The Difference Between Cocoa Butter and Salatrim® Lies in the Microstructure of the Fat Crystal Network

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ABSTRACT: The solid fat content of Salatrim® at 22°C is 9% higher than that of cocoa butter; however, its storage modulus, G′ (1 Hz), or solid-like character, is one order of magnitude lower (2.5 MPa vs. 52 MPa). This difference may be explained by structural differences in the microstructure of the fat crystal network of the respective fat systems. Polarized light micrographs of these two confectionery fats show that cocoa butter forms a fat crystal network characterized by discrete crystalline particles which aggregate to larger microstructures which then form a three-dimensional network, while Salatrim®'s network is composed of randomly arranged noncrystalline, translucent platelets. Rheological measurements on both fat networks yielded fractal dimensions of 2.37 for cocoa butter and 2.90 for Salatrim®. Image analysis of the microstructure of cocoa butter yielded a fractal dimension of 2.31; however, the microstructure of Salatrim® does not lend itself to fractal analysis *via* image analysis. It was observed that the microstructure of Salatrim® is random instead of fractal. The proposition is made that the macroscopic mechanical properties of Salatrim® are related to the mechanical properties of the platelets that make up the network and the nature of the links between the platelets. Furthermore, the random spatial distribution of the platelets does not provide an indication of the strength of the network. For cocoa butter, the macroscopic mechanical properties are integrally related to the fractal spatial distribution of the solid mass in the network, the nature of the links between the microstructures, and the strength of the microstructures.

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The melting profiles of cocoa butter and Salatrim® (obtained from Cultor Food Science, Inc., Ardsley, NY) are almost identical (Fig. 1); however, their functionality is very different. Physical indicators of functionality such as melting points, melting profiles, and solid fat contents (SFC) at different temperatures do not explain the differences in physical properties of these fats, nor do they offer insights into the macroscopic behavior of these fats. Since the usual physical indicators do not offer insight into the origin of the macroscopic differences between these two fat systems, we propose that the difference lies in the structural characteristics of these systems at the microstructural level.

The enormous influence of the microstructure of fat crystal networks on the macroscopic properties of fats is not a new concept, and in fact was recognized as early as 1987 (1). Heertje's work (2) on the visualization of the fat crystal network in plastic fats remains one of the most important contributions to this field. Furthermore, work in our laboratory on the effects of chemical and enzymatic interesterification on the physical properties of milk fat, palm oil, and lard showed that the microstructure of the fat crystal network plays a key role in determining the rheological properties of plastic fats (3).

The importance of the microstructural level of structure in fat crystal networks can only be understood when placed in the context of the roles the other levels of structure play in the determination of the macroscopic properties of the fat crystal network. The different levels of structure are defined during the formation of the fat crystal network from the melt. The growth of a typical confectionery fat crystal network such as cocoa butter can be visualized thus: the triglycerides present in the sample crystallize from the melt into particular polymorphic/polytypic states. These crystals then aggregate *via* a mass- and heat-transfer limited process into larger mi-

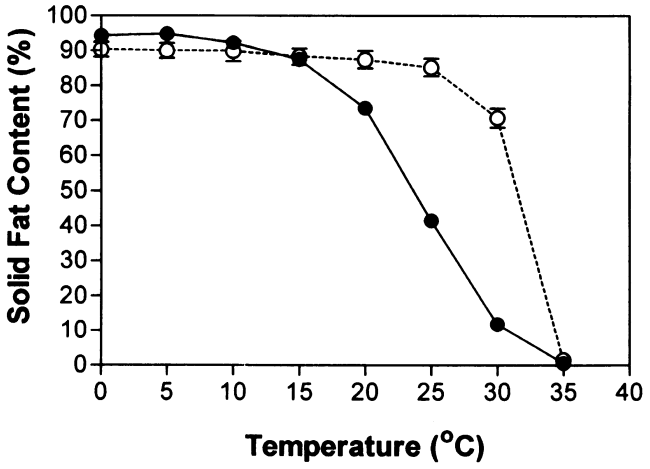

FIG. 1. Melting profiles of cocoa butter (●) and Salatrim® (Cultor Food Science, Inc., Ardsley, NY) (○).

