Mixed Monolayers of Acylglycerols on Sugar Aqueous Solutions

Julia de la Fuente Feria and Juan M. Rodríguez Patino
Dept. de Ingeniería Química, Facultad de Química, Universidad de Sevilla, 41012-Sevilla, Spain

Structural characteristics of mono- and distearin mixed monolayers spread on aqueous solutions containing sugar (glucose or sucrose) at 0.5 mol/L were studied as a function of temperature and interfacial and subphase compositions. The compression isotherms were obtained using an automated Langmuir-type film balance over a temperature range of 10 to 40°C. The results show an expansion in the structure of the mixed monolayer with sugar in subphase. Nonideal mixing of the component at interface was observed with positive deviation from the additivity rule. This fact is indicative of the presence of repulsive interactions between the two lipids on sugar aqueous solutions. The mixed monolayers with a monostearin molar fraction less than 0.8 are stable from a thermodynamic point of view. Mixed monolayer stability depends on monolayer composition, temperature, and surface pressure with different order of magnitude.

Introduction

In the past several years, extensive studies concerning the influence of emulsifiers (proteins and low molecular-weight emulsifiers or surfactants) on the stability of food dispersions have been reported (Dickinson and Stainsby, 1988; Dickinson and Walstra, 1993; Larsson and Friberg, 1990; Sjöblom, 1992; and Wan, 1990). These works showed that the stability of food emulsions and foams depends on the amount of emulsifier adsorbed and on the conformation adopted at the interface (Halling, 1981; Krog et al., 1985). The function of surfactants in foods is to facilitate large-scale, highly automated production of uniform products with improved shelf life that can withstand transport and storage (Leadbetter, 1990; Krog, 1990, 1992). Mono- and diglycerides are the most commonly used surfactants in the food industry (Als and Krog, 1991). The so-called monoglycerides produced by esterification of fats with glycerol are a blend of mono-, di- and triglycerides that contain from 40% to a maximum of 60% monoglyceride (Leadbetter, 1990; Krog, 1990, 1992). Concentrated monoglycerides are produced by a high-vacuum, thin-film molecular distillation process yielding products typically containing 95% monoglycerides. Distilled monoglycerides are widely used in many different types of foods (Krog, 1990, 1992; Krog et al., 1985). A major application is in the baking industry. Mono- and diglycerides are also used as the major emulsifier in the ice cream industry.

The study of mixed emulsifier films is important because it leads to an understanding of the preferential structuring at the interface and is of significant practical importance. The interactions and film characteristics (structure, elasticity, miscibility, etc.) of commercial or distilled monoglycerides at interface can be studied from mixed monolayers at the air–water interface. This information—which can be obtained from research on spread monolayers using Langmuir trough techniques (Krog et al., 1985)—may be used to improve the surface properties by means of an appropriate mixture of emulsifiers.

From a fundamental point of view, the actual structure of surfactant complexes at the oil–water or air–water interface is not well characterized and there is an incomplete understanding of all the factors that influence that structure. Furthermore, few relationships have been established that relate the structure of the mixed film to the environmental conditions (temperature, interfacial or subphase composition, etc.). A more complete understanding of these issues would facilitate the selection and use of emulsifiers that have appropriate interfacial properties.

This work is an extension of studies of mixed emulsifier monolayers spread on aqueous solutions (Fuente Feria and Rodríguez Patino, 1995a,b). Due to their practical interest in food products, the structural characteristics of monostearin

---

Correspondence concerning this article should be addressed to J. M. Rodríguez Patino.

and distearin mixed monolayers spread on aqueous solutions containing sugar (glucose or sucrose) have been studied as a function of temperature and interfacial and subphase compositions. It has been observed that all of these factors affect the structural characteristics, miscibility, and stability of mixed monolayers. The effect of sugar in subphase on monostearin monolayers has been previously studied (Rodríguez Patino et al., 1993).

Materials and Method

Chemicals

Monostearin (1-monooctadecanoyl-rac-glycerol) and distearin (dioctadecanoyl-rac-glycerol, mixed isomers, approximately 50% 1,3- and 50% 1,2-isomer) more than 99% pure were acquired from Sigma. The sugars and analytical-grade ethanol and hexane were acquired from Merck and used without further purification. The water used as subphase was purified by means of a Millipore filtration device (Mille Q). The absence of surface-active contaminants in the water and in the hexane–ethanol mixture used as the spreading solvent was verified.

Apparatus and procedure

Measurements of surface pressure, π, vs. average area per molecule, A, were performed on a fully automated Langmuir-type film balance (Lauda). The apparatus, techniques, and experimental conditions used to study $\pi-A$ isotherms for single and mixed films on aqueous sugar solutions at 0.5 mol/L have been described in detail elsewhere (Fuente Feria and Rodríguez Patino, 1995a,b; Rodríguez Patino et al., 1992a,b, 1993). For mixed films, the separate components were dissolved in hexane–ethanol solutions (9:1, v/v), then premixed volumetrically in the required ratio and spread at the air–water solution interface by means of a micrometric syringe at the lowest temperature (10°C). Aliquots of 250 μL (from 6.17×10^{16} to 6.83×10^{16} molecules, depending on the molar ratio of the mixture) were spread in each experiment. To allow for evaporation of the spreading solvent at 10°C, 15 min elapsed before each measurement were taken. To verify the reproducibility of the results we periodically tested the film balance, by spreading stearic acid on water and recording the $\pi-A$ isotherms at different temperatures. From these experiments we can conclude that the reproducibility of the results—in the absence of monolayer molecular loss—agree with the sensitivity of both the film pressure (±0.1 mN/m) and the molecular area (±0.005 nm^2/molecule) of the film balance.

Six mixtures with a molar ratio of monostearin, x_M, ranging between 0 and 1 (0, 0.2, 0.4, 0.6, 0.8, and 1) were studied. The experiments were carried out at temperatures ranging between 10 and 40°C. The temperature of the system formed by the spread film and the subphase was maintained constant within ±0.2°C by a Lauda K2R electronic thermostat. The concentration of sugar solutions is 0.5 mol/L. This concentration is usual in food formulations, especially for sucrose in cereal-based foods such as bread, cakes, and other bakery products (Leadbetter, 1990).

