Wetting of Fat Crystals by Triglyceride Oil and Water. 2. Adhesion to the Oil/Water Interface

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ABSTRACT: Fat crystals influence the stability of food emulsions, such as margarine, butter, or cream, if adsorbed to the oil/water interface. During the adsorption process, a new fat crystal/water interface is created, while the oil/water interface is lost. The driving force for adsorption is therefore the difference between the interactions between fat crystal/water and oil/water. In this work, we have estimated this interaction difference and compared it to the displacement energy for fat crystals from the oil/water interface to the oil. Our calculations have shown that fat crystal adsorption to the oil/water interface (expressed by contact angle θ) is determined by polar energy, excess of fat crystal/water over oil/water ($I_{sw} - I_{ow}$). The interfacial tension constitutes the resistance force for crystal adsorption to the interface. Polar interaction energy for fat crystal/water is stronger than the polar interaction energy for oil/water in all cases examined ($I_{sw} - I_{ow} > 0$). The difference corresponds to about 10^4–10^6 hydrogen bonds for a hypothetical fat crystal with a diameter of 1 μm. The displacement energy for fat crystals to oil is lower than the polar energy excess in most cases examined. Thus, an additional interaction between fat crystals and oil makes it easy to displace the crystals to the oil. There is also a relationship between the adhesion tension ($-\gamma_{ow} \bullet \cos\theta$) for the crystals at the oil/water interface and the interfacial tension γ_{ow} A straight line of slope −1 is achieved for systems with low interfacial tensions (γ_{ow}) and low polar energy excess ($I_{sw} - I_{ow}$).
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KEY WORDS: Adhesion energy, adhesion tension, contact angle, emulsifiers, fat crystals, Fowkes equation, interfacial tension, oil/water interface, polar interaction, Young equation.

Partially crystalline fat constitutes the continuous phase of food emulsions such as margarine, butter, or spreads. Solid fat crystals influence the stability of these emulsions if they are attached to the emulsion droplets. Generally speaking, colloidal particles contribute to the stability of emulsions if they are preferentially wetted by the continuous phase, and contribute to their instability if they are preferentially wetted by the droplet phase. In our previous study (1), wetting of fat crystals was determined by contact angle measurements in several systems of differing composition. The results show that fat crystals are rather nonpolar (contact angles through the oil ≈30°). Emulsifiers in oil and surface-active proteins in water make the crystals more polar.

The adhesion strength for fat crystals at the oil/water interface (represented by displacement energy) was calculated in our previous paper (1) by using an approach presented by Becher (2), Friberg (3), and Friberg *et al.* (4). In similar systems, Young (5) and modified Fowkes (6) equations were applied to relate the adhesion tension ($-\gamma_{ow} \bullet \cos\theta$) to the interfacial tension (γ_{ow}) (7,8). A straight line of a slope −1 was found and explained by unchanged wetting of fat crystals on addition of emulsifiers to the oil (constant contact angles, θ). In support of this thesis, the authors presented constant contact angles on addition of low concentrations of some monoglycerides and lecithins to systems that contained hardened palm oil, soybean oil, and water (8).

The aim of this work is to estimate the polar excess for fat crystals/water over oil/water, and compare it with the displacement energy for crystals from the oil/water interface to oil for a system that contains β′ fat crystals, vegetable oil, emulsifiers, and water.

EXPERIMENTAL PROCEDURES

Fat crystals. Fully hydrogenated palm oil (palm stearin) from Karlshamns AB (Karlshamn, Sweden) was the fat crystal used in our study. The palm stearin had a mole mass of 756 g/mol on average and an iodine value of 2.6. Its purity and density were not specified. It was stable in the polymorphic form β′, with a melting point of 57–59°C. The α-form appears in a rapidly cooled palm stearin and has a melting point of about 46°C. Fatty acid and triglyceride compositions of palm stearin were reported before (1).

Oils. A refined soybean oil from Karlshamns AB was used as the oil phase. The interfacial tension between this oil and distilled water was slightly above 30 mN/m. This high value indicated high purity with regard to surface-active components (monoglycerides <0.05%, diglycerides <0.7%, free fatty acids <0.03%, lecithins <1 ppm). The oil had an iodine value of about 130 (~2.5 double bonds per hydrocarbon chain on average) and a melting point of about −20°C. Fatty acid

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and triglyceride compositions of soybean oil were presented in our previous article (1).

