Formulation and Optimization of Sucrose Polyester Physical Properties by Mixture Response Surface Methodology

Chwen-Jen Shieh, Casimir C. Akoh*, and Philip E. Koehler

Department of Food Science and Technology, The University of Georgia, Athens, Georgia 30602-7610

ABSTRACT: The physical properties of sucrose polyester (SPE), prepared from different composite blends of fatty acid methyl esters (FAME) of safflower oil, palm oil, and peanut oil, were evaluated by mixture response surface methodology. Optimum combinations of fatty acids to achieve specific physical properties of SPE were determined. The SPE most similar in physical properties to peanut oil was obtained with a 55:45 molar ratio of mixed FAME from safflower oil and peanut oil. The physical properties of SPE were significantly affected by the degree of saturation and the average chainlength of their composite fatty acids.

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KEY WORDS: Formulation, mixture response surface methodology, optimization, physical properties, sucrose polyester.

The use of sucrose polyester (SPE) in foods as reduced or zero calorie fat and oil substitutes has already been brought to consumers' attention by the food industry (1). SPE is a fat-like material, miscible in triglycerides and insoluble in water. SPE consists of a mixture of hexa-, hepta-, and octaesters, formed by the reaction of sucrose with long-chain fatty acids. They are not hydrolyzed by the lipolytic enzymes in the intestinal tract, and thus they are not absorbed. SPE has the taste and consistency of conventional vegetable oil (2). It also has shown promise in reducing blood cholesterol and body weight of obese individuals (3).

The physical properties of SPE have been reported by Akoh and Swanson (1), Hamm (4), and Jandacek and Webb (5). The physical properties of dietary triglycerides depend on the fatty acid composition of the fat. The melting points of triglycerides are dependent on the fatty acid composition, chainlength, and degree of unsaturation of their fatty acid constituents. The melting point of SPE also reflects their fatty acid constituents. For example, sucrose octastearate is a high-melting material, whereas sucrose octaoleate is liquid at room temperature (5). The density, specific gravity, and refractive indices of SPE approximate those of commercial vegetable oil (6). However, the apparent viscosities of SPE are significantly greater than those of salad oil (1,5). Hamm (4) and Akoh and Swanson (1) suggested that the physical properties of SPE can be adjusted by varying the degree of unsaturation and chainlength of the fatty acids used in the synthesis to produce SPE with functional properties appropriate for food use. The effect of fatty acid composition on the physical properties of SPE has to be studied further.

Mixture response surface methodology (RSM) was developed by Claringbold (7) and Scheffé (8,9). Cornell (10) provided an excellent summary on this topic in a review with an update and bibliography (11). Mixture RSM has been successfully applied to optimize formulations in food research by several investigators. Examples include blending fruit juices (12), composite flours in formulation tortillas (13), and development of coffee whitener from peanut extract (14). However, it has not been used for optimizing the physical properties of SPE.

The objective of this study was to optimize the physical properties of SPE synthesized from composite fatty acid methyl esters (FAME) blends of safflower oil, palm oil, and peanut oil by using mixture RSM. Specific objectives were as follows: (i) to determine physical characteristics of SPE prepared from composite blends of FAME by mixture RSM, (ii) to determine FAME levels at which peanut oil can be replaced and result in SPE with physical characteristics comparable to 100% peanut oil, and (iii) to validate prediction models for selected attributes. Triangular contour plot and superimposing technique were employed to reach the objectives.

MATERIALS AND METHODS

Experimental design. A three-variable simplex centroid design was employed in which the number of points was $2^q - 1$, where q is equal to the number of variables, i.e., 3 (9). This design resulted in seven FAME mixtures (Table 1). The molar ratio (percentage) of composite blends of FAME was present at levels ranging from 0 to 100%. Two replications of the study were conducted.

