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ORIGINAL PAPER

Correlation of Rheological and Microstructural Properties in a Model Lipid System

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Abstract Model systems having different microstructures and rheological properties were obtained by controlled crystallization from a mixture of high-melting and low-melting lipids. Based on analysis of confocal scanning light microscopic images, the microstructural characteristics of the systems were quantified by use of different approaches including microstructure density, Euler characteristic, nearest-neighbor analysis, fractal dimension of microstructure interface, and fractal dimension by the particle-counting method (PCM). The solid-fat content (SFC) of semisolid lipid samples was measured by nuclear magnetic resonance (NMR) spectroscopy and rheological properties were analyzed by compressive penetration tests with a texture analyzer. As expected, SFC had a major impact on rheological properties, but lipid crystalline microstructure also had significant effects. Correlation analysis showed that rheological properties were highly correlated with the various quantitative microstructural parameters, with the exception of the fractal dimension by the PCM. Empirical models adequately correlated rheological properties with SFC and microstructure density. Compression modulus increased by a factor of about ten as SFC increased from 0.28 to 0.51. However, for systems with the same SFC, compression modulus was dependent on microstructure. At low SFC compression modulus increased by about a factor of seven over the range of microstructures formed, whereas at higher SFC compression modulus only increased by a factor of about two.

Keywords Crystal network · Microstructure · Rheological property · Solid-fat content · Microstructure density · Euler characteristic · Nearest neighbor analysis · Fractal dimension

List of Symbols

a	power order of solid fat content in Eqs. 4–7 (dimensionless)
b	coefficient in Eqs. 4–7 (dimensionless)
d_{min}	minimum size of fat crystal flocs in terms of equivalent diameter, 0.018 for model systems in this study (mm)
D	fractal dimension by particle counting method (dimensionless)
D_{L}	fractal dimension of microstructure interface boundaries (dimensionless)
D_{NN}	nearest-neighbor distance (mm)
G'	shear modulus in Eq. 2 (Pa)
l_0	zero-crossing length (mm)
x	microstructure factor in Eq. 1
δ_{n}	normalized microstructure density (mm^{-3})
ε	characteristic compressive modulus (Pa)
ϕ	solid-fat content (SFC) expressed in volume fractions between 0 and 1 (dimensionless)
γ	coefficient for correlation of rheological properties with solid fat content and microstructure factor in Eqs. 2 and 4–7 (unit depending on equations)
σ	deformation in compressive penetration test (%)
τ	force per unit area in compressive penetration test (Pa)

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Introduction

Semisolid lipid systems containing different (e.g. high-melting and low-melting) triacylglycerols (TAG) exist in many food products. The rheological, textural and functional properties of those fat systems are complex and influenced by many factors. This complex behavior is the result of the presence of a certain amount of solid crystalline fat, that forms a crystal network, and the rheological properties are directly related to the solid-fat content (SFC) of the systems [1]. It is generally accepted that processing conditions have significant impact on fat crystallization, and hence on aspects of structure development, at molecular and microscopic levels, and eventually govern the macroscopic properties [2–4].

Although SFC is a primary factor influencing rheological properties of a semisolid lipid system, two samples of a system with same SFC may have significantly different rheological properties due to their different microstructures, indicating that the microstructure of the system also has an impact on rheological properties [5]. Therefore, a rheological, mechanical or textural property (e.g., a modulus ε) of a fat is a function of SFC ϕ and some microstructural factor x.

$$\varepsilon = f(\phi, x) \tag{1}$$

Rheological properties of a semisolid fat system have been determined mainly by three methodologies: rheometrical, mechanical, or textural analyses. With a rheometer, samples are subject to a rotational shearing test and shear modulus G' is obtained from a relationship between shear stress and shear rate. With a texture analyzer or a mechanical analyzer, a variety of different tests including compression, spreading, bending, etc., can be performed and the corresponding modulus is determined from a relationship between stress and strain. These methods were successfully applied to fat property characterization in studies of lipid systems [6–8]. Like rheological properties, SFC of a lipid system is readily determined, in this case, by use of nuclear magnetic resonance (NMR). Thus, quantification of microstructure for a system with a fat crystal network becomes the key issue in determination of the relationship shown in Eq. 1.