Emulsifiers. The following nonionic emulsifiers were studied as additives to the oil: monounsaturated monoglyceride monoolein (purity >99%), saturated distilled monoglycerides (Dimodan PV), lactic acid esters of distilled monoglycerides (Lactodan P22), polyoxyethylene (20) sorbitan monostearate (ethoxylated sorbitan monostearate, Tween 60), pentaethyleneglycol mono-*n*-dodecylether (ethoxylated alkyl ether, Nikkol BL5SY), polyoxyethylene (12) castor oil (Breddol 691), and polyoxyethylene (20) castor oil (Breddol 697). The first three emulsifiers (monoglycerides and their esters) were supplied by Grinsted A/S (Braband, Denmark), polyoxyethylene sorbitan monostearate by ICI Surfactants (Everberg, Belgium), ethoxylated alkyl ether by Nikko Chemicals (Tokyo, Japan), and polyoxyethylene castor oils by Berol Nobel (Stenungsund, Sweden). We also examined some lecithins (zwitterionic emulsifiers): pure phosphatidylcholine from soybean oil (soybean PC, Epikuron 200) and three technical lecithins (Topcithin, sterncithin, and Metarin P). Soybean PC, Topcithin, and Metarin P were supplied by Lucas Meyer (Hamburg, Germany) and sterncithin by Stern Chemie (Hamburg, Germany).

The first four nonionic emulsifiers and all lecithins were of food grade, ethoxylated castor oils were for food products, and ethoxylated alkyl ether was a special chemical for research purposes. The emulsifiers were specified (purities, mole masses, hydrophilic–lipophilic balance (HLB) values, polar head groups, and hydrocarbon chains) in more detail previously (1).

Other chemicals. Doubly distilled water was used as the water phase in all experiments.

Methods. Polar energy excess for fat crystals/water and the displacement energy were calculated from interfacial tension and contact angles as determined by the methods presented below. Further experimental details were published before (1).

Interfacial tension: pendant drop method. Interfacial tension between oil and water was measured by a pendant drop method (9), where the tension is obtained from the shape of a droplet. The oil droplet was pressed upward from a bent needle into water. The droplet shape was recorded by a CCD video camera and analyzed on a Personal Computer with an image processing program Micro GOP 300 (CONTEXTVISION AB, Linköping, Sweden) by an algorithm of Finn Knut Hanssen (University of Oslo, Norway). Reproducibility by this method is ≤5%. It is especially suitable for recording the time dependence of interfacial tension for systems with slow equilibrium adsorption, where other methods (such as Wilhelmy plate or Du Noüys ring) fail.

Contact angles. Contact angles at the three-phase contact (water–fat crystal–oil) were measured through the oil phase with equipment described previously (1). A measuring plug was filled with fat and dipped in a water-filled glass vessel at room temperature. The oil droplet was pressed through the plug onto the fat crystal surface. The contact angles were taken with a goniometer, a conventional apparatus from Ramé-Hart Inc. (Mountain Lake, NJ).

Advancing contact angles were determined first by pressing an oil droplet (~100 µL) onto the water-covered fat crystal. The receding contact angles were determined afterward by sucking back ~50 µL of the oil phase. The advancing contact angles represent fat crystals initially covered by water and approaching the oil/water interface from the water side. The receding contact angles represent fat crystals that approach the interface from the oil side. The receding contact angles are particularly sensitive to the nonpolar areas on the surface (10) and are less sensitive to surface impurities, for example, water molecules.

Preparation of fat crystal surfaces for contact angle measurement. A base fat (tristearin of technical grade) was melted at 80°C. A niche in the measuring plug was filled with the melted base fat and allowed to crystallize in the air at room temperature. Palm stearin was melted at about 80°C, and the plug was dipped into it. The plug was refrigerated to obtain the α form of the top fat by quick supercooling. The presence of the α form was confirmed by differential temperature analyzer (DTA) and X-ray diffraction analysis, performed at Karlshamns AB. To get a palm stearin β′ surface, the samples were tempered for 90 min. The β′ form was confirmed by DTA and X-ray diffraction analysis.

RESULTS AND DISCUSSION

Interfacial tensions and contact angles. A summary of results is presented in Table 1. The interfacial tension between soybean oil and water is high (>30 mN/m), indicating high purity with respect to surface-active components. All emulsifiers examined adsorbed onto the soybean oil/water interface and lowered the interfacial tension. The degree of lowering depended on emulsifier type and concentration.