Precautions were taken to collect reliable $\pi-A$ isotherms using the continuous compression methods described previously (Rodríguez Patino et al., 1992b). This is especially important in the case of monolayers that are unstable in relation to molecular loss toward either the air or water adjacent phase. In previous works performed in this laboratory we have observed from relaxation experiments that monoglyceride monolayers spread on aqueous ethanol and sugar solutions are unstable (Fuente Feria and Rodríguez Patino, 1994a,b, 1995c). For this reason the choice of compression rate is very important (McArthur and Berg, 1979; Motomura et al., 1969). In the present work, the chosen compression rate was $6.2\cdot10^{-2}$ nm$^2\cdot$molecule$^{-1}\cdot$min^{-1}. This value was such that isotherms obtained from previous experiments with monoglyceride– water systems are practically repetitive (Fuente Feria and Rodríguez Patino, 1994a,b; Rodríguez Patino et al., 1992a). The value of the compression rate chosen ensures reproducibility in the present work on mixed films.

Results and Discussion

Monolayer structural characteristics

The isotherms obtained for each mixture spread on aqueous solutions of glucose and sucrose at 0.5 mol/L are shown as a function of monostearin content (x_M) in Figures 1 and 2. It can be seen that the dominant monolayer structure is liquid-condensed and liquid-expanded, and is a function of the surface pressure value and of the monostearin content, respectively, in the mixture. The existence of these structures in a mixed monolayer depends on the variables and is a function of the degree of interaction between molecules at the interface. In this work the assignment of each structure to a monolayer is based on the $\pi-A$ isotherm (Gaines, 1966). The existence of a plateau corresponding to the transition between both structures is more evident in pure-component monolayers. The phase change degenerates with monostearin–distearin films at $0.2 \le x_M < 0.8$, and at temperatures lower than 40°C. At these experimental conditions the plateau corresponding to the transition between liquid-condensed and liquid-expanded structure is not observed. The mixed films on aqueous sucrose solutions (Figure 2) are more expanded —the isotherms are displaced toward higher areas—than with glucose in the subphase (Figure 1).

Moreover, it can be seen (Figures 1 and 2) that there is a monolayer expansion when both the temperature and distearin content in the mixture are increased. Temperature has a similar effect on the elasticity modulus ($-d\pi/dA$), obtained directly from the slope of the isotherms (data not shown). That is, the elasticity modulus is higher with decreasing temperature. However, the content of monostearin in the mixture has the opposite effect. In fact, the monolayer is more compact—that is, the molecules are closer together—and the elasticity modulus increases as the content of monostearin in the mixture increases, especially at temperatures lower than 40°C.

The effect of the sugars on the structural characteristics of the mixed monolayers is similar to that observed previously in this laboratory with pure monostearin monolayers (Rodríguez Patino et al., 1993) and with fatty acids (Rodríguez Patino et al., 1992a). An expansion of the monolayer structure with sugars in the subphase is observed. This monolayer expansion has the opposite effect to the one observed with the same mixed monolayers spread on water (Fuente Feria and Rodríguez Patino, 1995a) or on aqueous ethanol solu-

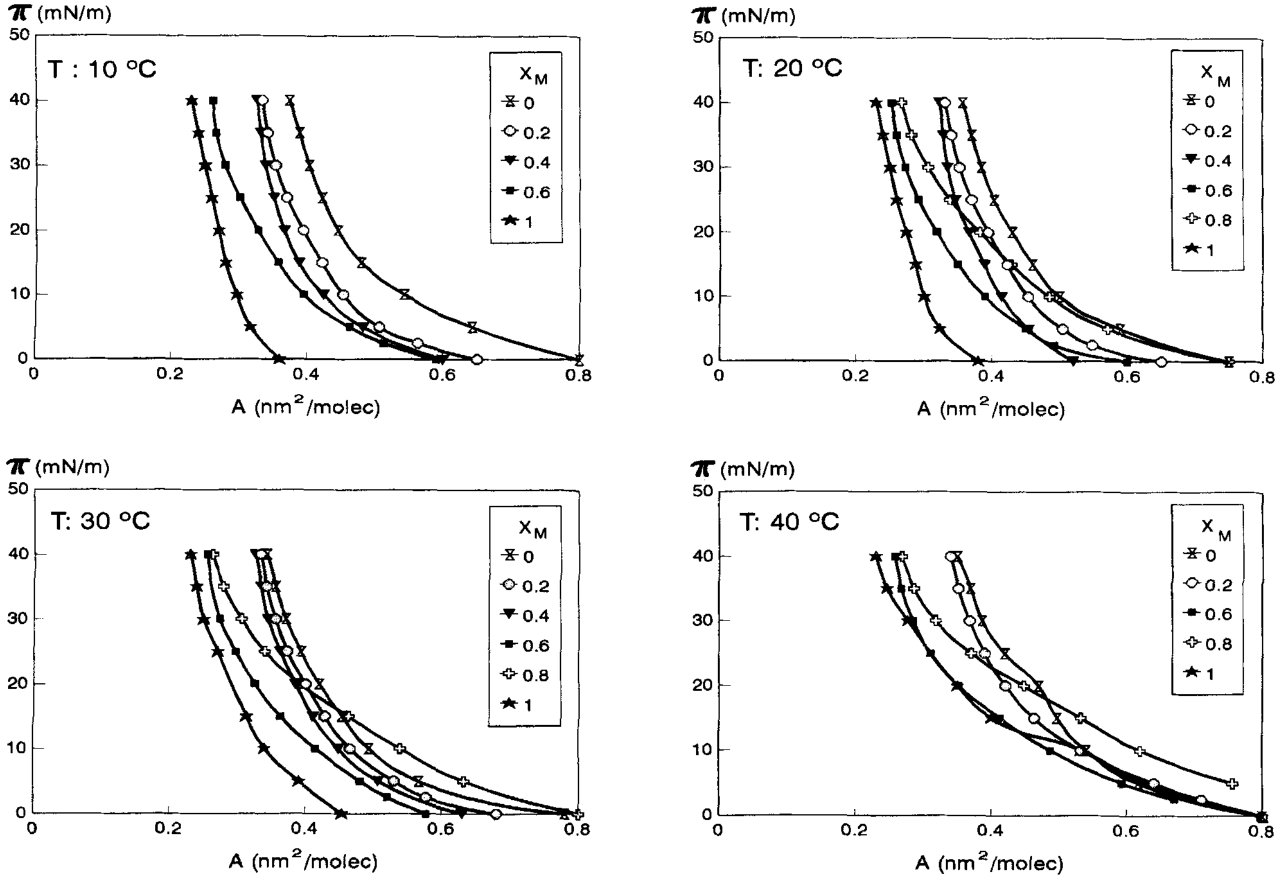

Figure 1. Surface pressure-area isotherms (compression curves) of monostearin–distearin mixed monolayers spread on 0.5 mol/L glucose at 10, 20, 30 and 40°C.

tions (Fuente Feria and Rodríguez Patino, 1995b), as can be seen in Figure 3.