Vreeker [9] suggested that fat crystal networks had a fractal nature and introduced the fractal dimension to quantify the relationship between the mass of a cluster and its size. In studies on colloidal gels, Shih et al. [10] developed a scaling theory where elastic constants for two regimes (strong and weak) of linkages between the flocs were defined. Recently, Marangoni and co-workers [11, 12] applied fractal concepts and scaling theory to viscoelastic lipid systems. According to this model, a relationship between shear modulus, G', and SFC ϕ may be expressed as

$$G' = \gamma \phi^{\frac{1}{3-D}} \tag{2}$$

where γ is a coefficient and D is fractal dimension representing the microstructural characteristics of the system. The number of particles in an image of fat crystal microstructure was found to be a linear function of fractal size after logarithmic transformation, with a slope of fractal dimension D. Thus, D can be obtained by use the of particle-counting method (PCM) via analysis of microscopic images for a fat crystal network system [12]. Correlations of the coefficient γ to parameters for primary particles, microstructures and intermicrostructural elements were further developed [13].

More recently, several other models for analyzing fat crystal microstructure were introduced and compared [14]. First, microstructure density, defined as the number of microstructure units per unit volume of the system, and its normalized value, provided a direct expression of the microstructure of a system. Second, the interface existing between microstructure units had a fractal nature and its fractal dimension was obtained by use of a Richardson plot [15] to describe the irregularity of the interface boundaries in a fat crystal network system in two-dimensional space. Third, the spatial point distribution based on the centroid of microstructure units was analyzed by use of Minkowski functionals [16] and a geometric algorithm of Dirichlet tessellation and Delaunay triangulation [17] to obtain Euler characteristic parameters and nearest-neighbor parameters, respectively, to quantitatively express the spatial relationship of fat crystals and crystal flocs. It was shown that each of these microstructural parameters correlated well with changes in the crystalline nature of model lipid systems.

In the current work, rheological and microstructural properties of semisolid model lipid systems containing high-melting and low-melting TAGs were quantitatively characterized. Empirical models to correlate rheological properties with SFC and microstructural characteristics of the fat crystal network were developed. One of the main aims of this study was to document the relative effects of SFC and microstructure on rheological properties.

Experimental

Materials

Model lipid systems were prepared from a mixture of low-melting and high-melting lipids. Purified sunflower oil (SFO) was used as a low-melting lipid with a melting point of −29.3 °C. Palm oil was fractionated to obtain a solid fraction (palm stearin, PS), which used as the high-melting lipid with a melting point of 56.4 °C. The TAG composition of these lipids was analyzed with a Hewlett–Packard (Wilmington,

Table 1 Triacylglycerol (TAG) composition of groups (g/100 g identified TAGs) of palm stearin (PS) and sunflower oil (SFO)

TAG group	PS	SFO
Short-chain, ≤C40	3.8	2.7
C42 + C44	0.1	0.0
Tri-saturated long-chain in C46–C54	37.7	0.1
Di-saturated long-chain in C46–C54	37.8	2.6
Mono-saturated long-chain in C46–C54	17.4	21.8
Tri-unsaturated long-chain in C46–C54	3.3	72.8

DE, USA) 5890 gas chromatograph (GC), with the major TAG groups shown in Table 1. SFO may be considered a long-chain unsaturated (18:2) TAG class, since its major TAGs, constituting about 73% of the mass, were LLL and LOL (L, linoleic; O, oleic). PS was a mixed tri-saturated and di-saturated fat, because its major TAGs, constituting about 93% of the mass, were PPP, POP and OOP (P, palmitic).

Processing

To prepare semisolid lipid samples, PS and SFO were mixed at different mass ratios (4:6, 5:5, 6:4, and 7:3). The mixtures were melted and stabilized at 80 °C for 1 h to destroy any crystal memory and Nile Red (Sigma, St Louis, MO, USA) was added at a level of 0.005% to provide labeling and contrast in the confocal scanning light microscopy (CSLM) images. The melt was cooled statically in a jacketed stainless steel beaker by circulated cooling water from a bath. As the sample reached the target crystallization temperature, which was dependent on high-melting to low-melting ratio, agitation was applied for 30 s to induce nucleation, followed by static growth of fat crystals at the same temperature. This primary crystallization lasted about 2 h. Then, slurry samples were placed on depressions with a thickness of 1.5–2.0 mm on microscope slides for imaging and also filled into texture analyzer sample cups and NMR tubes for analysis. Samples were stored in a temperature-controlled chamber at 0 °C for secondary crystallization for 24 h.