Fat crystals in pure oil are nonpolar (contact angles $\theta < 90°$), and adsorbed to the oil/water interface ($\theta > 0°$). Advancing contact angles were higher than receding angles, indicating that the crystals approaching the oil/water interface from the water side are more polar than the crystals approaching the interface from the oil side. All emulsifiers examined adsorbed to the fat crystals and increased the contact angles (more polar crystals). The degree of increase depended greatly on the emulsifier and its concentration. More detailed results have been presented elsewhere (1).

Polar energy excess for fat crystals/water over oil/water: modified Young and Fowkes equations. Wetting of solids by oil and water is described by the Young equation (5), which represents a balance of interfacial tensions:

$$\gamma_{so} - \gamma_{sw} = -\gamma_{ow} \cdot \cos\theta \qquad [1]$$

where θ is the contact angle measured through the oil phase; γ is the interfacial tension between solid/oil (*so*), solid/water (*sw*), and oil/water (*ow*), respectively (Fig. 1).

The interfacial tension is the sum of contributions from

TABLE 1

Interfacial Tension Between Soybean Oil and Water, and Contact Angle at Three-Phase Contact: Fat Crystal–Oil–Water for Systems That Contained Double-Distilled Water, Refined Soybean Oil, and Palm Stearin β′ Crystal

Emulsifier	Concentration (%)	Interfacial tension (mN/m)	Advancing contact angle θa (°)	Receding contact angle θr (°)
None	—	31	32	12
Monoolein	0.2	21	43	17
Saturated monoglyceride	0.2	5.5	—	—
	2	4	125	100
Lactic acid esters of monoglyceride	0.2	23	45	23
	2	8	132	85
Lecithins				
Soybean phosphatidylcholine[a]	0.1	1.2	115	110
	1	1	160	143
Metarin P[a]	0.1	8.6	40	20
	1	0.9	65	35
Topcithin[a]	0.1	—	20	10
	1	0.9	120	45
Sterncithin[b]	0.1	—	15	8
	1	1	75	30
Ethoxylated alkyl ether	0.2	21	36	16
	2	2.1	135	69
Ethoxylated sorbitan monostearate	0.2	2.1	160	95
	2	0.7	175	175
Ethoxylated (12) castor oil	0.2	0.3	18	9
	2	~0	90	90
Ethoxylated (20) castor oil	0.2	0.3	3	3
	2	~0	90	90

[a] From Lucas Meyer (Hamburg, Germany).

[b] From Stern/Chimie (Hamburg, Germany).

two adjacent monolayers in contact (6) as schematically presented in Figure 2. An equation for interfacial tension has been derived by Fowkes (6) for systems dominated by dispersion forces, and modified to include the polar interaction with the water phase (11):

$$\gamma_{1,2} = \gamma_1 + \gamma_2 - 2 \cdot \sqrt{\gamma_1^d \cdot \gamma_2^d} - I_{1,2} \qquad [2]$$

where γ_1 and γ_2 are surface tensions of two adjacent phases 1 and 2; γ_1 and γ_2 consist of a dispersive (van der Waals) contribution (γ_1^d,γ_2^d), and a polar contribution (γ_1^p,γ_2^p). One term $\sqrt{\gamma_1^d \cdot \gamma_2^d}$ represents the van der Waals attraction of interfacial monolayer 1 by the bulk phase 2 (opposes term γ_1). The second term $\sqrt{\gamma_1^d \cdot \gamma_2^d}$ represents the van der Waals attraction of interfacial monolayer 2 by the bulk phase 1 (opposes term γ_2). $I_{1,2}$ represents the attraction of the nonpolar phase (oil) by the polar phase (water) due to polar interaction. $I_{1,2}$ opposes the γ_1 and γ_2 terms.