Materials. Safflower oil and peanut oil were purchased locally (Athens, GA). Palm oil was kindly donated by Fuji Vegetable Oil, Inc. (Savannah, GA). Sucrose, potassium hydroxide, sodium hydroxide, molecular sieve 4Å, and hydrochloric acid were purchased from J.T. Baker Co. (Phillipsburg, NJ).

*To whom correspondence should be addressed at Department of Food Science and Technology, Food Science Building, The University of Georgia, Athens, GA 30602-7610.

TABLE 1
Percentage Composition of Seven Sucrose Polyester Formulations Representing a Mixture Design Experiment Utilizing Blends of Fatty Acid Methyl Ester (FAME) Prepared from Safflower, Palm, and Peanut Oil[a]

Formula number	Component of FAME		
	Safflower oil	Palm oil	Peanut oil
1	100	0	0
2	0	100	0
3	0	0	100
4	50	50	0
5	50	0	50
6	0	50	50
7	33.3	33.3	33.3

[a]Mixture design results in seven formulations.

Engelhard F-160 bleaching clay was supplied by Engelhard Corporation (Jackson, MS). All organic solvents and potassium carbonate were from Fisher Scientific (Norcross, GA).

Synthesis and purification. SPE was synthesized by the method of Boutte and Swanson (15), and the reaction condition was based on the investigation of Shieh *et al.* (16), in which optimum parameters were as follows: reaction time of 11.5 h, synthetic temperature of 144°C, and substrate molar ratio of 11.4:1 (FAME/sucrose). The average molecular weight of FAME was calculated based on the fatty acid composition as determined by gas–liquid chromatography (GLC) (16). In a typical synthesis, 30 g free sucrose (0.088 mol) and 1.00 mol FAME (294.11 g FAME of safflower oil, 288.21 g FAME of palm oil, and 294.32 g FAME of peanut oil) were reacted in a 2-L round-bottom flask attached to a Büchi rotary evaporator (Büchi, Postfach, Switzerland). The speed of the rotary evaporator was set at ~200 rpm. The amount of potassium hydroxide was 2.5%, and methanol was 40% by weight of FAME plus sucrose. Potassium hydroxide was dissolved in methanol first, and FAME was added and stirred at room temperature for 30 min, after which sucrose was added. The reactants were heated to 85°C for 15 min while rotating at 200 rpm at atmospheric pressure to form a soap and to remove most of the methanol. The remaining methanol was removed by applying intermittent vacuum until foaming subsided. Then full vacuum was applied, and the mixture was heated to 144°C. After 2 h, 0.5% (w/w) potassium carbonate was added to catalyze the transesterification (ester exchange) reaction. At 11.5 h, the reaction mixture was cooled to 80°C and neutralized with 2–3 mL concentrated acetic acid. The crude product was washed five times with 1.5 L water (70°C), and five times with 500 mL 95% ethanol (80°C). Then, SPE was stirred and bleached with Engelhard F-160 bleaching clay (15%, w/w) at 100°C for 1.5 h under medium vacuum (~100 mm Hg) in a rotary evaporator. The mixture was dissolved in hexane, and clay particles were removed by filtering the SPE solution through Whatman No. 5 filter paper. A KDL-4 short-path distillation apparatus (UIC, Joliet, IL) was used to remove excess FAME and sucrose esters of low degree of substitution. Conditions for short-path distillation were as follows: evaporator heat of 150°C, condenser temperature of −2.5°C, wiper speed of 500 rpm and vacuum less than 0.02 mm Hg. SPE was passed through short-path distillation twice. SPE was not steam-deodorized.