Effects of processing conditions on crystallization, and hence on SFC and microstructure, were studied in preliminary tests to determine the conditions for primary crystallization (temperatures, agitation intensity for induced nucleation, etc.) to obtain semisolid fat samples with similar SFCs but different microstructures for lipid mixtures of the same PS:SFO ratio. Mixtures containing more high-melting fat had higher supersaturation level than systems with less high-melting fat; therefore, to keep similar supersaturation level for systems with different mass ratio of PS to SFO, mixtures containing more PS were crystallized at higher temperatures. To form semisolid products with distinguishable microstructures, primary crystallization temperature and agitation speed for induced nucleation were set at different levels depending on the PS:SFO ratios. The crystallization temperatures were 35, 36, 39, and 40–47 °C for lipid mixture with PS:SFO ratios of 4:6, 5:5, 6:4, and 7:3, respectively. The agitation speed for induced nucleation also varied as needed, up to about 1,000 rpm, to generate samples with different microstructures for a given PS:SFO ratio and crystallization temperature. Note that the exact crystallization conditions are not critical since the goal was simply to create samples with a range of crystalline microstructures, SFC, and rheological properties. Three to five replicates were conducted for each experimental condition (crystallization temperature and agitation rate).

Characterization of Semisolid Lipid Samples

Microstructural images of semisolid fat samples were obtained by use of a Bio-Rad 1024 CSLM system (Bio-Rad Laboratories, Hemel Hempstead, UK) with a Nikon Eclipse Microscope (Nikon, Melville, NY, USA). Samples on the microscope slide were scanned at a temperature of 0 °C. A 4× objective was employed and the frame of acquired images had a dimension of 512 × 512 pixels or 3.367 × 3.367 mm, which gave sufficient resolution for microstructural comparisons [14]. Based on the CSLM images, quantification of the microstructure of these model lipid mixture samples was performed using different approaches, described in our previous work [14]. These included microstructure density, δ_n, Euler characteristic (zero-crossing length, l_o), nearest-neighbor distance, D_{NN}, fractal dimension of microstructure interface, D_I, and fractal dimension by the PCM, D.

SFC of duplicate samples after storage for 24 h at 0 °C was determined with a Bruker minispec pc-120 NMR system (Bruker, Milton, Ontario, Canada).

A TA-XT2 (Stable Micro Systems, Surrey, UK) texture analyzer was used to characterize the rheological properties of the semisolid fat samples after the secondary crystallization. The texture analyzer sample cup had a top diameter of 46 mm, a bottom diameter of 30 mm, and a height of 30 mm. A plastic cylindrical probe with a diameter of 12.7 mm was used for a compressive penetration test to a depth of 10 mm with a speed of 3.0 mm/s. Triplicate tests for each sample were performed at 0 °C and force versus distance/time was recorded, from which force applied to unit area versus deformation were obtained.

A Pareto chart was constructed to document the role of each affecting factor on the rheological properties of these semi-solid fat systems. According to this statistical approach [18], the magnitude of the effect of SFC or microstructure property (factor) on the rheological properties (response) is the absolute value of the difference between average of response to a high level of the factor and average of response to a low level of the factor. Thus, a larger Pareto effect indicates that the variation of a factor results in a larger change of response. In addition, correlation analysis was performed on the data (Microsoft Excel Analysis ToolPak [19]) to obtain correlation coefficients which indicated whether certain parameters were related or not.

Results and Discussion

A typical relationship between force per unit area (τ) and deformation (σ) for compressive penetration is shown in Fig. 1. In all samples, a linear region was observed during the early stage of the test and a maximum value usually occurred near the end of the test when, for certain samples, the plastic fat sample collapsed. The maximum force per unit area might be regarded as a characteristic parameter of rheological properties for the samples. However, in this study, the characteristic compression modulus ε, was obtained from the slope of the linear region.

$$\varepsilon = \frac{\Delta\tau}{\Delta\tau} \tag{3}$$

The parameter ε was used to represent the rheological characteristics of those systems for analysis of relationships among rheological, phase, and microstructural properties. Values of ε for each of the lipid systems are given in Table 2.

As examples of typical CSLM microstructure, images of the model lipid systems with different PS:SFO ratios of 5:5 and 7:3 with different microstructure densities (low, medium, and high) are shown in Fig. 2. The fat crystal network in terms of microstructure interface boundaries are also shown. For a given PS:SFO ratio, different microstructure densities were obtained by changing the agitation rate and extent during primary crystallization. Mean particle size decreased when agitation extent increased, as seen in the images from top to bottom in Fig. 2. Also, for a given composition (PS:SFO of 5:5 or 7:3) mean crystal size and, hence, the size of microstructure units, was very different, which led to different microstructure densities. However, systems with different composition (e.g., PS:SFO of 5:5 versus 7:3) could have very similar microstructure densities, as shown in the same row of images.