As in McArthur and Berg (1979), differences in the physical properties (density, viscosity, and surface tension) of aqueous solutions of sugars and water, at the concentrations studied in this work, are too small to justify differences between structural characteristics of monostearin–distearin mixed monolayers spread on these aqueous solutions. Because the sugar molecules have no particular affinity to the air–water interface—surface tensions of aqueous glucose and sucrose solutions are higher than that of pure water at the same temperature (Rodríguez Patino and Martín Martínez, 1994)—the expansion of the monolayer can be attributed to the interactions between the monolayer and sugar molecules in the subphase.

The formation of hydrogen bonds between the polar head of the lipids and the hydrophillic groups of the sugar molecules may be the principal cause of these interactions near the surface (Rodríguez Patino et al., 1993; Taylor and Rowlinson, 1955). The existence of interactions between lipid and sugar molecules can decrease the van der Waals interactions between lipid hydrocarbon chains with similar consequences. Differences between structural characteristics of mixed monolayers spread on glucose and sucrose solutions suggest that the packing of water molecules around a glucose or sucrose molecule is different. This reasoning agrees with the thermodynamic properties of aqueous sugar solutions (Taylor and Rowlinson, 1955). The excess heat, entropies, free energies, and volumes indicate that there is stronger hydrogen bonding between glucose and water molecules than between sucrose and water molecules. But, as was discussed with monostearin spread on aqueous sugar solutions, the greater strength of the glucose–water association can be compensated for by the larger number of potential hydrogen-bonding groups in sucrose (Rodríguez Patino et al.,1993). Thus if sucrose–water interactions are weaker than those of glucose–water, the sucrose–lipid head polar interactions will be stronger, and in turn the interactions between the lipid hydrocarbon chains will be weaker. So the greater expansion of the mixed films spread on aqueous sucrose solutions could be due to these effects. We can see that the structural characteristics of mixed films depend on the two-dimensional interactions between the molecules of the monolayer as well as on the three-dimensional interactions between the monolayer molecules and solutes in the subphase. These two types of interactions are not independent.

Monolayer miscibility

The miscibility between the monolayer components is an important characteristic of mixed films related to the exist-

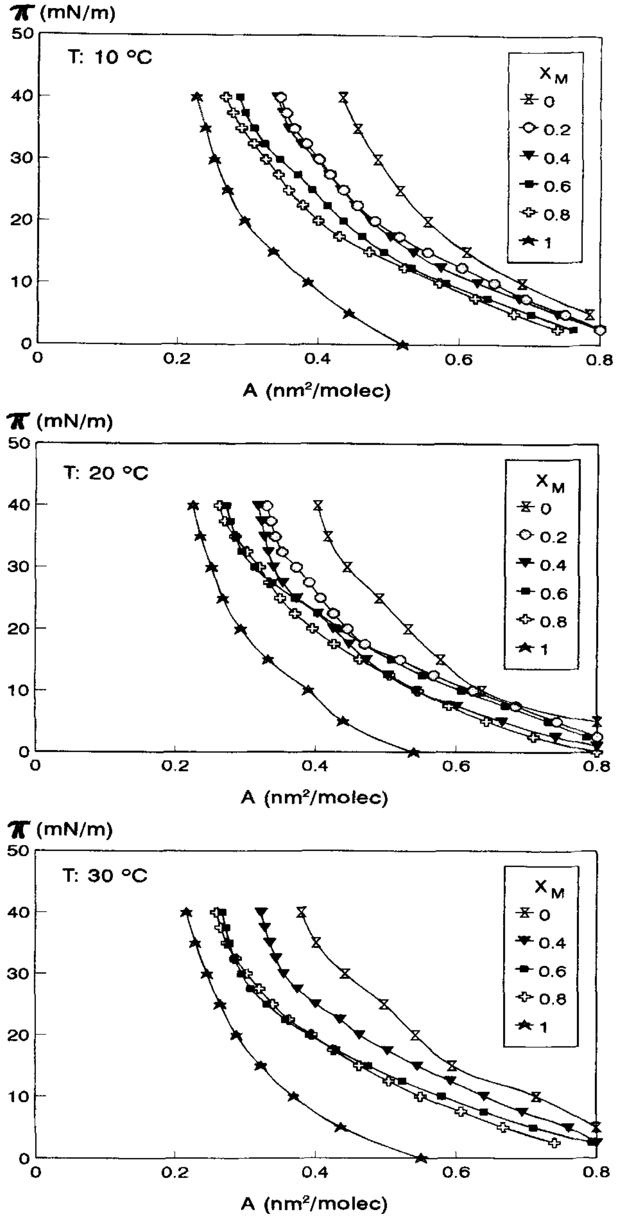

Figure 2. Surface pressure-area isotherms (compression curves) of monostearin, distearin, and their mixtures spread on 0.5 mol/L sucrose.

ence of intermolecular interactions at interface (Cadenhead and Müller-Landau, 1980; Costin and Barnes, 1975; Gabrielli and Baglioni, 1980; Tomoaia-Cotisel and Chifu, 1983) that can be deduced from the excess area, A_{exc}. The excess area is calculated following Eq. 1:

$$A_{\text{exc}} = A - (x_1 A_1 + x_2 A_2), \qquad (1)$$

where A, A_1 and A_2 are the molecular areas of the mixture, which can be evaluated directly from the $\pi - A$ isotherms, and the pure component monolayers, respectively, for a given surface pressure, and x_i is the molar fraction of each component in the mixture.