Analysis and measurements. The fatty acid composition of SPE was obtained by transesterification of SPE with 1.0 M methanolic NaOH and analyzed by GLC (1). Theoretical molecular weight was calculated assuming SPE had eight long-chain fatty acids attached ($C_{14:0}$–$C_{18:3}$), as determined from fatty acid composition by GLC analysis. Refractive indices of SPE were determined with a Bausch and Lomb refractometer (Rochester, NY), equipped with a constant temperature water bath (40°C). Specific gravity was calculated by using the density of SPE at 40°C divided by the density of water at 40°C. Melting points were measured with standard AOCS (1994) method Cc 1-25 (17). The temperature, lower than 0°C, was reached by cooling 100% ethanol with dry ice (solid carbon dioxide). The viscosity of SPE was measured with a Brookfield viscometer (Model RVTD; Stoughton, MA), and viscosity values were expressed in centipoise (cp) at 40°C. The oxidative stability index (OSI) was determined as described by Akoh (18).

Statistical analysis. The data were analyzed with the Statistical Analysis System (19). Multiple regression analysis (Proc Reg) was used to fit a quadratic canonical polynomial model, described by Scheffé (9), as follows: $Y = \beta_1X_1 + \beta_2X_2 + \beta_3X_3 + \beta_1\beta_2X_1X_2 + \beta_1\beta_3X_1X_3 + \beta_2\beta_3X_2X_3$ where Y is a dependent variable (each property of SPE), β_1, β_2, β_3, $\beta_1\beta_2$, $\beta_1\beta_3$, $\beta_2\beta_3$ are the corresponding parameter estimates for each linear and cross-product term produced for the prediction models, X_1 is the FAME prepared from safflower oil, X_2 is the FAME prepared from palm oil, X_3 is the FAME prepared from peanut oil. The intercept and quadratic terms were removed from the models in accordance with procedurés described by Cornell (20,21). The intercept is not included in the analysis because the mixture components should be equal to 100% of the mixture. The stepwise option was employed to eliminate insignificant variables in a model.

Optimization. The predicted models were used to generate triangular contour plots. In this study, ranges of acceptability used in formulation optimization were determined for the melting point, viscosity, and OSI by means of the following criteria: (i) melting point less than 10°C, (ii) viscosity less than 200 cp, and (iii) OSI greater than 1 h. The optimum area obtained by superimposing the acceptable areas represented the combination of mixtures that would meet pre-set criteria for an acceptable formulation.

Model verification. The predicted values for each dependent variable were calculated from the corresponding reduced models. The chi-square test described by Ott (22) was performed to determine whether the observed values were significantly the same as the predicted values from the predicted model.

RESULTS AND DISCUSSION

The fatty acid profile and physical properties of SPE and peanut oil are illustrated in Table 2. The unsaturated fatty

TABLE 2
Observed Experimental Values and Predicted Values for Some Properties of Sucrose Polyester Prepared from Composite Blends of FAME of Safflower, Palm, and Peanut Oil

	Fatty acid profiles						Physical properties				
Formula no.[a]	Saturated fatty acid (%)	Unsaturated fatty acid[b] (%)	MUFA (%)	PUFA[c] (%)	Average chainlength	Theoretical molecular weight[d]	Refractive index (40°C)	Specific gravity (40°C/40°C)	Melting point (°C)	Viscosity (40°C) (cp)	Oxidative stability index (h)
1	11.34	88.66	17.81	70.86	17.834	2298.0	1.4737	0.9334	−8.61	140	0.85
	11.28[e]	88.69	17.97	70.80	17.805	2297.8	1.4736	0.9339	−8.74	139	0.66
2	48.68	51.32	40.73	10.60	17.107	2230.6	1.4672	0.9493	29.72	489	5078
	48.84	51.28	40.86	10.41	17.103	2229.5	1.4671	0.9498	29.59	488	5.76
3	19.99	80.01	49.92	30.08	17.985	2317.6	1.4703	0.9460	20.56	293	1.28
	20.21	79.21	49.31	29.98	17.942	2317.3	1.4702	0.9465	20.42	292	1.08
4	29.97	70.03	29.46	40.57	17.466	2264.5	1.4716	0.9480	13.61	286	0.95
	30.06	69.99	29.41	40.61	17.454	2263.7	1.4716	0.9462	14.13	289	1.00
5	13.95	86.05	33.63	52.42	17.781	2295.3	1.4730	0.9473	1.67	185	0.48
	13.91	86.09	33.64	52.29	17.873	2293.2	1.4730	0.9455	2.19	488	0.87
6	34.98	65.02	44.98	20.04	17.356	2273.8	1.4691	0.9410	20.83	305	6.35
	34.52	65.25	45.05	20.20	17.370	2273.4	1.4691	0.9391	21.36	308	6.40
7	25.90	74.10	36.50	37.60	17.579	2270.1	1.4715	0.9395	13.33	252	2.95
	25.96	74.00	36.04	37.90	17.547	2274.9	1.4714	0.9436	12.16	246	2.84
Peanut oil	13.38	81.62	50.05	31.57	17.949	873.90	1.4633	0.9100	5.36	57	3.53