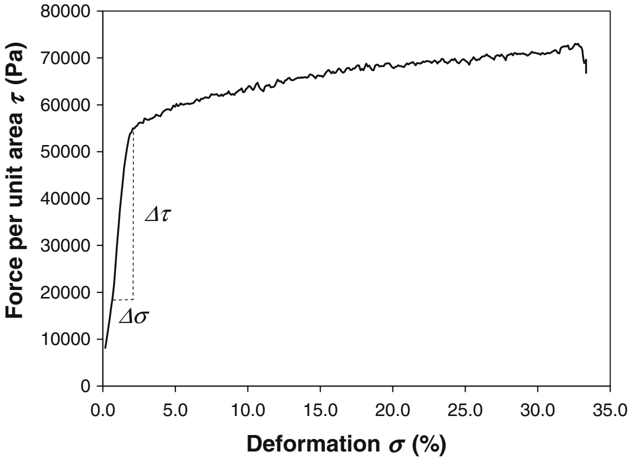

Fig. 1 Typical relationship between force per unit area (τ) and deformation (σ) for the compressive penetration test

SFC, Microstructure, and Compression Modulus

Microstructural properties of CLSM images for each system were analyzed according to the methods described in previous work [14]. Table 2 summarizes the average values of the microstructural parameters for the different model lipid systems, along with the experimentally determined SFC and compression modulus. An increase of 10% high-melting fat led to an increase of about 8% SFC in the final products over the range studied. Also, for a certain mass ratio of PS:SFO, the SFC of systems undergoing crystallization at different agitation rate were very similar with a coefficient of variation smaller than 3%.

From Table 2, SFC and the corresponding compression modulus are significantly different for different lipid systems with different PS:SFO ratios. On the other hand, SFC values in a group with the same PS:SFO ratio were almost the same, whereas compression modulus and microstructural parameters were significantly different. This was confirmed by analysis of variance (ANOVA) at 95% confidence, which resulted in p values less than 0.05 for microstructure density, fractal dimension of microstructure interface, zero-crossing length, nearest neighbor distance, and compression modulus.

These results show that SFC has a significant effect on rheological property, although the impact from microstructural factors cannot be neglected. A Pareto chart (Fig. 3) describing the magnitude of the effect from different factors, as generated by statistical analysis, documents the relative importance of each parameter. As seen in Fig. 3, SFC has the largest effect on compression modulus, but the microstructural properties also contribute a significant effect. The four microstructural parameters (δ_n, D_L, l_o, and D_{NN}) each have approximately the same level of significance on compression modulus.

To further clarify the relative effects of SFC and the crystalline microstructure parameters, correlation analysis was performed between compression modulus and SFC or microstructural parameters for all systems. Correlation analysis (Table 3) was performed on the entire data set,

Table 2 Average values of solid-fat content (ϕ), compression modulus (ε), and microstructural parameters for different ratios (4:6, 5:5, 6:4, and 7:3) of palm stearin (PS) and sunflower oil (SFO)