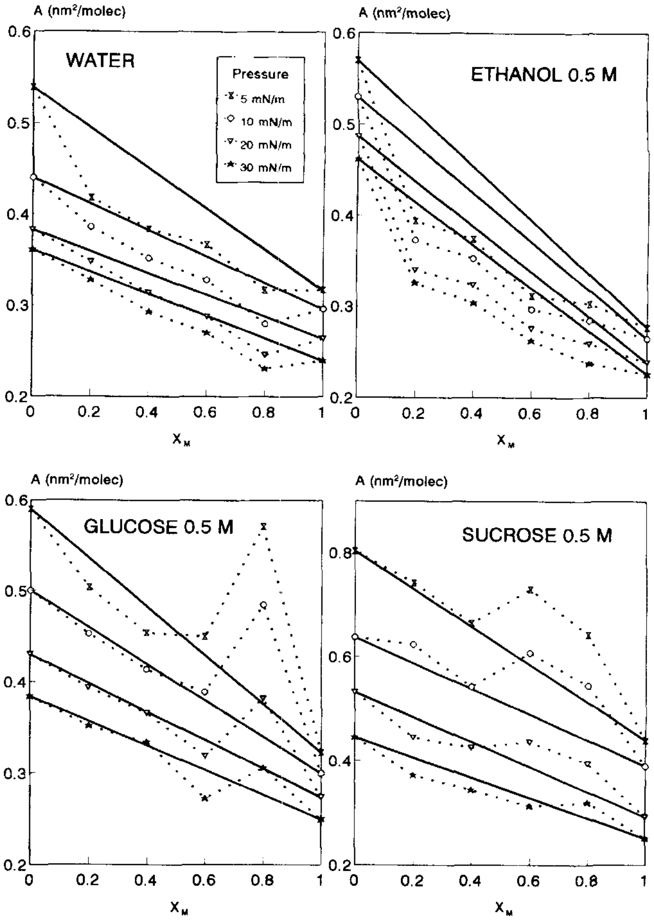

Figure 3. Molecular area vs. monostearin molar fraction for monostearin–distearin mixed monolayers spread on water, 0.5 mol/L ethanol, 0.5 mol/L glucose, and 0.5 mol/L sucrose as a function of the surface-pressure value at 20°C.

The excess-area–monolayer composition dependence for mixed films on aqueous solutions of glucose and sucrose is shown in Figures 4 and 5. The values of A_{exc} can be positive or negative depending on the monolayer composition, temperature, and surface pressure. An expansion of the mixed monolayer with respect to the ideal mixture is observed, especially in mixed monolayers with a molar fraction of monostearin $x_M = 0.8$. The positive values of A_{exc} are attributed to the miscibility and nonideality of the system with monolayer expansion (Kulkarni and Katti, 1982). The interactions between monolayer molecules, which would be affected by the presence of sugars in the subphase, suggest that the mixing process is affected by the surface interactions as well as by the packing at the interface. Positive deviations from the additivity rule, especially in mixed monolayers with high monostearin content, is indicative that repulsive interactions between the two lipids at interface are dominant (Cadenhead and Müller-Landau, 1980; Mestres et al., 1992; Tomoaia-Cotisel and Chifu, 1983) and depend on the surface pressure and to a lesser extent on the temperature.

It can be seen in Figures 4 and 5 that a variation in the surface pressure, at constant temperature and monostearin molar fraction, causes variations in the A_{exc} values. At posi-

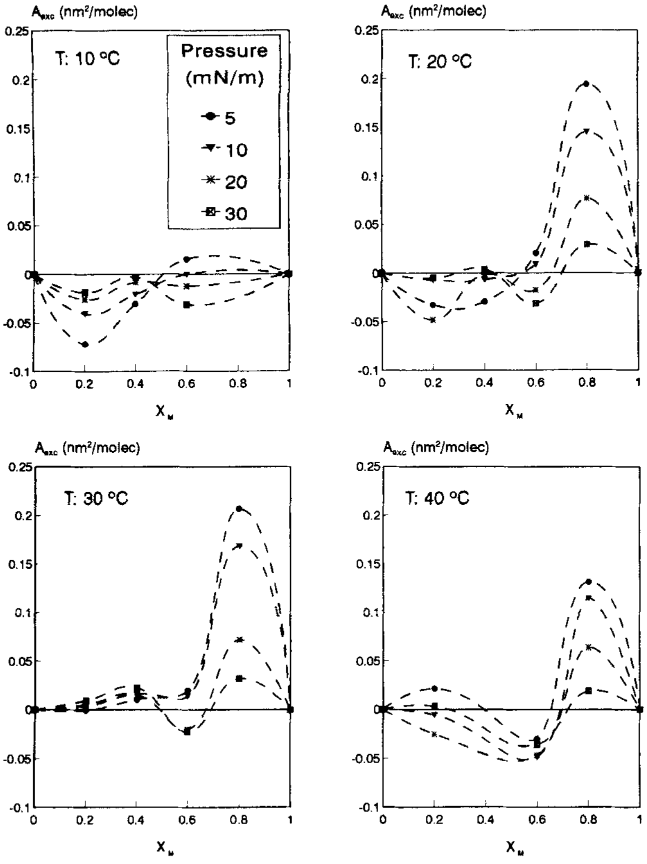

Figure 4. Excess area vs. monostearin molar fraction for monostearin–distearin mixed monolayers spread on 0.5 mol/L glucose as a function of surface pressure.

tive A_{exc} values, an increase in π is followed by a decrease in the A_{exc} value. That is, at the higher surface pressures, both the interaction in the mixed film and the packing of molecules at interface increase.

Thermodynamic parameters

The interactions between monolayer components in a mixed film and the stability of a mixed film in relation to pure components can be studied from a thermodynamic point of view. The magnitude of these interactions can be quantified from the excess free energy values or from the values of either the interaction parameter, $\alpha(\pi)$, or the energy corresponding to these interactions, Δh.

The values of excess free energy have been calculated using the Goodrich method (Goodrich, 1956), following the Pagano and Gershfeld criterion (Pagano and Gershfeld, 1972), and Eq. 2, from data shown in Figures 4 and 5. The integral is resolved analytically through a fourth-order polynomial equation that relates the values of A_{exc} to π:

$$\Delta G^{\text{exc}} = \int_{\pi \to 0}^{\pi} A_{\text{exc}} \cdot d\pi . \tag{2}$$

The interaction parameter (α) at different pressures and temperatures and the energy corresponding to these interactions (Δh) were evaluated using Eqs. 3 and 4 (Joos and Demel, 1969; Mestres et al., 1992).

$$\alpha = \frac{\Delta G^{\text{exc}}}{RT(x_1 x_2^2 + x_2 x_1^2)} \tag{3}$$

$$\Delta h = \frac{RT\alpha}{Z} . \tag{4}$$

To calculate interaction energies, the coordination number (Z) has been considered constant and equal to 6 according to the equivalences given by Quickenden and Tan (1974). The variations of α and Δh with composition, temperature, and pressure are similar. That is, the higher the interaction parameter, the higher the interaction energy. From results shown in Figure 6 it can be deduced that at 10°C a minimum in Δh exists at $x_M = 0.2$, the absolute value of which in-

