and was supplied by C&T

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**Table 1.** Properties of Hydrogenated Soybean Oil<sup>a</sup>

oil <sup>b</sup>	temp (°C)	pressure (psi)	IV	reaction time (min)	trans (%)	selectivity		mp (°C)	fatty acid composition (%)					solid fat content (%) at				
						Ln	Lo		C16	C18	C18:1	C18:2	C18:3	10 °C	21.1 °C	26.7 °C	33.3 °C	40 °C
A	221	20	65	50	39.7	— <sup>c</sup>	—	41.2	11.3	13.6	75.2	0	0	73.7	54.1	44.7	22.3	3.3
B	140	200	66.2	152	16.6	2.4	4.8	46.4	10.7	22.1	55.4	10.5	0.1	46.5	31.2	27	20.9	9.8
C	150	200	63.6	72	17.4	1.6	5.0	48.2	10.7	23.5	55.4	8.8	0.3	51.4	35	31.6	24.3	12.7
D	160	200	70.2	33	16.8	1.6	5.3	46.1	10.7	19.4	56.4	11.8	0.5	43.7	27.6	23.8	17.6	9.2
E	170	200	68.8	21	17.9	1.5	5.5	48.2	10.7	20.1	56.8	10.8	0.5	46.4	30.4	26.7	19.5	10.3

<sup>a</sup> Nickel was used at 0.02% for all hydrogenations. <sup>b</sup> Letters represent temperature–pressure combinations. <sup>c</sup> Cannot be determined.

**Table 2.** Properties of Blended Oils

oil blend <sup>a</sup>	trans (%)	drop point (°C)	solid fat content (%) at					g of TFA/14 g serving <sup>b</sup>			
			10 °C	21.1 °C	26.7 °C	33.3 °C	40 °C	80% fat	70% fat	60% fat	50% fat
A	11.9	31.4	17.2	8.6	5.0	1.0	0.0	1.33	1.16	1.00	0.83
B	5.0	37.7	11.1	6.3	5.0	3.1	1.3	0.56	0.49	0.42	0.35
C	5.2	39.4	11.8	7.9	6.1	3.6	1.8	0.58	0.51	0.47	0.36
D	5.8	36.9	9.5	6.0	4.4	2.2	0.6	0.65	0.51	0.44	0.41
E	5.4	37.8	10.3	6.6	5.1	2.6	1.5	0.60	0.53	0.45	0.38
commercial spread oils <sup>c</sup>	5.3–14.5	31.6–33.1	10.0–18.0	4.4–8.0	2.8–4.7	0.9–1.7	0				

<sup>a</sup> Oil blends A–E were prepared by mixing 30% hydrogenated (A–E in **Table 1**) basestock with 70% liquid soybean oil. <sup>b</sup> Fat content for blend shown as percent of blended oil in final product. <sup>c</sup> Partially hydrogenated/liquid soybean based.

Refinery (Charlotte, NC). It had the following fatty acid composition as determined by gas–liquid chromatographic analysis of the methyl esters: palmitic, 10.5%; stearic, 4.7%; oleic, 23.5%; linoleic, 52.2%; linolenic, 7.5%. It had a calculated IV of 130.7. The catalyst was a commercial nickel catalyst supported on alumina 26.5% nickel and was supplied by Calicat E-479D, Mallinckrodt (Erie, PA). The final nickel concentration in the hydrogenation was 0.02 wt % of oil. Hydrogenations were carried out in a 2-L stainless steel stirred autoclave (Parr Instruments, Moline, IL) described in a previous publication (8). After the hydrogenations were completed, the catalyst was removed by filtration. Compositional analyses were made by capillary gas chromatography. The TFA composition was determined using AOCS Official Method Ce 1f-96 (9). One hundred milligram SBO samples were transesterified and analyzed by gas chromatography (GC) as described previously (10). Solid fat contents and dropping points were determined using AOCS Official Methods Cd 16-81 and Cc 18-80, respectively (9, 11).

## RESULTS AND DISCUSSION

The compositional and physical properties of soybean oil hydrogenated at temperatures ranging from 140 to 170 °C and a hydrogen pressure of 200 psi are shown in **Table 1**. For comparison, a commercially prepared margarine stock (IV 65) is included. Under commercial conditions of 221 °C, 20 psi, and 0.02% nickel catalyst concentration, extensive *trans* formation occurs. The experimental oils (B–E) show significant reduction in *trans* fatty content of the order of >50%.