PS:SFO	MDL	ϕ	$\varepsilon \times 10^{-4}$ (Pa)	δ_n^a (mm^{-3})	l_0^a (mm)	D_L^a	$D_{NN}{}^a$ (mm)	D^a	Image
4:6	VL	0.276 ± 0.003	0.543 ± 0.126	34 ± 10	0.2075 ± 0.0032	1.087 ± 0.004	0.2576 ± 0.0364	2.021 ± 0.184	
	VL	0.266 ± 0.001	1.014 ± 0.599	44 ± 1	0.1738 ± 0.0208	1.093 ± 0.005	0.2120 ± 0.0022	2.097 ± 0.072	
	L	0.278 ± 0.001	1.853 ± 0.582	54 ± 15	0.1594 ± 0.0025	1.101 ± 0.002	0.2004 ± 0.0282	2.019 ± 0.205	
	M	0.283 ± 0.006	3.473 ± 0.381	168 ± 15	0.0928 ± 0.0314	1.124 ± 0.010	0.1192 ± 0.0052	1.990 ± 0.081	
5:5	VL	0.346 ± 0.002	3.011 ± 0.508	34 ± 7	0.1958 ± 0.0009	1.092 ± 0.003	0.2443 ± 0.0239	2.109 ± 0.288	
	L	0.348 ± 0.005	3.596 ± 0.121	53 ± 7	0.1666 ± 0.0007	1.104 ± 0.006	0.2161 ± 0.0160	1.932 ± 0.135	b
	M	0.357 ± 0.000	4.121 ± 0.589	120 ± 30	0.1233 ± 0.0044	1.112 ± 0.007	0.1581 ± 0.0161	2.002 ± 0.083	b
	H	0.367 ± 0.005	9.431 ± 0.910	244 ± 4	0.0795 ± 0.0105	1.142 ± 0.011	0.0998 ± 0.0010	1.997 ± 0.131	b
	H	0.367 ± 0.006	8.969 ± 1.962	252 ± 8	0.0821 ± 0.0075	1.145 ± 0.004	0.1038 ± 0.0009	2.023 ± 0.089	
6:4	L	0.433 ± 0.001	9.218 ± 1.580	82 ± 19	0.1512 ± 0.0021	1.103 ± 0.006	0.1868 ± 0.0235	1.957 ± 0.071	
	M	0.447 ± 0.002	11.312 ± 1.879	194 ± 12	0.0952 ± 0.0022	1.127 ± 0.004	0.1180 ± 0.0022	1.953 ± 0.035	
	H	0.429 ± 0.014	13.516 ± 1.381	309 ± 44	0.0779 ± 0.0181	1.145 ± 0.007	0.1013 ± 0.0116	2.016 ± 0.070	
7:3	L	0.507 ± 0.000	14.127 ± 0.890	50 ± 7	0.1739 ± 0.0016	1.096 ± 0.002	0.2139 ± 0.0119	2.094 ± 0.184	
	L	0.499 ± 0.002	16.929 ± 1.736	86 ± 2	0.1111 ± 0.0028	1.105 ± 0.007	0.1399 ± 0.0013	2.004 ± 0.150	b
	M	0.501 ± 0.001	22.439 ± 3.044	118 ± 22	0.1141 ± 0.0096	1.136 ± 0.004	0.1432 ± 0.0128	1.915 ± 0.085	b
	H	0.511 ± 0.000	26.308 ± 1.426	246 ± 12	0.0773 ± 0.0068	1.149 ± 0.009	0.1007 ± 0.0070	1.937 ± 0.044	b
	VH	0.510 ± 0.004	30.261 ± 1.659	357 ± 17	0.0733 ± 0.0022	1.176 ± 0.010	0.0923 ± 0.0024	1.992 ± 0.075	

MDL microstructure density level, *VL* very low, *L* low, *M* medium, *H* high; *VH* very high

[a] δ_n, normalized microstructure density; l_0, Euler zero-crossing length; D_L, fractal dimension of microstructure interface boundaries; D_{NN}, nearest neighbor distance; D, fractal dimension by particle counting method