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crostructures. The aggregation process continues until a continuous three-dimensional network is formed. The macroscopic rheological properties of the network are influenced by all levels of structure defined during the formation of the network—i.e., the structure of the individual triglycerides, the structure of the individual crystalline units formed, the polymorphic nature of the network, and particularly by the microstructural level of structure, since the microstructural level is closest to the macroscopic world. Any perturbation of the fat crystal network is liable to affect the microstructural organization before any other level of structure.

The study of crystal polymorphism and morphology has increased our understanding of the properties of cocoa butter (4–8). However, the *in situ* microstructure of the fat crystal network had never been visualized or quantified before some recent advances in our laboratory (9,10). Hicklin *et al.* (6) showed that crystal morphology cannot easily be predicted from knowledge of the polymorphic state of cocoa butter triacylglycerides (TAG). Kellens *et al.* (11) showed that the same crystal polymorphic forms in tripalmitin can give rise to different microstructures. This points to the fact that the relationship between polymorphism and macroscopic properties is, at best, of an indirect nature. The theoretical prediction of polymorphism from triglyceride composition remains a daunting task that has not been undertaken because of the variety of variables involved in a typical confectionery fat. Therefore, consideration of the microstructural level of structure in the determination of the macroscopic properties of fat crystal networks is of crucial importance. Obviously, the complete picture would necessarily be formed by considering all levels of structure and their interdependence. The goal is to quantitatively link triglyceride structure, polymorphism, and microstructure. The melting profiles of fat crystal networks are related to triglyceride structure and polymorphism, since the melting of the network results in complete breakage of these levels of structure. However, rheological measurements performed on the fat crystal network, especially in the linear viscoelastic region of the network, do not result in the breakage—or, sometimes, even stressing—of levels of structure below the level of the microstructure. Therefore, it is conceivable that fats with almost identical melting profiles can have very different rheological characteristics if their microstructures are sufficiently different.

In this research paper, differences in the microstructure of the fat crystal network between cocoa butter and Salatrim® are reported. We propose that differences in the macroscopic rheological properties between these two fats can be explained by the drastic differences in their fat crystal network microstructures. This has important implications in the manufacture of confections.

MATERIALS AND METHODS

Materials and sample preparation. Mixtures of cocoa butter and Salatrim® with canola oil were prepared in 5% (w/w) increments, from 100 to 75% (w/w) confectionery fat/canola oil. The confectionery fats were melted above 80°C to destroy any crystal memory, mixed with canola oil and stirred until thoroughly mixed.

SFC determination. SFC was determined with a Bruker PC/20 series NMR Analyzer (Bruker, Milton, Ontario, Canada). The blends were introduced into nuclear magnetic resonance tubes, chilled in a refrigerator for 1 h at 5°C and allowed to crystallize for 48 h at room temperature (21–23°C). One determination on each of two replicate tubes was performed ($n = 2$). The averages and standard deviations are reported.

Polarized light microscopy (PLM). A few drops of melted (80°C) samples of 85% (w/w) cocoa butter and Salatrim® in canola oil were placed on a microscope slide preheated to 80°C. A coverslip preheated to the same temperature was immediately placed on top of the sample. Care was taken to create an intermediate thickness film—thin enough to be able to observe the structure, but not so thin as to create artifacts (5). Care was taken to ensure that the plane of the coverslip was parallel to the plane of the slide, so that the thickness of the sample was uniform (10). The slides were chilled in a refrigerator for 1 h at 5°C and allowed to crystallize for 48 h at room temperature (21–23°C). Under polarization, the anisotropic solid phase of the network refracts light in a different manner than the isotropic liquid phase. The liquid phase appears black, while the solid phase is colored. Grayscale PLM photographs of cocoa butter/canola oil and Salatrim®/canola oil prepared as described above are shown in Figures 2A and 2C. The solid phase appears as white or a shade of grey in these photographs. The entire solid mass of the thin samples is represented in the pictures, although some of the solid entities are out of focus due to the depth of the sample. A dynamic thresholding method (10) was used to create thresholded images of the grayscale photographs of the samples. Thresholded photographs render the image into two colors—white for the solid phase, black for the liquid phase. Figures 2B and 2D show the thresholded images for cocoa butter/canola oil and Salatrim®/canola oil mixtures, respectively. The thresholded images give a better representation of the total number of different solid microstructural elements present in the solid sample, and of the spatial distribution of these microstructural elements.