[a]Components of composite FAME formulas are shown in Table 1. See Table 1 for abbreviation.
[b]Unsaturated fatty acid includes mono- and polyunsaturated fatty acid (MUFA and PUFA, respectively).
[c]PUFA includes two and three double bonds of fatty acid.
[d]Average molecular weight based on fatty acid composition as determined by gas–liquid chromatography.
[e]Underline indicates predicted value from the model listing in Table 3.

acids included monosaturated and polyunsaturated fatty acids. The theoretical molecular weights of SPE were between 2200 and 2300, based on their fatty acid composition determined by GLC, assuming that SPE contains eight long-chain fatty acids. The theoretical molecular weights of SPE were all significantly larger than that of the peanut oil. The refractive indices of SPE (all greater than that of peanut oil) were positively related to their unsaturated fatty acid amounts, which were always higher than that of peanut oil. The specific gravities of SPE were higher than that of the peanut oil also. The reason why the molecular weight, refractive index and specific gravity were greater than that of peanut oil was that SPE contains approximately eight fatty acids whereas peanut oil contains three fatty acids in one molecule. The range of melting points was wide, from −8.61 to 29.72°C, varying with the different fatty acid compositions. The viscosities of all formulations were significantly higher than that of peanut oil (57 cp). The OSI of SPE were less than that of peanut oil (3.53 h), except for formulas #2 (prepared from 100% palm oil) and #6 (prepared from 50% palm oil). The fatty acid composition of palm oil was high in saturated fatty acid (~50%). The SPE of formula #2 (prepared from palm oil) was semi-solid, and all of the others were liquid at room temperature (22°C). It was observed that SPE must contain over 50% unsaturated fatty acid to be liquid at ambient temperature. The color of SPE was golden yellow, similar to that of peanut oil. Overall, the apparent properties of SPE were similar to those of peanut oil, except viscosity. Hamm (4) suggested that modifying the degree of saturation and average chainlength of the fatty acids used in the synthesis of SPE could affect the physical properties of SPE. Our results, determined by triangular contour plots, confirm earlier reports (4,6).

Results of the regression analyses are presented in Table 3, which lists parameter estimates for the prediction models for fatty acid profiles and the physical properties of SPE. The stepwise regression option was specified to eliminate the insignificant variables (all variables in the model were significant at the 0.15 level). Triangular contour plots, produced from the parameter estimates from each of the attributes, were generated. The composite fatty acid blend is better understood through the triangular contour plots, as shown in Figure 1. For example, the SPE prepared from palm oil was high (~50%) in saturated fatty acid (Fig. 1A), and SPE prepared from safflower oil was high in unsaturated fatty acid, shown in Figure 1B.