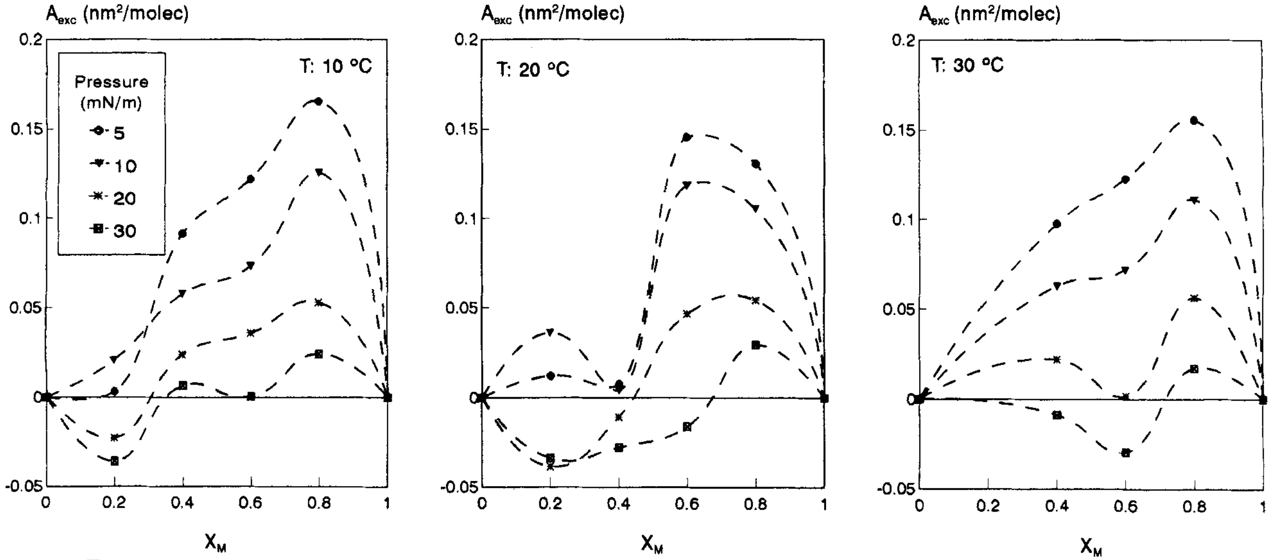

Figure 5. Excess area vs. monostearin molar fraction for monostearin–distearin mixed monolayers spread on 0.5 mol/L sucrose as a function of surface pressure.

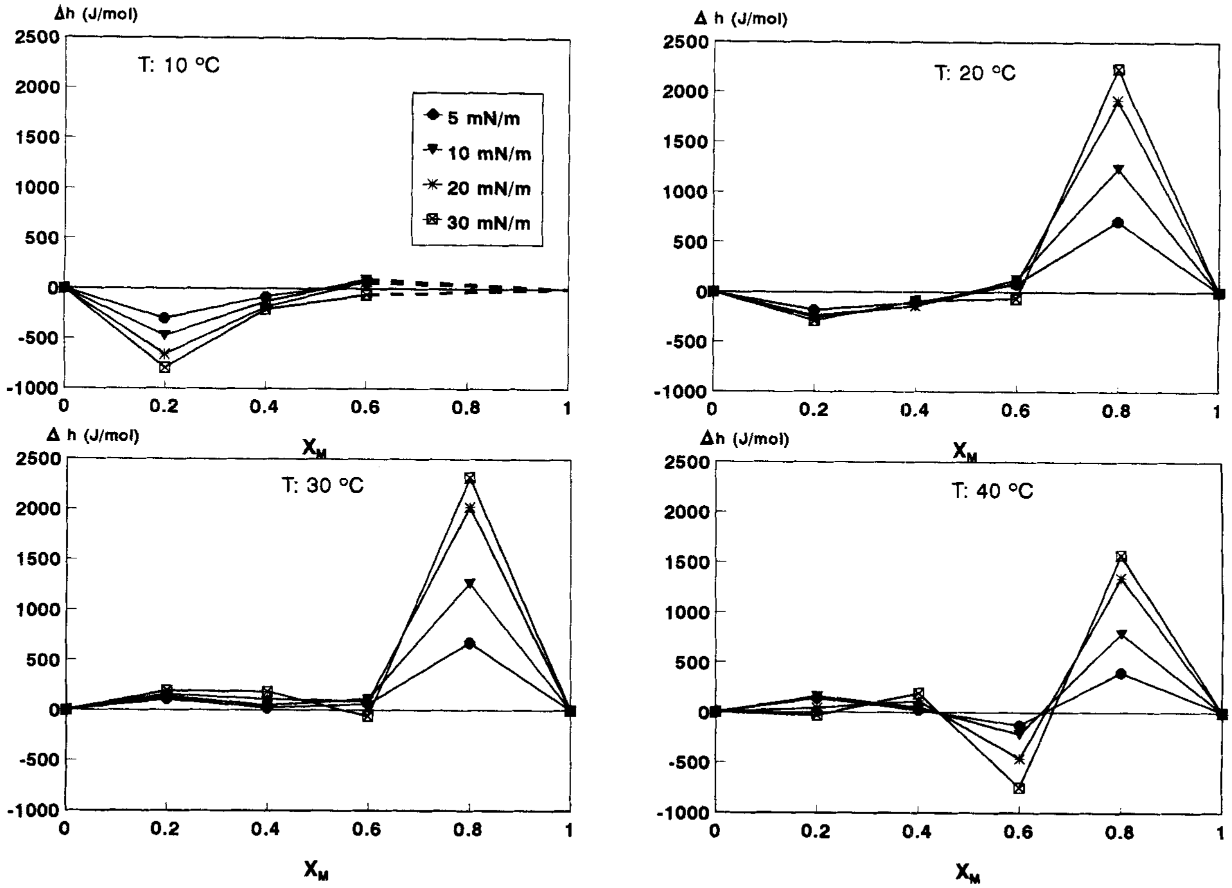

Figure 6. Interaction energy between monostearin and distearin molecules spread on 0.5 mol/L glucose vs. monolayer composition at different surface pressures.

creases with pressure. From results shown in Figures 6 and 7 it can be deduced that there is a maximum in Δh values at $x_M = 0.8$, the value of which increases with pressure. The Δh value in the maximum is practically independent of the temperature no matter what the subphase composition is (Figures 6 and 7). All these data agree with conclusions derived from deviations from the additivity rule (Figures 4 and 5).