The components 1 and 2 in Equation 2 may be replaced by solid and oil (*SO*), solid and water (*SW*), and oil and water (*OW*), respectively, and combined with Equation 1. Because vegetable oil (*O*) and fat crystals (*S*) are nonpolar, one can assume that $I_{SO} = 0$, and $\gamma_O = \gamma_o^d$. After rearrangement, we obtain the following equation:

$$-\gamma_{OW} \cdot \cos\theta = -\gamma_{OW} + 2 \cdot (\sqrt{\gamma_o^d} - \sqrt{\gamma_w^d}) \cdot (\sqrt{\gamma_o^d} - \sqrt{\gamma_s^d}) + I_{sw} - I_{ow} \qquad [3]$$

For a given system (a combination of oil phase, water phase, and solid), the expression: $2 \cdot (\sqrt{\gamma_o^d} - \sqrt{\gamma_w^d}) \cdot (\sqrt{\gamma_o^d} - \sqrt{\gamma_s^d})$ may be treated as a constant, called *C*. We then get:

$$-\gamma_{ow} \cdot \cos\theta = -\gamma_{ow} + I_{sw} - I_{ow} + C \qquad [4]$$

or

$$I_{sw} - I_{ow} = \gamma_{ow} \cdot (1 - \cos\theta) - C \qquad [5]$$

The magnitude of this constant may be estimated as follows; γ_w^d is equal to ~22 mN/m (6,8,12); γ_o^d for soybean oil has not been determined. Values for oils are spread over the interval 32–36 mN/m. Two values are applied in our calculation: γ_o^d = 32.5 mN/m for oleic acid (12), and γ_o^d = 35.4 mN/m for cottonseed oil (13). The γ_s^d value for any type of fat crystal is not available in the literature. The density difference between the crystals (*S*) and oils (*O*) can be estimated as $\rho_S\rho_O \approx 1.1$. Because γ_s^d originates from van der Waals interaction, it is assumed to be proportional to the Hamaker constant as a first approximation ($\gamma^d \propto A_H$). The Hamaker constant depends on the density as follows (15):

$$A_H \propto \rho^2 \qquad [6]$$

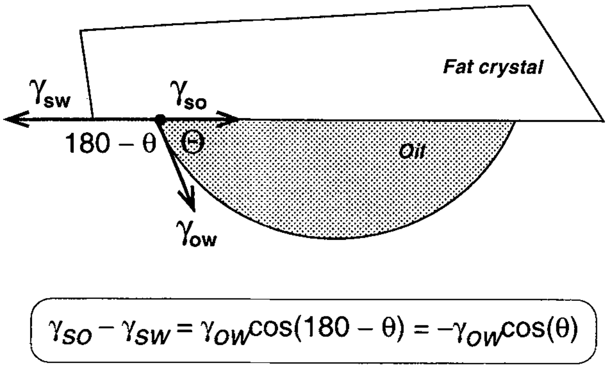

FIG. 1. Balance of tensions at fat crystal/oil/water interface and the Young equation (Ref. 5).

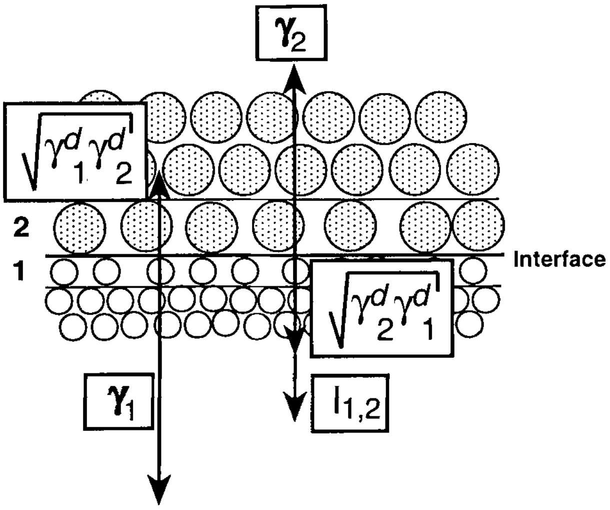

FIG. 2. Schematic sketch of the interface between two phases in contact. Arrows represent energy contributions to interfacial tensions.

Thus:

$$\gamma_s^d \approx \left(\frac{\rho_s}{\rho_o}\right)^2 \cdot \gamma_o^d \approx 1.21 \cdot \gamma_o^d \qquad [7]$$

With γ_o^d values for oleic acid and cottonseed oil, the γ_s^d is equal to 39.3 and 42.8 mN/m, respectively. The corresponding values for constant C are −1.1 and −1.5, with a mean value of −1.3 mN/m. Equations 4 and 5 can be written as:

$$1 - \cos\theta = \frac{I_{sw} - I_{ow} - 1.3}{\gamma_{ow}} \qquad [8]$$

Equation 8 shows that an increase in polar interaction for fat crystal/water (I_{sw}) leads to an increase in (1 − cos θ), a decrease in cos θ, and an increase in contact angle θ (highly polar crystals). A high I_{sw} term counteracts spreading of oil droplets on the water-covered fat crystal and is a driving force for fat crystals toward the water phase. The opposite, high polar interaction for oil/water (I_{ow}), leads to low (1-cos θ), high cos θ, and low contact angle θ (nonpolar crystals). High I_{ow} is a force that resists crystal penetration of the oil/water interface. The interfacial tension γ_{ow} represents also a resistance of the oil/water interface against fat crystal penetration (if γ_{ow} is high, high polar interaction excess $I_{sw} - I_{ow}$ is necessary to increase the contact angle θ).