Dynamic rheological testing. The rheological measurements were performed using a CarriMed CSL^2 500 Rheometer (TA Instruments, Mississauga, ON, Canada) with a 2-cm parallel plate attachment. One of the attachment plates is a Peltier plate, which allows the samples to be analyzed at specific temperatures (in this case, our samples were analyzed at 20°C). Liquefied blends of cocoa butter and Salatrim® with canola oil were poured into molds in order to ensure uniform diameter and thickness for the samples. The diameter of the resulting samples was the same as the diameter of the attachment plates on the CarriMed machine. The thickness of the sample is an important parameter, since a too-thick sample makes it impossible to attain a uniform strain field through

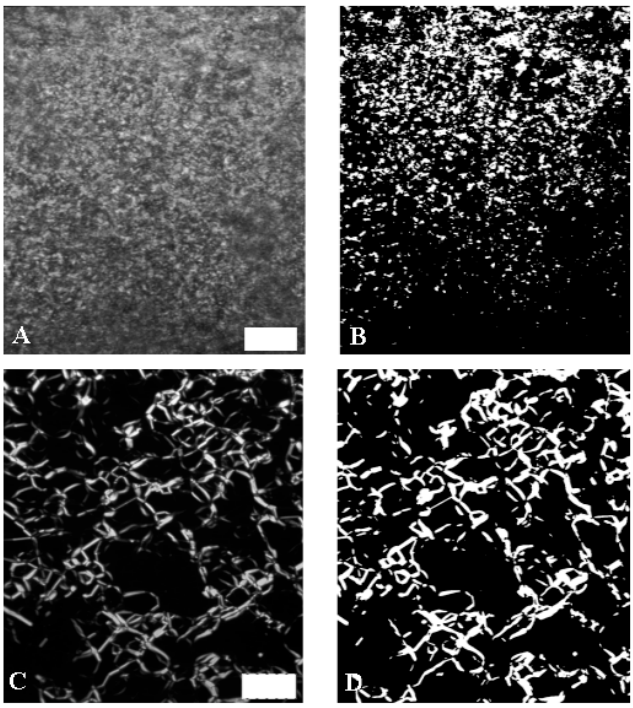

FIG. 2. Polarized light micrograph grayscaled images of (A) 85% w/w cocoa butter/canola oil and (C) 85% w/w Salatrim®/canola oil. (B) and (D) show thresholded images of (A) and (C), respectively. The horizontal length of the inset bar represents 0.05 mm.

the sample, while a too-thin sample results in interference due to particulates. We used a sample thickness of 3200 μm. Once in the molds, the lipid material was allowed to crystallize for 1 h at 5°C, followed by 48 h at 21–23°C. Samples were then removed from the molds, placed on a tray, and stored at 18–20°C immediately prior to analysis.

For Salatrim®, to prevent slippage between the sample surface and the surface of the sample attachment of the CarriMed, 50 grit sandpaper was attached to both the sample attachment plate and the surface of the Peltier plate with Krazy Glue™ (Borden, Inc., Columbus, OH). The sample was then compressed to approximately 10% of its original thickness, utilizing the software interface of the CarriMed. The compression force was applied in an exponentially increasing manner at a rate of 50 mm/s. The reason for this compression was to ensure that the sandpaper was thoroughly embedded into the sample, thereby preventing surface slippage.

For cocoa butter, each sample was glued directly onto the attachment plate and the Peltier plate with Krazy Glue™. This was done by removing the attachment plate from the machine, turning its flat surface upward, applying a small amount of glue, and carefully laying the sample onto it. Once the glue was cured, the attachment plate was reattached to the machine. A small amount of glue was then applied onto the Peltier plate after the instrument had set its bias. The ram was then quickly raised so that it just made contact with the sample. Since there was no compression of the sample, reproducibility was greatly increased. Unfortunately, Krazy Glue™ does not stick to Salatrim®, and therefore, sandpaper had to be used for this fat, as described above.

After the samples were mounted according to the different methods described above, the rheometer was run through an oscillatory stress program, with applied stresses ranging from 1 to 31800 Pa at a frequency of 1 Hz. This stress program was performed in order to determine the boundaries of the linear viscoelastic region (LVR). One has to exercise caution in the determination of the LVR, since the instrument response must be 100% in order to obtain an accurate value. For the CarriMed CSL^2 500, a strain of approximately 0.02% is the minimal detection limit. On establishing the LVR, frequency sweeps were carried out over a frequency range of 0.1 to 10 Hz at a strain level of 0.2% for cocoa butter and 0.5% for Salatrim®.