The thermodynamic stability of mixed monolayers compared to that of the pure components is determined by the values of the total free energy of the mixing process. The analysis is based on the assumption that contributions from low-pressure data is not significant to the outcome of the analysis (Costin and Barnes, 1975). The practical lower limit of the integration is zero when monolayer components may be considered to mix ideally (Pagano and Gershfeld, 1972). According to the theory developed by Goodrich (1956), the evaluation of the total free energy of mixing, ΔG_M^π, can be made, quite simply, directly from the $\pi - A$ isotherms of pure and mixed monolayers (Eq. 5). The second term of the equation corresponds to the free energy for an ideal mixture. Equation 5 can be applied for any pressure lower than the equilibrium surface pressure (ESP) of the pure components (Kulkarni and Katti, 1982; Gaines, 1966, 1982). The ESP data of monostearin and distearin on aqueous solutions of glucose and sucrose of 0.5 mol/L, at temperatures ranging between 10 and 40°C are higher than 30 mN/m (Rodríguez Patino and Martín, 1994):

$$\Delta G_M^\pi = \int_{\pi \to 0}^{\pi} A_{\text{exc}} \cdot d\pi + RT(x_1 \ln x_1 + x_2 \ln x_2). \qquad (5)$$

From data shown in Figures 8 and 9, it can be deduced that a mixed monolayer composition of $x_M = 0.8$ decreases the stability of the monolayers formed by monostearin and distearin molecules spread on aqueous sugar solutions. At lower monostearin contents, the mixed monolayers are thermodynamically stable. The composition with maximum stability, which is coincident with the minimum in the $\Delta G_M^\pi - x_M$ plot, is practically independent of the surface pressure, but depends on the temperature. The composition of the maximum stability of the mixed monolayers is displaced toward higher monostearin content when the temperature increases.

The thermodynamic stability of mixed monolayers as compared to the ideal mixture can be explained if different zones are limited as a function of the monostearin content in the mixture. A model of association previously elaborated (Fuente Feria and Rodríguez Patino, 1995a,b) could be useful in this work because it takes into account both the monolayer expansion by sugar in the subphase and interactions between sugar and lipid molecules at the interface. As the sugar molecules have no particular affinity to the air–water interface, the lipid–sugar interactions are only possible in the zone just below the surface. So these interactions are greater when the monostearin molecules submerge in the subphase; this lipid is more hydrophillic than distearin. This phenomenon is

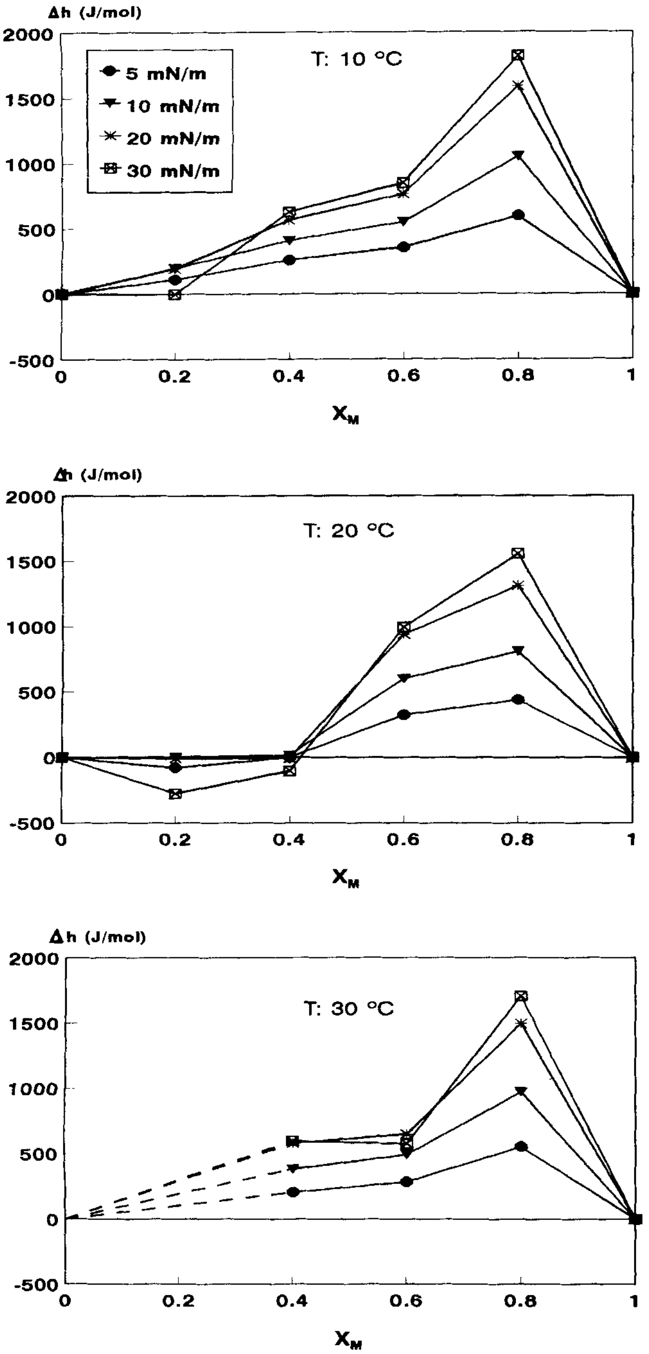

Figure 7. Interaction energy between monostearin and distearin molecules in mixed monolayers spread on 0.5 mol/L sucrose vs. monolayer composition at different surface pressures.

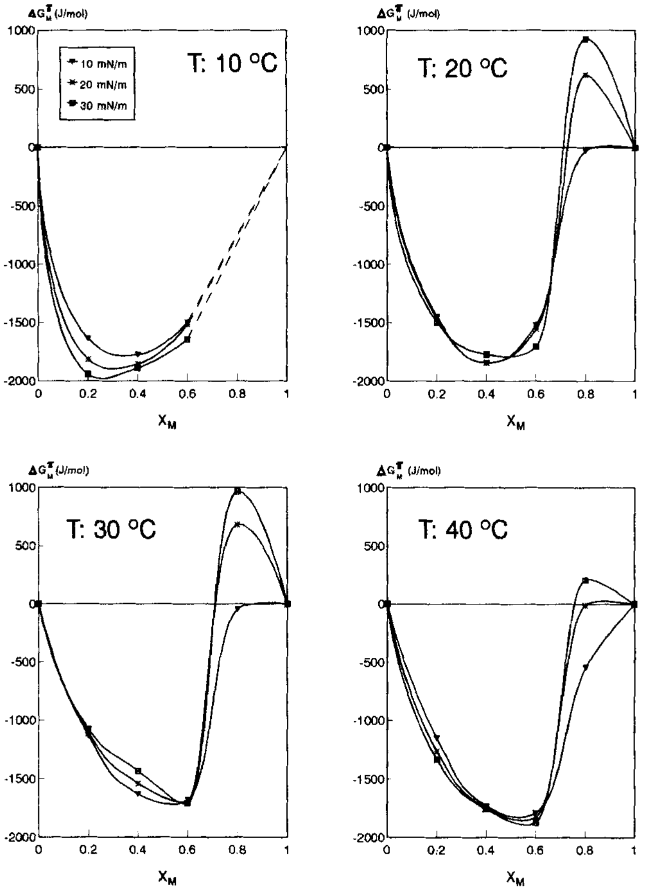

Figure 8. Total free energy of mixing of monostearin-distearin mixed monolayers on 0.5 mol/L glucose as a function of monolayer composition and surface pressure.