The triangular contour plots of the physical properties of SPE are shown in Figure 2. The contour behavior of the refractive index (Fig. 2A) was similar to that of the unsaturated fatty acid (Fig. 2B) and opposite to that of saturated fatty acid. It is apparent that the refractive index increases with an increase in the degree of unsaturation. The refractive index of SPE was comparable to that of liquid sucrose octaesters reported by Akoh and Swanson (1) and Jandacek and Webb (5). Our results suggest that refractive index could be used to predict the degree of saturation and unsaturation of SPE. There

TABLE 3
Parameter Estimates for Variables Used in Prediction Models for the Properties of Sucrose Polyester from Composite Blends of FAME of Safflower, Palm, and Peanut Oil[a]

	Fatty acid profiles						Physical properties				
FAME	Saturated fatty acid	Unsaturated fatty acid	MUFA	PUFA	Average chainlength	Theoretical molecular weight	Refractive index	Specific gravity	Melting point	Viscosity	Oxidative stability index
Safflower oil (SF)	11.283	88.692	17.970	70.805	17.805	2297.824	1.4734	0.934	−8.741	139.347	0.662
Palm oil (PM)	48.838	51.279	40.856	10.409	17.103	2229.521	1.467	0.950	29.589	488.347	5.763
Peanut oil (PN)	20.211	79.212	49.312	29.983	17.942	2317.272	1.470	0.946	20.424	292.347	1.087
SF × PM	—	—	—	—	—	—	0.005	0.018	14.833	−100.947	−8.857
SF × PN	−7.352	8.553	—	7.576	—	−57.488	0.004	0.021	−14.617	−112.947	—
PM × PN	—	—	—	—	−0.609	—	0.002	−0.036	−14.597	−330.947	−11.893

[a]See Tables 1 and 2 for other abbreviations.

was no relationship between the specific gravity (Fig. 2B) and theoretical molecular weight (Fig. 2C) and degree of saturation and unsaturation. The contour behavior of melting point and viscosity (Figs. 2D and E) was obviously similar to that of the degree of saturated fatty acid (Fig. 1A) and somewhat related to average chainlength (Fig. 1E). This revealed that the melting point and viscosity were affected by the degree of saturation and chainlength of their fatty acid composition. The higher the saturated fatty acid content, the higher the melting point and viscosity. Figure 3, produced from the prediction model, shows that the major physical properties (melting point, viscosity, and OSI) of SPE were affected by the degree of saturation and average chainlength. Figure 3A revealed that the melting point of SPE was obviously affected by the degree of saturation and average chainlength. Viscosity is also dependent on the chainlength and the degree of saturation (6). Jandacek and Webb (5) reported that apparent viscosity of sucrose octaesters synthesized from homogeneous saturated fatty acids increased with fatty acid chainlength. The contour behavior shown in Figure 3B revealed that viscosity was affected by the average chainlength and the degree of saturation, supporting our previous reports (6). OSI of SPE