Data analysis. Apparent storage G′ and loss G″ moduli values were obtained from both the stress sweeps and the frequency data. Two determinations on each of two replicate samples were performed ($n = 2$). The averages and standard deviations are reported.

Fractal analysis. Fractal dimension values were derived from the slopes (m) of the log-log plots of the storage modulus (G′) against the SFC, as described by Vreeker *et al.* (12) and Marangoni and Rousseau (3). The weak link regime of Shih *et al.* (13) was assumed for our high SFC fats. The parameter γ was derived from the *y* intercept of the log-log plot of G′ vs. SFC. This parameter has been related to the Young's modulus of the individual particles that make up the network (14).

Fractal dimensions were also determined from image analysis of the PLM image of cocoa butter, using a method developed by Narine and Marangoni (10). In brief, this method assumes a model in which the microstructural level of structure of the fat crystal network is composed of individual microstructural elements forming a three-dimensional heat- and mass-transfer limited aggregate. The fractal dimension calculated from the PLM image of the microstructural network is a measure of the spatial distribution of the microstructural elements.

RESULTS AND DISCUSSION

The SFC of Salatrim® and Salatrim®/canola oil blends is about 9% higher than that of cocoa butter and cocoa butter/canola oil blends (Fig. 3). Since the shear storage modulus of a plastic fat is essentially a measure of its elasticity, or solid-like character, from these results one would expect the shear storage modulus (G′) to be higher in Salatrim® than in cocoa butter. However, as shown in Figure 4A, the G′ of cocoa butter is one order of magnitude higher than that of Salatrim® (Fig. 4B). Although sandpaper was utilized during the rheological analysis of Salatrim®, but not during the analysis of cocoa, we found that the rheological results for cocoa butter are the same regardless of whether sandpaper is used. This would seem to indicate that the difference in the

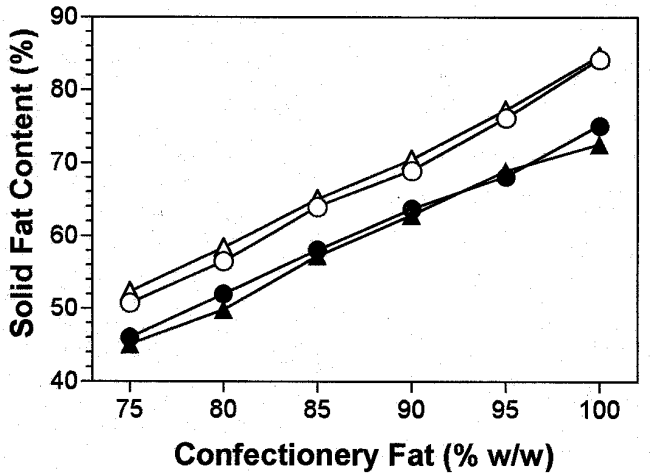

FIG. 3. Solid fat content of cocoa butter and Salatrim® blends with canola oil. Samples were crystallized statically at 5°C for 1 h, followed by room temperature (21–23°C) incubation for 24 or 48 h. (●) cocoa butter after 24 h; (○) Salatrim® after 24 h; (▲) cocoa butter after 48 h; (△) Salatrim® after 48 h.

values of G′ are not due to differences in the way the sample was analyzed, i.e., with or without sandpaper. As well, the ratio of the loss modulus to the storage modulus, or the ratio of the viscous to the elastic components (G″/G′), in Salatrim® is twice that of cocoa butter (Fig. 4C). Cocoa butter can hold very high amounts of liquid oil within its structure (>50%). This characteristic may be described as a very high "oil trapping capacity." Salatrim® can barely hold 8% oil within its crystal network, and hence displayed a low oil trapping capacity. One would expect that if the difference in the rheological measurements of Salatrim® and cocoa butter was due to differences in viscous drag, as opposed to differences in solid network, the observed ratios of G″/G′ would be smaller for Salatrim®. Therefore, the differences in shear storage modulus may be attributed to the differences in the solid phase of the network. We have already indicated the similarity in melting profiles of these two fats, as well as the inability of differences in SFC to explain the differences in the elastic nature of these two fats. Therefore, we propose that the differences are linked to the enormous differences in the microstructures of these fats.