produced either in the monostearin-rich or in the distearin-rich zone. In the monostearin-rich zone, competition between greater head-group repulsion and greater hydrocarbon chain interactions exists because the molecules are close together.

Sugars in the subphase produce either positive deviations of ideality at $x_M = 0.8$ (Figures 4 and 5) or an expansion in $\pi - A$ isotherms toward larger areas (Figures 1 and 2). These phenomena agree with a decrease in the interactions between the hydrocarbon chains and/or an increase in the interactions between the lipid polar group and sugar. The fact that higher monostearin content in the mixture leads to greater thermodynamic instability in mixed films (Figures 8 and 9) could be explained by either effect.

Moreover, the temperature has no influence on the stability or instability of mixed monostearin-distearin monolayers on aqueous solutions of glucose or sucrose.

Conclusions

Structural characteristics, miscibility, and stability of mixed monostearin-distearin monolayers spread on aqueous solutions of glucose and sucrose at 0.5 mol/L have been studied as a function of the temperature and the interfacial and subphase compositions. The results show that sugars in the subphase cause an expansion in the structure of the mixed films. Nonideal mixing of the components at the interface was observed with positive deviations from the additivity rule, especially at higher proportions of monostearin or at lower pressures. This behavior has been attributed to miscibility and to the lipid-sugar intermolecular repulsive interactions at interface. The mixed monolayers on aqueous sugar solutions are stable at compositions lower than $x_M = 0.8$. The location of

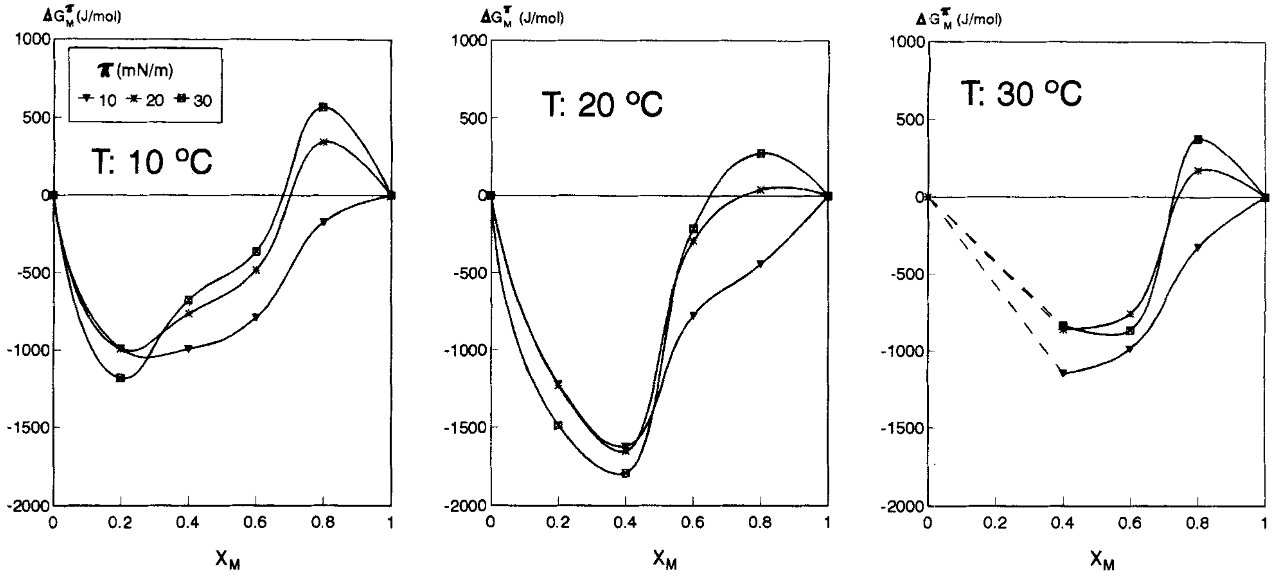

Figure 9. Total free energy of mixing of monostearin–distearin mixed monolayers on 0.5 mol/L sucrose as a function of the surface pressure.

the maximum stability depends on the temperature. However, this variable has no influence on the degree of monolayer stability. The influence of pressure on the stability of mixed films is significant. The stability of a given mixed monostearin–distearin monolayer increases with pressure.

Acknowledgment

The authors wish to acknowledge financial assistance from Plan Nacional I+D CICYT (IN93-0353) and DGICYT (PB93-0923).

Notation

ΔG^{id} = ideal free energy, J/mol
R = constant of ideal gas, J/K·mol
T = temperature

Literature Cited

Als, G., and N. J. Krog, "Emulsifiers as Food Processing Aids," *Proc. World Conf. on Oleochemicals into the 21st Century*, T. H. Applewhite, ed., Amer. Oil Chemists' Soc., Champaign, IL, p. 67 (1991).

Cadenhead, D. A., and F. Müller-Landau, "Molecular Accommodation and Molecular Interactions in Mixed Insoluble Monomolecular-Films," *J. Colloid Interf. Sci.*, **78**, 269 (1980).

Costin, I. S., and G. T. Barnes, "Two-Component Monolayers: II," *J. Colloid Interf. Sci.*, **51**, 106 (1975).

Dickinson, E., and G. Stainsby, eds., *Advances in Food Emulsions and Foams*, Elsevier, London (1988).

Dickinson, E., and P. Walstra, eds., *Food Colloids and Polymer: Stability and Mechanical Properties*, Royal Society of Chemistry, Cambridge, England (1993).