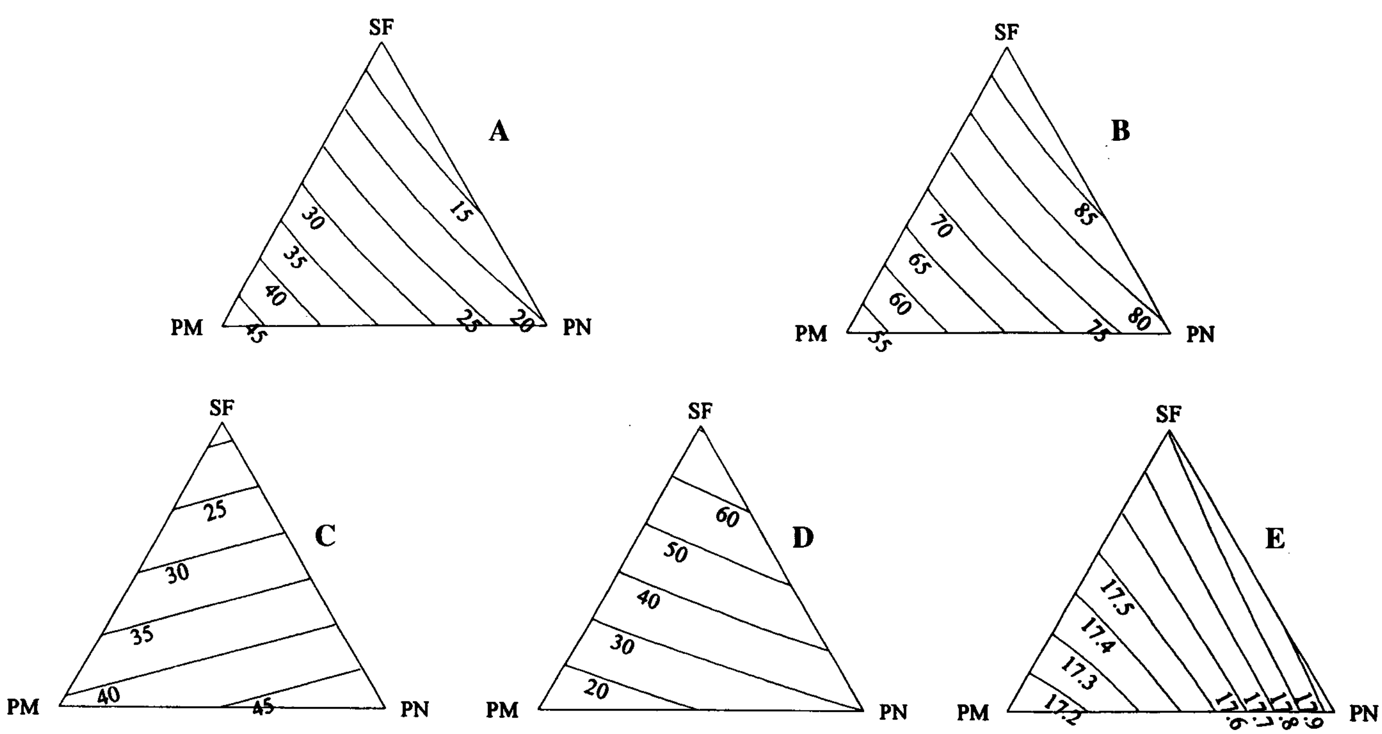

FIG. 1. Triangular contour plots for fatty acid profiles for blends containing composite fatty acid methyl ester prepared from safflower oil (SF), palm oil (PM), and peanut oil (PN). A, Percentage of saturated fatty acid; B, percentage of unsaturated fatty acid; C, percentage of monosaturated fatty acid; D, percentage of polyunsaturated fatty acid; E, average chainlength.