PLM of Salatrim® and cocoa butter showed striking differences in crystal network characteristics (Figs. 2A, B, C, and D). At low magnification (100×) cocoa butter is composed of large particles, some as large as 120 µm (image not shown). A dense core is surrounded by less tightly packed crystalline material. This morphology was observed before (5). Imaging under a PLM at high magnification (400× and 1000×) shows that these large entities are made up of an aggregation of smaller crystalline microstructural elements with an average diameter of 5 µm. The large particles seen under low magnification are the microstructures formed by aggregation of the small crystalline microstructural elements. The addition of canola oil did not alter the morphology of cocoa

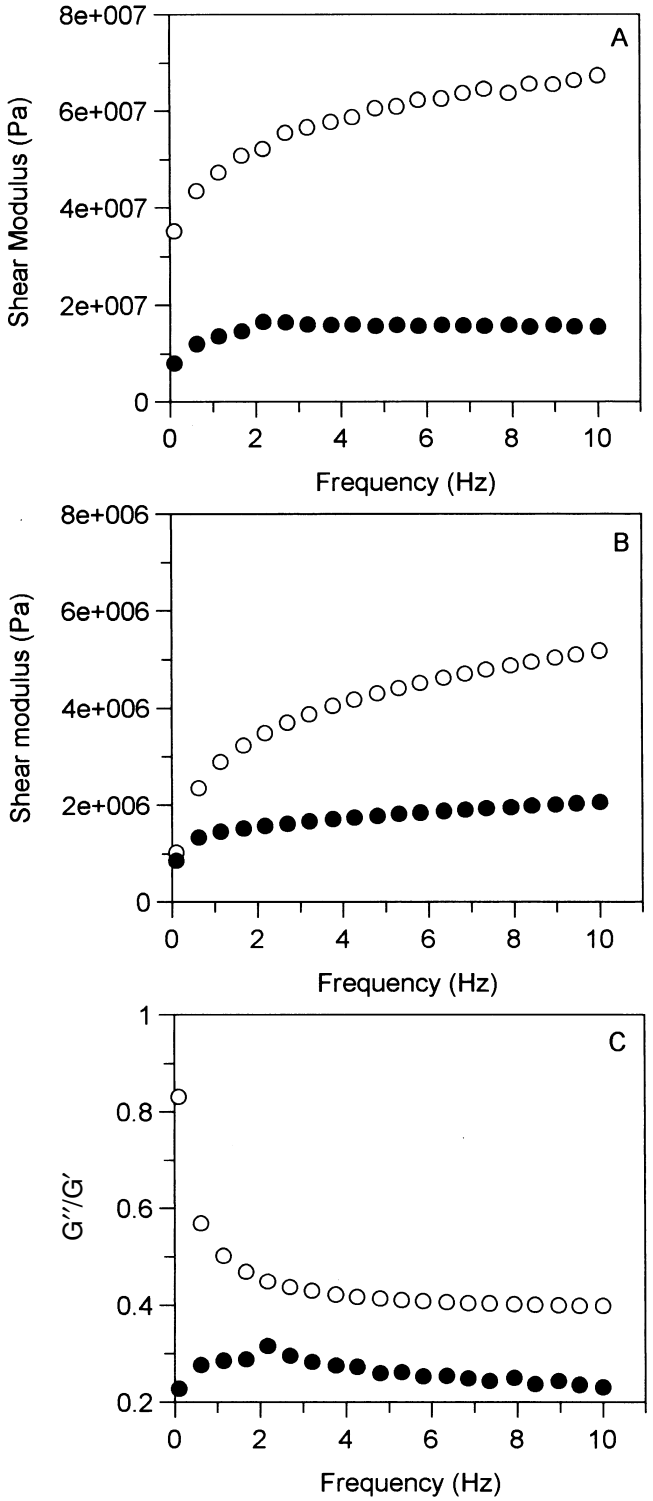

FIG. 4. Dynamic oscillatory rheological analysis. Frequency dependence of G′ and G″ of (A) cocoa butter at a 0.2% strain level and (B) Salatrim® at a 0.5% strain level. The symbol (●) corresponds to G″, while the symbol (○) corresponds to G′. (C) Frequency dependence of the tanδ, or ratio of viscous to elastic components, for cocoa butter (●) and Salatrim® (○).

butter or Salatrim®, for in the course of this work, different w/w percentages of confectionery fat/canola oil were imaged (not shown). The main effect of canola oil addition was to de-

crease the SFC of the network, thus improving image resolution.