[b] Example images shown in Fig. 2

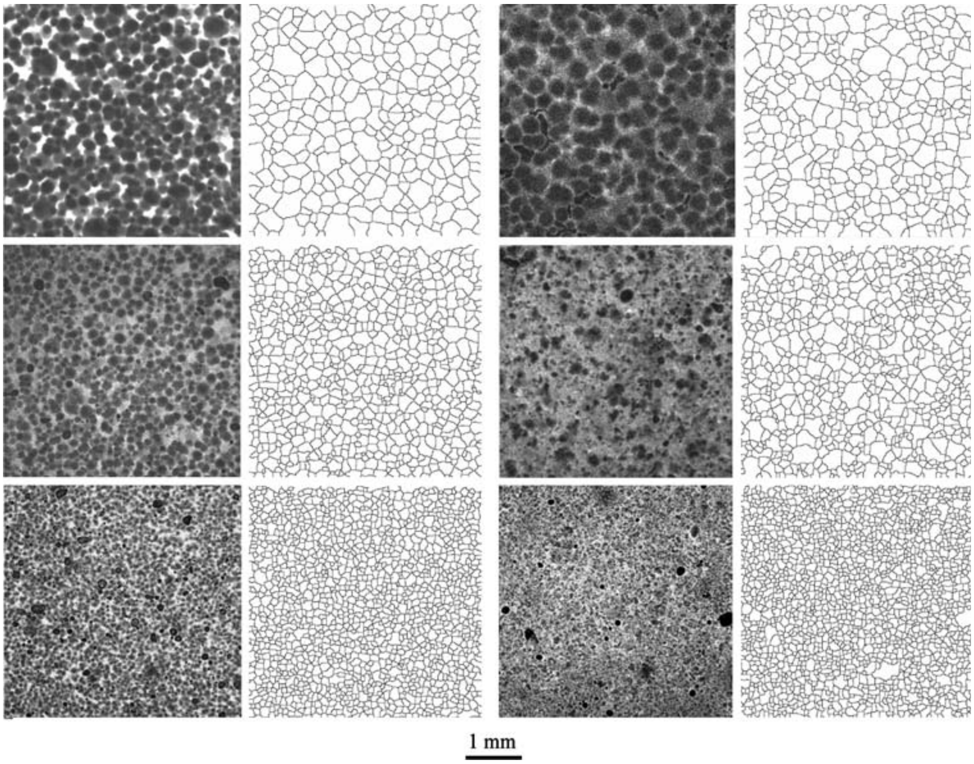

Fig. 2 Example images of model lipid systems (palm stearin (PS), and sunflower oil (SFO)) having different microstructural characteristics with the original CLSM image on the *left* and the processed image for microstructure units with interface boundaries on the *right*: *left systems* PS:SFO = 5:5 and *right systems* PS:SFO = 7:3. *Top*, *middle*, and *bottom rows* represent low, medium, and high microstructure densities, respectively. For average solid-fat content, compressive modulus and microstructural parameters of each system, refer to Table 2. Frame dimension: 3.367 × 3.367 mm

and for systems in a group having the same PS:SFO ratio (thus, the same SFC). In terms of all systems, compression modulus is highly dependent on SFC with a correlation coefficient of about 0.9, a much higher effect than for crystal microstructure. However, very high correlations (mostly higher than 0.9, except fractal dimension by PCM)

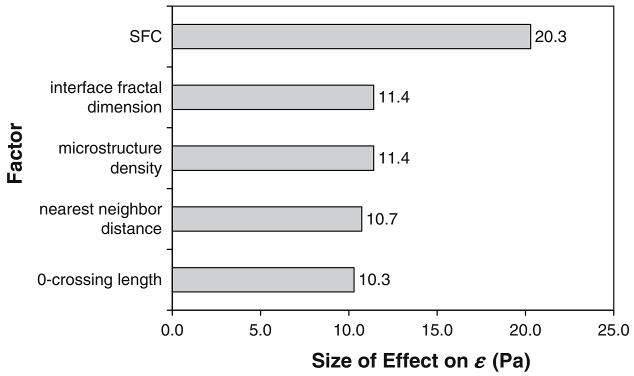

Fig. 3 Pareto chart showing the relative magnitudes of factor effect of different microstructure on compressive modulus

were found for systems with the same SFC. Again, SFC is the most important parameter affecting mechanical properties, but for systems with the same SFC, the microstructural characteristics are also important to predicting compression modulus. Relatively low and inconsistent correlation coefficients between compression modulus and fractal dimension by PCM (Table 3) indicate that this measure of microstructure does not correlate very well with compression modulus. This result is consistent with our previous work [14].

Relationships Among Modulus, SFC, and Microstructure Characteristics

As indicated in Table 3, for systems with constant SFC, there is very good correlation between compression modulus and the microstructural properties. To quantify these relationships, nonlinear regression was performed by use of the SAS statistical analysis system according to the following empirical equations.

$$\varepsilon = \gamma \phi^a \delta_n^b \tag{4}$$

$$\varepsilon = \gamma \phi^a \frac{1}{e^{b(2-D_L)}} \tag{5}$$

$$\varepsilon = \gamma \phi^a \frac{1}{e^{b(l_0-d_{\min})}} \tag{6}$$

$$\varepsilon = \gamma \phi^a \frac{1}{e^{b(D_{NN}-d_{\min})}} \tag{7}$$

The $d_{\min}$ value, the equivalent diameter of the minimum fat crystal flocs, was found to be 0.018 mm from image analysis of the PS:SFO system studied in our previous work [14]. Note that Eqs. 4–7 are empirical correlations that were found to provide reasonable fits to the data although they have little, if any, fundamental meaning. Nevertheless, such correlations are useful models for characterizing relationships and predicting results. These correlations also help to document the magnitude of each effect on rheological properties.

The parameters γ, a and b were obtained from the SAS nonlinear regression analysis and their values are shown in Table 4. These relationships, which are shown graphically in Figs. 4–7, show that compression modulus is a function of two independent variables, SFC and a microstructural parameter. Model-predicted lines based on average SFC for Eqs. 4–7 show excellent fits to the experimental data.