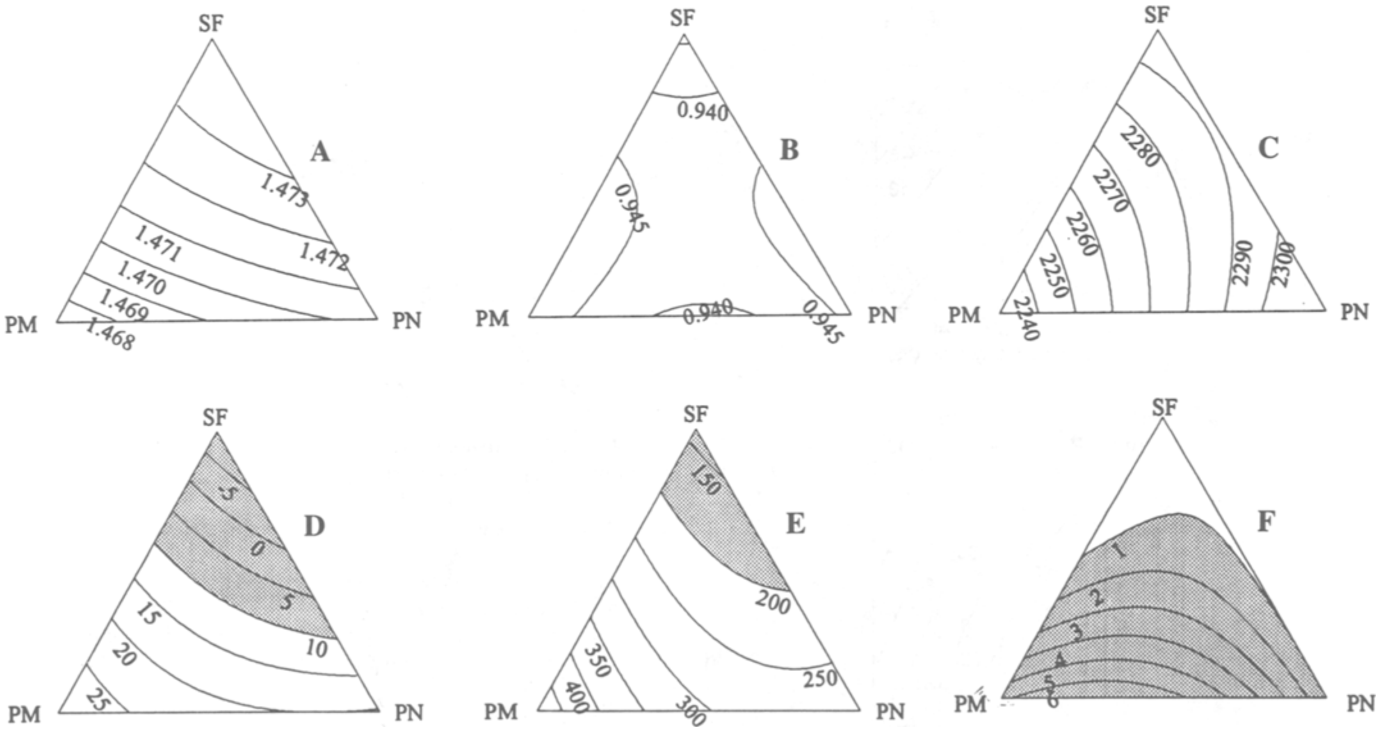

FIG. 2. Triangular contour plots for theoretical molecular weight and physical properties for blends containing composite fatty acid methyl ester prepared from SF, PM, and PN. A, Refractive index; B, specific gravity; C, theoretical molecular weight; D, melting point; E, viscosity; and F, oxidative stability index. The shaded regions represent the accepted area. See Figure 1 for abbreviations.

was only slightly affected by the degree of saturation (compare the contour plots of Fig. 2F and Fig. 1A) and was not affected by the average chainlength of SPE (Fig. 3C). Akoh (18) reported that the stability of SPE depends on the following: (i) the fatty acid composition and degree of unsaturation; (ii) processing conditions; (iii) the presence or absence of residual natural antioxidants; and (iv) storage conditions and nature of abuse. Overall, the physical properties of SPE can be adjusted by modifying the fatty acid chainlength and saturation to simulate the fatty acid composition and/or simulate the physical properties of peanut oil.

The major physical properties (melting point, viscosity, and OSI) of SPE were optimized to stimulate those of peanut oil and to obtain the optimum ratio of starting vegetable oil FAME for SPE synthesis. The accepted areas were chosen based on the actual physical properties of peanut oil shown in Table 2. The accepted properties were as follows: melting point (<10°C), viscosity (<200 cp), and OSI (>1 h). Because all viscosities of SPE were significantly higher than that of peanut oil, the accepted area for viscosity was defined to be less than 200 cp, which was reasonably close to that of peanut oil (57 cp). Optimum areas for the major physical properties were outlined on surface contour plots as shaded regions in Figure 2. Acceptable regions for formulations producing values within the optimum criteria ranges are shown in Figure 4, obtained by superimposing Figures 2D, E, and F. The shaded area in Figure 4 represents the region in which peanut oil could be successfully replaced with the synthesized SPE. Furthermore, the FAME mixture of safflower oil and peanut oil (molar ratio 55:45), which was close to the shaded area, was suggested as the optimum formula in this study because only two kinds of FAME, not three, would be required to synthesize the SPE with the desired physical properties.