The fat crystal network in Salatrim® is composed of small, randomly arranged, noncrystalline, translucent platelets, about 10 μm in diameter (Fig. 2C). These striking differences in microstructure can be readily explained from knowledge of triglyceride structure and polymorphic behavior. Salatrim® is a synthetic, random fat, containing short- and long-chain fatty acids. Due to the high asymmetry in the molecular structure of the triglycerides present in Salatrim®, packing into a crystalline structure is extremely difficult, and hence, no discrete crystalline polymorphic behavior was observed. Instead, there seems to be the formation of a solid phase characterized by weak interactions between mismatched triglycerides. Therefore, instead of the formation of microstructural elements consisting of crystalline solid, Salatrim®'s network seems to grow in one solid phase, with platelet growth discontinuing when the symmetry of the random triglycerides present is insufficient to form strong enough interactions to the platelet surface molecular symmetry. The random spatial distribution of the platelets points to very weak links between them, since if the links were sufficiently strong a definite fractal or Euclidean interplatelet distance would be observed. Whenever strong interparticulate forces are present in a three-dimensional network, the three-dimensionality of the system forces regular or pattern-forming interparticulate distances. Therefore, the random nature of the triglycerides in Salatrim® results in the formation of a weak, random network.

On the other hand, cocoa butter triglycerides do form defined crystal lattices upon crystallization (4). The network is formed as many initial centers of nucleation become larger crystalline entities, the larger crystalline entities then aggregate in a mass and heat transfer-limited aggregation process into the microstructural network described above. The lattice thus formed is a fractal lattice. Therefore, the highly ordered nature of the triglycerides in cocoa butter results in the formation of a network with defined structural elements and a characteristic spatial distribution of solid mass.

It is possible to quantify the spatial distribution of solid mass in the fat crystal network using scaling relationships between the storage modulus (G′) and the SFC (3,12). From this type of analysis, it is possible to derive a fractal dimension and certain network characteristics, as well as a parameter, γ, dependent on the size of the primary particles and on the interactions between them (14), if there indeed exists a scaling relationship between G′ and SFC. Figure 5 shows the results of a rheological determination of the fractal dimension of the fat crystal networks in cocoa butter and Salatrim®. It must be mentioned here, however, that fractal dimensions calculated rheologically in this manner are physically significant only if the physical structure of the network is fractal in nature, i.e., self-similar on different length scales. The microstructural network of cocoa butter is a striking example of a fractal, microstructural network. However, from the PLM image of Salatrim®, this network does not appear to be fractal. The rheological analysis for fractal dimension is only included in this communication as a warning of the pitfalls associated with this type of analysis. Although such analysis can be extremely useful, one must be careful of the fact that unless the network itself is fractal, any apparent scaling relationship must be viewed with caution. It is often difficult to discern whether a straight line from a Log-Log plot is straight due to physical implications of scaling behavior (or a power-law relationship) or due to the fact that almost any Log–Log plot appears faintly linear.

The structure of Salatrim® at the scales equivalent to the microstructural level of cocoa butter is a random structure, as opposed to a fractal structure. The rheologically-calculated fractal dimension of 2.90 is, at first glance, commensurate with a low elastic constant (10); however, we believe this to be misleading for reasons mentioned above. In contrast, the rheologically-calculated fractal dimension of the cocoa butter network is 2.37 (weak link regime), characteristic of structures formed by reaction-limited mechanisms (15). The rheological analysis is supported by image analysis of the PLM image of the microstructure of cocoa butter, where the fractal dimension was calculated to be 2.31. The rheological analysis was performed assuming a model (weak-link model) developed by Shih *et al.* (13) for colloidal gels and extended by Narine and Marangoni (10) to microstructural networks of fat crystals where the links between the microstructures are mechanically weaker than the microstructures themselves. In this model, any small deformation of the network places stress upon the links between the microstructures rather than upon the individual elements.