Equations 4–7 can be regarded as a combination of three parts, a coefficient γ, a power law term for SFC with a power order of a, and a term containing a microstructural parameter with a corresponding coefficient b. The coefficient, γ, or the maximum modulus as SFC approaches unity (volume fraction), depends on the nature of components of a system (i.e., density and hardness of fat crystals, viscosity of liquid phase, interaction characteristics between the two phases, etc.) [12, 13]. The value of the exponent for SFC, a, represents the magnitude of the effect of a change in SFC on compression modulus. Higher values of a indicate a

Table 3 Correlation coefficients between compression modulus (ε), solid-fat content (ϕ), and microstructure for different ratios (4:6, 5:5, 6:4, and 7:3) of palm stearin (PS) and sunflower oil (SFO)

System	All systems	In group			
		PS:SFO = 4:6	PS:SFO = 5:5	PS:SFO = 6:4	PS:SFO = 7:3
SFC	–	0.276 ± 0.007	0.357 ± 0.010	0.434 ± 0.009	0.505 ± 0.006
ϕ	0.898	0.708	0.954	−0.222	0.594
δ_n^a	0.620	0.954	0.977	1.000	0.956
l_0	−0.615	−0.987	−0.920	−0.952	−0.891
D_L	0.735	0.998	0.980	0.994	0.995
D_{NN}	−0.622	−0.979	−0.930	−0.939	−0.898
D	−0.622	−0.612	−0.122	0.846	−0.613

[a] See Table 2 for description of microstructure parameters

Table 4 Relationships among compression modulus (ε), solid-fat content (ϕ), and microstructural parameters for mixtures of palm stearin (PS) and sunflower oil (SFO) based on SAS non-linear regression at the 95% confidence level

Coefficient ± standard error				
Equation no.	Relationship	γ	a	b
4	δ_n^a	$(5.76 \pm 1.90) \times 10^5$	3.91 ± 0.30	0.34 ± 0.04
5	D_L	$(4.20 \pm 2.98) \times 10^9$	3.47 ± 0.24	8.69 ± 0.87
6	l_0	$(5.29 \pm 1.27) \times 10^6$	3.78 ± 0.33	7.06 ± 1.09
7	D_{NN}	$(5.59 \pm 1.34) \times 10^6$	3.77 ± 0.32	5.90 ± 0.89

[a] See Table 2 for description of microstructure parameters

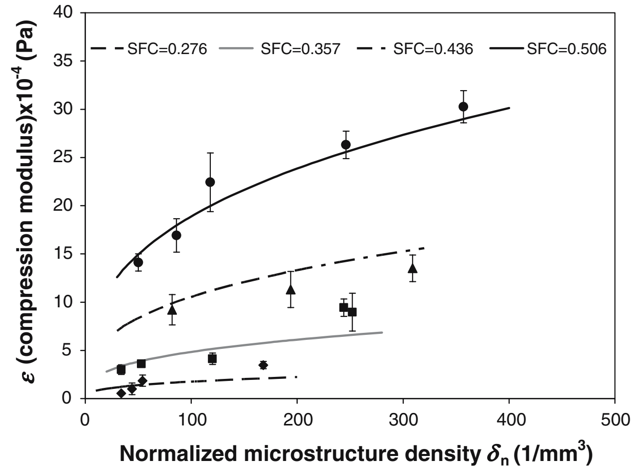

Fig. 4 Relationship between compressive modulus and normalized microstructure density for model systems with different solid-fat content (SFC). The *lines* represent the best fit to Eq. 4

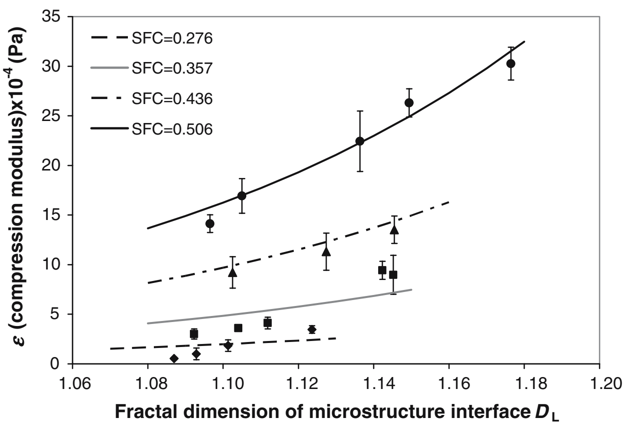

Fig. 5 Relationship between compressive modulus and fractal dimension of microstructure interface boundaries for model systems with different solid-fat content (SFC). The *lines* represent the best fit to Eq. 5

larger effect. The microstructural coefficient, b, is an empirical representation of the effect of a particular property on the compression modulus.