Equation 5 was used to estimate the polar energy excess ($I_{sw} - I_{ow}$) for fat crystal/water over oil/water, and how it depends on additives. Figure 3 shows the influence of emulsifiers in the oil on the polar energy excess for palm stearin β′ and water. The polar energy excess was calculated both for crystals approaching the oil/water interface from the water side (advancing contact angles) and for crystals approaching the oil/water interface from the oil side of the emulsion (receding contact angles). Generally speaking, polar energy excess ($I_{sw} - I_{ow}$) describes a crystal's tendency to adsorb to the oil/water interface, and interfacial tension (γ_{ow}) represents the interfacial resistance to crystal adsorption.

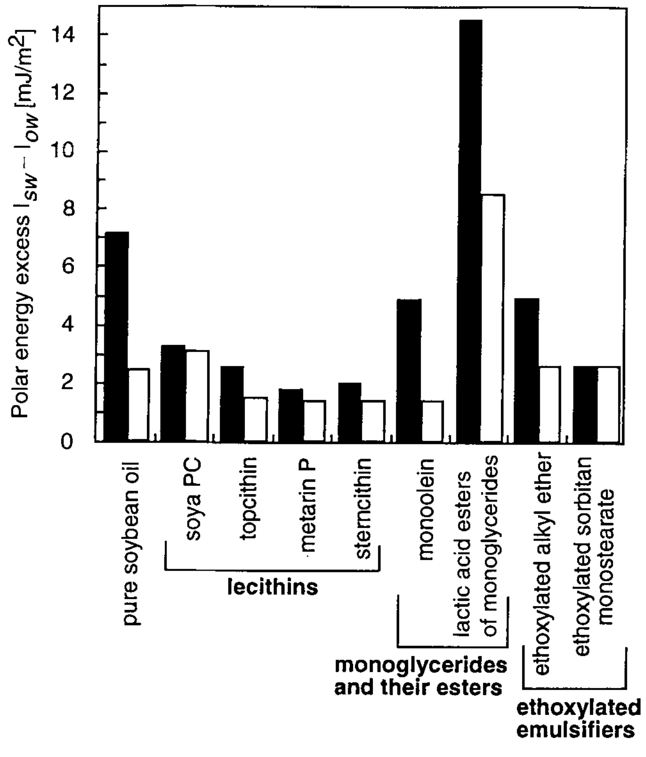

FIG. 3. The influence of emulsifiers (1–2%) in soybean oil on the polar energy excess ($I_{SW} - I_{OW}$) for fat crystals (palm stearin β′)/water over oil/water. The energy excess is calculated from advancing contact angles (fat crystals are initially pressed into the water phase) (■), and from receding contact angles (fat crystals are initially in the oil phase) (□). PC, phosphatidylcholine. Soya PC, Topcithin, and Metarin P from Lucas Meyer (Hamburg, Germany); sterncithin from Stern Chemie (Hamburg, Germany).

The polar energy excess is of the order of magnitude of 1–10 mJ/m^2. For a hypothetical particle of a diameter 1 µm, this energy excess is about $3 \cdot 10^{-15}$–$3 \cdot 10^{-14}$ J, corresponding to about 10^5–10^6 hydrogen bonds more per fat crystal unit surface area compared to the oil/water unit surface area. This energy is usually higher when the crystals approach the oil/water interface from the water phase, compared to approaching it from the oil phase, most likely because of a higher polarity of the crystal surface in water compared to oil. Polar energy excess is always positive. Consequently, fat crystals always have the tendency to adsorb to the oil/water interface, even in the absence of emulsifiers.

Lactic acid esters of monoglycerides (2%) gave the highest polar energy excess, followed by monoolein and ethoxylated alkyl ether. These emulsifiers are rather nonpolar and have a stronger tendency to adsorb to the fat crystal/oil interface compared to the oil/water interface. The polar energy excess is also high when no emulsifier is present in the oil. Thus, the fat crystal surface interacts more strongly with water than a liquid oil layer does. The lowest polar energy excess is given by hydrophobic lecithins, which have almost no preference for the fat crystal/water interface compared to the oil/water interface.