Therefore, there are fundamental differences between the structures of Salatrim® and cocoa butter at the scales which, for reasons discussed above, are most affected by small-deformation rheological measurements. From the description of the differences in structure, it seems that small-deformation

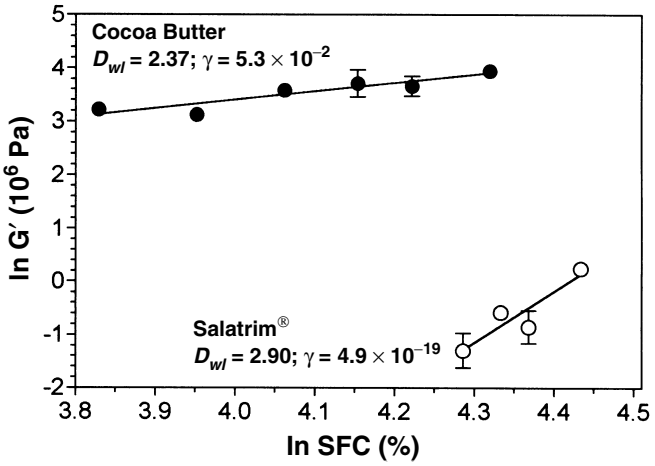

FIG. 5. Scaling relationship between the storage modulus (G′ @ 1 Hz) and the solid fat content for cocoa butter and Salatrim®/canola oil blends. *D* corresponds to the fractal dimension in the weak link (*wl*) regime. The constant γ is directly proportional to the Young's modulus of the particles that make up the network.

rheological measurements on Salatrim® stress the platelets that make up the network and the weak links between the platelets. As described above, these links must be very weak, since they represent the boundary to the growth of the platelets; we propose that this is due to the asymmetry of the triglycerides present and the subsequent inability of the mismatched triglycerides to form interactions strong enough to continue the growth of the platelet. On the other hand, cocoa butter forms a well-defined microstructural network, with the distance between microstructural elements statistically similar. This suggests that the interactions between the microstructural elements are quite strong and defined, leading to a three-dimensional arrangement with similar intermicrostructural distances. As mentioned before, the shear elastic modulus of Salatrim®'s network is one order of magnitude lower than that of cocoa butter at different SFC. The explanation that the entities (links between microstructures) being stressed during the measurement of the macroscopic shear elastic modulus in cocoa butter are stronger (higher elastic constant) than those being stressed in the Salatrim® network (links between platelets) is therefore a very probable cause of this difference, bearing in mind that the melting profiles and SFC of these two confectionery fats do not explain these differences. An additional consideration is the fact that the microstructural elements themselves in cocoa butter are hard, crystalline particles, whereas the platelets in Salatrim® are noncrystalline, relatively soft particles. In the event of the small-deformation measurements actually causing stress on the microstructural entities themselves, therefore, the cocoa butter network would be harder.

It is important to mention that it was previously established that the hardness of a fat, as determined by large-scale rheological analyses such as cone penetrometry, is directly correlated to the hardness determined by sensory analysis (16–19). Furthermore, measurements in our laboratory indicate that there is a direct relationship between the elastic modulus (storage shear modulus) of a fat and its hardness index, as determined by cone penetrometry measurements. Thus the elastic modulus of a fat crystal network is an indicator of the macroscopic consistency of that network. Therefore, from our measurements, cocoa butter is a much harder fat than Salatrim®. It would therefore seem that a highly organized, fractal crystal network structure with a high G'/G'' ratio is required to achieve proper hardness and immobilization of nontriglyceride components. Additionally, a fractal structure would be very efficient in trapping oil, or nontriglyceride components, since cocoa butter can store much more oil than Salatrim®, as described above. One can imagine exogenous components being trapped within the network and between microstructural elements upon crystallization, since there is much more space within the microstructural network. A space-filling random structure such as the one present in Salatrim® does not have the ability to trap much exogenous material.

Apart from offering an explanation for the differences between cocoa butter and Salatrim®, an examination of the microstructural characteristics of these two networks also offers interesting insights into preferred microstructural characteristics of confectionery fats. Cocoa butter is a much-utilized confectionery fat, and can be thought of as a good candidate for the representation of a perfect confectionery fat, while Salatrim® fails in many instances to justify its use as a confectionery fat. Therefore, the differences between these two fats outline the preferred characteristics of a good confectionery fat.

The very important "snap" characteristic of a good chocolate product is related to the mechanical failure of the network along a microstructural link fracture plane. With a network such as Salatrim®, the "snap" would not be achieved, since the random nature of the spatial arrangement of the platelets does not allow fracture planes to be defined along the links between the platelets, instead, the network would bend due to the bending of the platelets themselves. Therefore, the formation of a fractal microstructural network generally produces harder, brittle fats. Consequently, it is important that a confectionery fat adhere to the weak-link model described earlier.