Observed and predicted values from the prediction models for the fatty acid profile and physical properties are also presented in Table 2. The chi-square tests indicated that the observed values were significantly the same (P-value > 0.95, degrees of freedom = 6) as the predicted values for all models. This indicates that parameter estimates presented in Table 3 can be used to predict the actual properties of SPE.

Mixture response surface methodology can be used to determine effects of variation in levels of FAME prepared from safflower oil, palm oil, and peanut oil regarding the fatty acid profiles and physical properties of seven SPE formulations. Optimizing based on three attributes—melting point, viscosity, and OSI—indicated that a mixture of FAME (molar ratio 55:45 of FAME prepared from safflower oil and peanut oil) reacted with sucrose can be used to synthesize an SPE with physical properties most similar to that of peanut oil. These findings and this experimental methodology could be useful in the synthesis of SPE from composite FAME to simulate any oil for various food applications.

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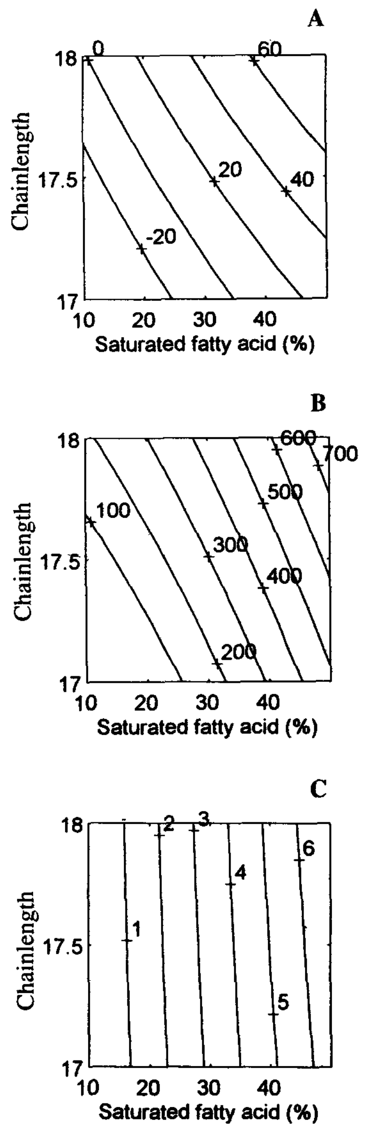

FIG. 3. The contour plots of the major physical properties affected by the degree of saturation and average chainlength. A, Melting point (°C); B, viscosity (cp); and C, oxidative stability index (OSI) (h).

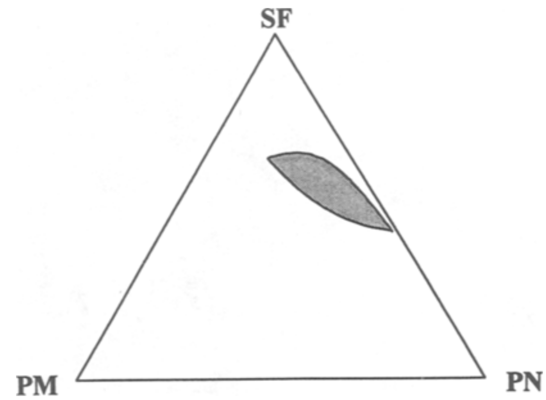

FIG. 4. Optimum regions obtained by superimposing contour plots of major physical properties (melting point, viscosity, and oxidative stability index) for blends containing composite fatty acid methyl ester prepared from SF, PM, and PN. See Figure 1 for abbreviations.

College of Agricultural and Environmental Sciences, The University of Georgia.

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