Fuente Feria, J. de la, and J. M. Rodríguez Patino, "Destabilization of Monoglyceride Monolayers at the Air-Aqueous Subphase Interface: 1. Kinetics," *Langmuir*, **10**, 2317 (1994a).

Fuente Feria, J. de la, and J. M. Rodríguez Patino, "Binary Mixture of Monostearin-Distearin Monolayers at the Air-Water Interface," *AIChE J.*, **41**, 1955 (1995a).

Fuente Feria, J. de la, and J. M. Rodríguez Patino, "Interactions of Ethanol in Subphase with Monostearin-Distearin Mixed Monolayers," *Langmuir*, **11**, 2163 (1995b).

Fuente Feria, J. de la, and J. M. Rodríguez Patino, "Relaxation Behavior of Emulsifiers at the Air-Water Interface as a Function of Film Elasticity," *Progress and Trends in Rheology IV*, C. Gallegos, ed., Steinkopff Verlag, Darmstadt, Germany, p. 185 (1994b).

Fuente Feria, J. de la, and J. M. Rodríguez Patino, "Destabilization of Monoglyceride Monolayers at the Air-Aqueous Subphase Interface: 2. The Role of the Film Elasticity," *Langmuir*, **11**, 2090 (1995c).

Gabrielli, G., and P. Baglioni, "Mixed Monolayers of Polydimethilsiloxane with Cellulose Acetate," *J. Colloid Interf. Sci.*, **73**, 582 (1980).

Gaines, G. L., *Insoluble Monolayers at Liquid-Gas Interface*, Interscience, New York (1966).

Gaines, G. L., "The Thermodynamic Equation of State for Insoluble Monolayers: III. Mixed Monolayers," *J. Colloid Interface Sci.*, **85**, 16 (1982).

Goodrich, F. C., "Molecular Interactions in Mixed Monolayers," *Proc. Int. Cong. Surface Activity*, Vol. 1, Butterworths, London, p. 85 (1956).

Halling, P. J., "Protein-Stabilized Foams and Emulsions," *CRC Crit. Rev. Food Sci. Nutr.*, **15**, 1551 (1981).

Joos, P., and R. A. Demel, "The Interactions Energies of Cholesterol and Lecithin in Spread Mixed Monolayers at the Air-Water Interface," *Biochim. Biophys. Acta*, **183**, 447 (1969).

Krog, N. J., "Food Emulsifiers and Their Chemical and Physical Properties," *Food Emulsions*, 2nd ed., K. Larsson and S. E. Friberg, eds., Marcel Dekker, New York, p. 127 (1990).

Krog, N. J., "The Role of Low-Polar Emulsifiers in Protein-Stabilized Food Emulsions," *Emulsions—A Fundamental and Practical Approach*, J. Sjöblom, ed., Kluwer, Dordrecht, The Netherlands, p. 61 (1992).

Krog, N. J., T. H. Riison, and K. Larsson, "Applications in the Food Industry: I," *Encyclopedia of Emulsion Technology*, P. Becher, ed., Vol. 2, Marcel Dekker, New York, p. 321 (1985).

Kulkarni, W. S., and S. S. Katti, "Surface Viscosity and Pressure-Area Isotherms of Mixed Monolayers of Hexadecanol with Docosanol and Hexadecoxy Ethanol with Docosanoxy Ethanol at 25.0°C," *J. Colloid Interf. Sci.*, **89**, 40 (1982).

Larsson, K., and S. E. Friberg, eds., *Food Emulsions*, 2nd ed., Marcel Dekker, New York (1990).

Leadbetter, S. L., "Emulsifying Agents in the Food Industry: a Literature Survey," *Food Focus*, No. 9, Leatherhead Food R.A., Brit. Food Mfg. Ind. Res. Assoc. (1990).

McArthur, B. W., and J. C. Berg, "Surface Equation of State for Insoluble Monolayers on Aqueous Solutions," *J. Colloid Interf. Sci.*, **68**, 201 (1979).

Mestres, C., M. A. Alsina, M. Espina, L. Rodríguez, and F. Reig, "Miscibility of Dipalmitoylphosphatidylcholine and Sulfatides in Mono- and Bilayers," *Langmuir*, **8**, 1388 (1992).

Motomura, K., A. Shibata, M. Nahamura, and R. Matuura, "Fatty Acid Monolayers in the Expanded State: II. The Effect of Compression Velocities," *J. Colloid Interf. Sci.*, **29**, 623 (1969).

Pagano, R. E., and N. L. Gershfeld, "Physical Chemistry of Lipid Films at the Air-Water Interface: II," *J. Phys. Chem.*, **76**, 1238 (1972).

Quickenden, T. I., and G. K. Tan, "Random Packing in Two-Dimensions and the Structure of Monolayers," *J. Colloid Interf. Sci.*, **48**, 382 (1974).

Rodríguez Patino, J. M., J. de la Fuente Feria, and C. Gómez Herrera, "Fatty Acid Films Spread on Aqueous Solutions of Compounds Containing Alcohol Radicals: Structure and Stability," *J. Colloid Interf. Sci.*, **148**, 223 (1992a).

Rodríguez Patino, J. M., M. Ruíz Domínguez, and J. de la Fuente Feria, "Monostearin Monolayers Spread on Aqueous Solutions Containing Ethanol," *J. Colloid Interf. Sci.*, **154**, 146 (1992b).

Rodríguez Patino, J. M., M. Ruíz Domínguez, and J. de la Fuente Feria, "The Effect of Sugars on Monostearin Monolayers," *J. Colloid Interf. Sci.*, **157**, 343 (1993).

Rodríguez Patino, J. M., and R. Martín Martínez, "Spreading of Acylglycerols on Aqueous Surfaces at Equilibrium," *J. Colloid Interf. Sci.*, **167**, 150 (1994).

Sjöblom, J., ed., *Emulsions–A Fundamental and Practical Approach*, Klumer, Dordrecht, The Netherlands (1992).

Taylor, J. B., and J. S. Rowlinson, "The Thermodynamic Properties of Aqueous Solutions of Glucose," *Trans. Farad. Soc.*, **51**, 1183 (1955).

Tomoaia-Cotisel, M., and E. Chifu, "Xantophyl Films: II. Two-Component Monolayers of Some Xantophylis and Egg Lecithin at the Air-Water Interface," *J. Colloid Interface Sci.*, **95**, 355 (1983).

Wan, P. J., ed., *Food Emulsion and Foams: Theory and Practice*, AIChE Symp. Ser. (1990).

Manuscript received Nov. 28, 1994, and revision received Aug. 10, 1995.