Contraction is another important property of confectionery fats. Without it, demolding would not be possible. The contraction zones we observed in low-magnification PLM images of cocoa butter would not be possible if the network was not composed of aggregates of hard microstructural elements separated by finite distances. Without the packing of crystals into these highly organized microstructures, contraction probably would not occur. In fact, one of the most problematic characteristics of Salatrim® is the difficulty it presents in demolding.

An incomplete list of criteria for the ideal confectionery fat should therefore include: (i) a material with a high G' and a low G''; (ii) a fat crystal network with a fractal nature; (iii) a fat crystal network composed of aggregated microstructural particles; and (iv) weak-link network characteristics. Fractal rheological analysis of the fat crystal network in fats, combined with simple polarized light microscopy, could be used in the design of high-quality confectionery fats.

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REFERENCES

1. deMan, J.M., and A.M. Beers, Fat Crystal Networks: Structure and Rheological Properties, *J. Texture Studies 18*:303–318 (1987).
2. Heertje, I., Microstructural Studies in Fat Research, *Food Structure 12*:77–94 (1993).
3. Marangoni, A.G., and D. Rousseau, Is Plastic Fat Rheology Governed by the Fractal Nature of the Fat Crystal Network? Interesterification Decreases the Fractal Dimension of Butterfat–Canola Oil Blends, *J. Am. Oil Chem. Soc. 73*:991–994 (1996).

4. Wille, R.L., and E.S. Lutton, Polymorphism of Cocoa Butter, *Ibid. 43*:491–496 (1966).
5. Manning, D.M., and P.S. Dimick, Crystal Morphology of Cocoa Butter, *Food Microstructure 4*:249–265 (1985).
6. Hicklin, J.D., G.G. Jewel, and J.F. Heathcock, Combining Microscopy and Physical Techniques in the Study of Cocoa Butter Polymorphs and Vegetable Fat Blends, *Ibid. 4*:241–248 (1985).
7. Koyano, T., I. Hachiya, and K. Sato, Fat Polymorphism and Crystal Seeding Effects on Fat Bloom Stability of Dark Chocolate, *Food Structure 9*:231–240 (1990).
8. van Malssen, K., R. Peschar, and H. Schenk, Real-Time X-Ray Powder Diffraction Investigations on Cocoa Butter. I. Temperature-Dependent Crystallization Behavior, *J. Am. Oil Chem. Soc. 73*:1209–1215 (1996).
9. Narine, S.S., and A.G. Marangoni, Microscopic and Rheological Studies of Fat Crystal Networks, *J. Crystal Growth* (in press, 1999).
10. Narine, S.S., and A.G. Marangoni, Fractal Nature of Fat Crystal Networks, *Phys. Rev. E 59(2)* (1999).
11. Kellens, M., W. Meeussen, and H. Reynaers, Study of the Polymorphism and the Crystallization of Tripalmitin: A Microscopic Approach, *J. Am. Oil Chem. Soc. 69*:906–911 (1992).
12. Vreeker, R., L.L. Hoekstra, D.C. den Boer, and W.G.M. Agterof, The Fractal Nature of Fat Crystal Networks, *Colloids Surfaces 65*:185–189 (1992).
13. Shih, W.H., W.Y. Shih, S.I. Kim, J. Liu, and A. Aksay, Scaling Behavior of the Elastic Properties of Colloidal Gels, *Phys. Rev. A 42*:4772–4779 (1990).
14. Bremer, L.G.B., B.H. Bijsterbosch, R. Schrijvers, T. van Vliet, and P. Walstra, On the Fractal Nature of the Structure of Casein Gels, *Colloids Surfaces 51*:159–170 (1990).
15. Meakin, P., Fractal Aggregates, *Adv. Colloid Interface Sci. 28*:249–331 (1988).
16. Haighton, A.J., The Measurement of the Hardness of Margarine and Fats with Cone Penetrometers, *J. Am. Oil Chem. Soc. 36*:345–348 (1959).
17. Hayakawa, M., and J.M. de Man, Interpretation of Cone Penetrometer Consistency Measurements of Fats, *J. Texture Studies 13*:201–210 (1982).
18. Lefebvre, J., Finished Product Formulation, *J. Am. Oil Chem. Soc. 60*:247–252 (1983).
19. Rousseau, D., and A.G. Marangoni, The Effects of Interesterification on Physical and Sensory Attributes of Butterfat and Butterfat—Canola Oil Spreads, *Food Research International* (in press, 1999).

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