Displacement of the fat crystals from the oil/water interface. Stabilization of emulsions by solid particles at the oil/water interface is strong if the amount of energy to displace the particles from the interface is high. The displacement energy represents the adhesion strength for particles at the interface. This energy depends on contact angle and is expected to be highest for the angle 90° (2–4).

Displacement energy can be calculated for droplets stabilized by spherical monodisperse particles. The displacement energy to the oil phase (E_{oil}), normalized by division with a particle surface area, is a function of the interfacial tension between oil and water (γ) and the contact angle (θ) for fat crystal/oil/water (2–4):

$$\frac{E_{\text{oil}}}{\pi \cdot r^2} = -\gamma_{ow} \cdot (1 - \cos\theta) \qquad [9]$$

Equation 9 was used for calculations of displacement energies for fat crystals to the oil (1).

Polar energy excess vs. displacement energy. Results in Figure 4 show that the displacement energy to the oil is of the same order of magnitude as the polar energy excess for fat crystal/water over oil/water. Thus, the polar energy excess seems to determine how easy it is to displace fat crystals from the interface to the oil. In most cases examined, displacement energy is lower than polar energy excess, especially when this excess is relatively low. A strong interaction between fat crystals and oil phase must exist in this case, making it easy to displace the crystals from the interface to the oil. There are some deviations from this rule when the displacement energy is the same or slightly higher than the polar energy excess. Therefore, a large fraction of the energy to displace a fat crystal to the oil is needed for breaking the polar interaction crystal/water and replacing it with polar oil/water interaction. For 2% lactic acid esters of monoglyceride, the displacement energy is about double the polar interaction energy due to a high activation energy for displacement (high γ_{ow}) and/or due to additional nonpolar interaction that keeps the crystals at the droplet interfaces.

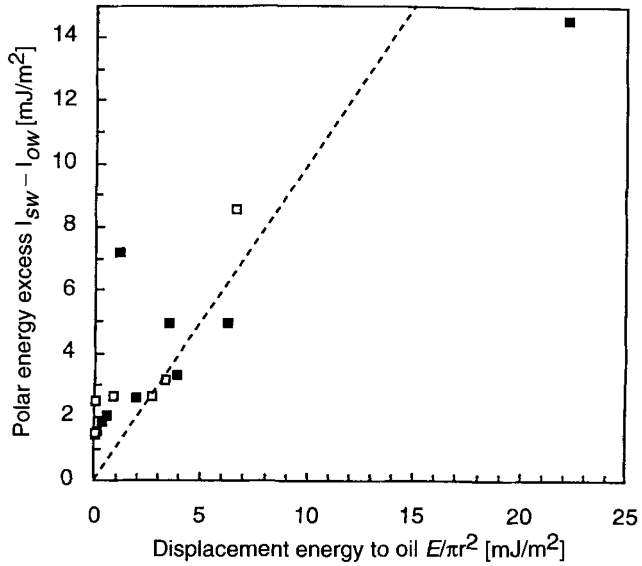

FIG. 4. A comparison of displacement energy ($E/\pi r^2$) (Eq. 9) for fat crystals at the oil/water interface to the oil, and polar energy excess ($l_{sw} - l_{ow}$) (Eq. 5) for fat crystals/water over oil/water for systems containing emulsifiers in the oil (1–2%). Displacement energy and polar energy excess were calculated for palm stearin β′ from advancing contact angels (fat crystals initially pressed into the water phase) (■) and from receding contact angles (fat crystals are initially in the oil phase) (□). The dotted line represents the case when the polar energy excess is equal to the displacement energy.

Adhesion tension ($-\gamma_{ow}\cos\theta$) vs. interfacial tension (γ_{ow}). A common way to present Equation 4 is to plot the adhesion tension $-\gamma_{ow} \cdot \cos\theta$ vs. the interfacial tension γ_{ow} (8). Graphs for advancing and receding contact angles are presented in Figure 5 for palm stearin β′ crystals at the soybean oil/water interface. Figure 5A shows that the data for receding contact angle form a straight line through the origin of slope −1, which describes the lower limit of contact angle $\theta = 0°$. This is in agreement with previous results (7,8). Data for advancing contact angle form a parallel line, which intersects the adhesion tension axis at 4 mJ/m^2. A straight line with slope −1 may be due to the constant contact angle θ in Equation 5, despite the addition of different emulsifiers to the oil. The unchanged contact angles on addition of low concentrations of emulsifiers, such as monoglycerides and hydrophobic lecithins, were fond in some studies (8). However, these contact angles should be 0° to get the slope −1, which was not the case.