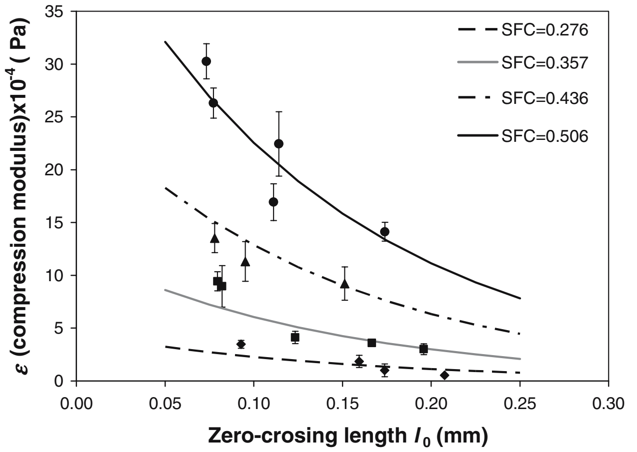

Fig. 6 Relationship between compressive modulus and zero-crossing length for model systems with different solid-fat content (SFC). The *lines* represent the best fit to Eq. 6

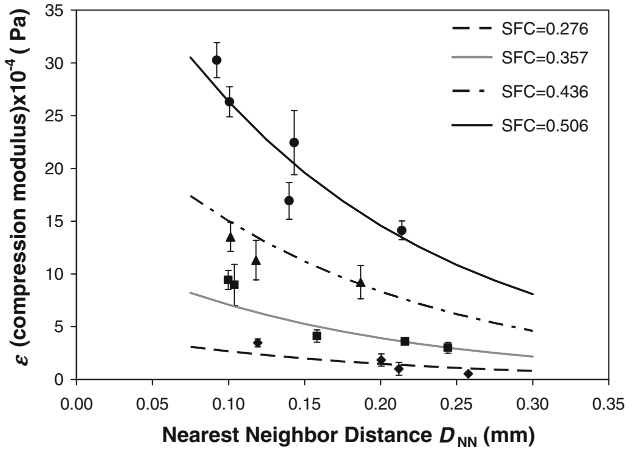

Fig. 7 Relationship between compressive modulus and nearest neighbor distance for model systems with different solid-fat content (SFC). The *lines* represent the best fit to Eq. 7

In Eq. 4, the power order b reflects the extent of the effect of microstructure density on compression modulus. Since both SFC and microstructure density impact compression modulus through an exponential model, the term with the higher power order has the larger effect on modulus. In the systems studied in this work (Table 4), power order a is more than ten times higher than power order b. Thus, the effect of SFC on compression modulus is much larger than that of microstructure for these systems. In addition, systems with higher microstructure density have a greater effect on the compression modulus for a given SFC. However, as discussed in our previous work [14], microstructure density, and hence normalized microstructure density, is limited by an ideal microstructure density that depends on average size of fat crystal flocs.

In Eqs. 5–7, the values of coefficient b also signify the extent of effect from the microstructural parameter (D_L, l_0 or D_{NN}) on compression modulus. Unlike the power order in Eq. 4, a larger b value for a system indicates the corresponding microstructural parameter has smaller effect on rheological property than that for a system with smaller b value. Since fractal dimension D_L represents irregularity of microstructure interface boundaries in two-dimensional space, its value is larger than unity. Higher D_L indicates microstructure interface boundaries are more irregular due to closer particles, which suggests a stronger interaction between particles and hence, a higher compression modulus. Equations 6 and 7 have the same mathematical form and quite similar values of coefficients, indicating zero-crossing length and nearest neighbor distance are very close in terms of quantitatively representing microstructure and correlating with the rheological properties. As l_0 or D_{NN} increases, compression modulus decreases because interactions are reduced due to larger distance between particles.

In this work, the relative effects of SFC and lipid crystal microstructure on mechanical properties have been documented for a model lipid system. Microstructures of natural lipid systems vary significantly depending on formulation and processing conditions for different lipid mixtures, resulting in differences in mechanical properties. In these systems, SFC had the dominant effect on mechanical properties. However, microstructure also played an important role, especially for systems with identical SFC. Empirical equations were shown to model reasonably well the relationships between SFC, microstructural parameters and mechanical properties. These models could be used to empirically predict mechanical properties once microstructural elements were quantified.

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