Previous publications from our laboratory showed that a temperature of 120 °C and pressures up to 500 psi yield products with even lower TFA content (7, 8). However, under these conditions, slow rates of reaction were observed. At a pressure of 200 psi and temperatures of 140–170 °C, the time required to obtain IVs of 65–70 is reduced to <45 min and is actually shorter than that observed in commercial practices. However, because commercial reaction vessels differ in their size and geometry compared with our laboratory reactor, reaction times may not be directly comparable. Under the conditions employed in the experimental oils (B–E), some selectivity is lost as evidenced by increased stearic acid content and by the fact that 8–10% linoleic acid remains at IVs of ~70. The commercially prepared margarine stock contains little if any linoleic acid at

IV 65. Published data indicate that under selective conditions used for margarine, shortening, and salad oil production, linolenate selectivity ratios of ~2 are observed (5). The experimental oils B–E show linolenate selectivities of ~1.5–1.6. The solid fat content of commercial margarine stocks exhibits steep, sharply melting profiles with a melting point of ~41 °C. The experimental oils (B–E) show melting points in the 46–48 °C range largely because of the increased stearic acid content. The solid fat contents of the experimental oils (B–E) are somewhat flatter than that of the commercial sample. Although this is undesirable in spreads, it would be an advantage in shortening formulations. Nonetheless, these hydrogenated oils appear to be suitable as soft margarine stocks.

The properties of blends made from the hydrogenated oils (30%) and liquid soybean oil (70%) are shown in **Table 2**. The solid fat contents and melting points of the blends qualify them as soft tub margarines and spreads. In addition, the blended oils contain only ~5–6% TFA, which qualifies them as zero grams of *trans* fat per serving spreads at fat levels of 70% or below.

Thus, with the acceptable reaction rates observed at higher pressures and lower temperatures along with >56% reduction in TFA formation, it should be possible to prepare low *trans* spread oils via hydrogenation with commercially available nickel catalysts. The basestocks of 17% TFA shown in **Table 1** should prove to be suitable for reduced *trans* shortening as well. Blends of the basestocks, liquid oils, and stearines will produce shortening fats below the usual levels of 20–25% TFA found in commercial products.

In commercial practice, hydrogenation converters can be operated at pressures up to ~50 psi and would require refitting to handle the higher pressures of 200 psi used in this study. However, rapid reaction rates, the benefit of TFA suppression, and the ready availability of commercial catalysts suggest that the technology may be effective for producing reduced *trans* oils. Further work is underway.

## LITERATURE CITED

- (1) FDA. Food labeling: *Trans* fatty acids in nutrition labeling. *Fed. Regist.* **2003**, 68, 41433–41506.

- (2) List, G. R. Processing and reformulation for nutritional labeling of *trans* fatty acids. *Lipid Technol.* **2004**, 16 (8), 173–177.
- (3) List, G. R. Decreasing *trans* and saturated fatty acid content in food oils. *Food Technol.* **2004**, 58, 23–31.
- (4) O'Brien, R. D. *Fats and Oils: Formulating and Processing for Applications*; CRC Press: Boca Raton, FL, 2004; pp 235–289.
- (5) Patterson, H. B. W. *Hydrogenation of Fats and Oils: Theory and Practice*; AOCS Press: Champaign, IL, 1994.
- (6) Hsu, N.; Diosady, L. L.; Rubin, L. J. Catalytic behavior of palladium in the hydrogenation of edible oils II. Geometrical and positional isomerization characteristics. *J. Am. Oil Chem. Soc.* **1989**, 66, 232–236.
- (7) List, G. R.; Neff, W. E.; Holliday, R. L.; King, J. W.; Holser, R. Hydrogenation of soybean oil triglycerides: Effect of pressure on selectivity. *J. Am. Oil Chem. Soc.* **2000**, 77, 311–314.
- (8) King, J. W.; Holliday, R. L.; List, G. R.; Snyder, J. M. Hydrogenation of soybean oil in supercritical carbon dioxide. *J. Am. Oil Chem. Soc.* **2001**, 78, 107–113.
- (9) AOCS. *Official Methods and Recommended Practices of the AOCS*, 5th ed.; Firestone, D., Ed.; AOCS Press: Champaign, IL, 2003; methods Ce 1f-96, Cc 18-80, Cd 16-81.
- (10) House, S. D.; Larson, P. A.; Johnson, R. R.; DeVries, J. W.; Martin, J. W. Gas chromatographic determination of total fat extracted from food samples using hydrolysis in the presence of antioxidant; *J. Assoc. Off. Anal. Chem.* **1994**, 77, 960–965.
- (11) List, G. R.; Steidley, K. R.; Palmquist, D.; Adlof, R. O. Solid fat index (SFI) vs. solid fat content (SFC): a comparison of dilatometry and pulsed NMR for solids in hydrogenated soybean oil. In *Crystallization and Solidification of Lipids*; Widlak, N., Hartel, R., Narine, S., Eds.; American Oil Chemists' Society, AOCS Press: Champaign, IL, 2001; pp 146–152.

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