The relationship observed in Figure 5A appears only for low contact angles and low polar energy excess ($I_{sw} - I_{ow}$), as indicated by Figure 5B.

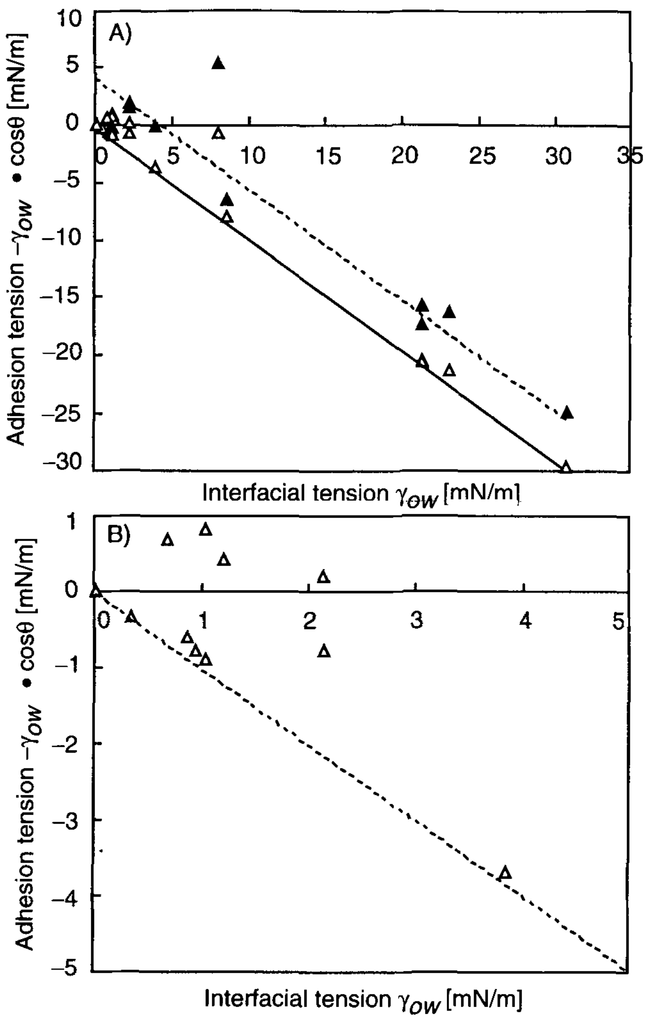

FIG. 5. Adhesion tension ($-g_{ow}$ • cos θ = $g_{so} - g_{sw} = -g_{ow} + I_{sw} - I_{ow} + C$) (Eqs. 2 and 4) for palm stearin β′ at the oil/water interface vs. interfacial tension (g_{ow}) between refined soybean oil and distilled water. A) All samples. Adhesion tension is calculated from receding (△) and advancing contact angles (▲), respectively. B) Samples close to origin (represent low interfacial tension and/or high contact angle). Adhesion tension is calculated from receding contact angles (△).

Fat crystal adsorption to oil/water interface (expressed by contact angle θ) is determined by the difference between polar interaction energies for fat crystal/water and oil/water ($I_{sw} - I_{ow}$, called polar energy excess). Interfacial tension constitutes a resistance force for crystal adsorption to the interface. The polar interaction energy for fat crystal/water is stronger than the polar interaction energy for oil/water in all cases examined ($I_{sw} - I_{ow} > 0$). The difference corresponds to about 10^5–10^6 hydrogen bonds for a hypothetical fat crystal of 1 μm diameter. The displacement energy for fat crystals to oil is lower than the polar energy excess for most cases examined. Thus, an additional nonpolar interaction between crystals and oil make it easy to displace them to the oil.

There is also a relationship between adhesion tension ($-\gamma_{ow}$ • cos θ) for the crystals at the interface and the interfacial tension γ_{ow}. A straight line with a slope −1 is achieved in systems with low interfacial tensions γ_{ow} and low polar energy excess ($I_{sw} - I_{ow}$).

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