of interfacial surface formation was less when two emulsifiers were used.

As indicated by the results, if two strongly hydrophilic emulsifiers are used, the resultant emulsion does not have good particle size, probably because of the limited number of oil-anchoring alkyl groups.

Summary

The bonds between some water-soluble emulsifiers and water are hydrogen bonds with low energies of dissociation, of the order of 7 kcal./mole. In the absence of other factors such bonds can be broken by the kinetic energy of motion at elevated temperatures.

The solubility of several emulsifiers was determined, and emulsions containing these emulsifiers at concentrations 2 to 4 times the amount required to make a monomolecular film of the oil droplets were made.

To provide emulsion stability at homogenization and sterilization temperatures the emulsifiers must be more hydrophilic than many oil-in-water emulsifiers that are satisfactory in ordinary use and must have an increased affinity for water in the temperature range of 5° to 120°. For a given type of emulsifier containing a given alkyl group, an optimum weight percentage of polyoxyethylene groups is required.

The solubility of an amine type emulsifier with the same alkyl group and approximately the same weight percentage of polyoxyethylene groups per molecule is greater than that of the corresponding amide compound, which, in turn, is more soluble than the corresponding ester type of emulsifier, because of differences in chemical type. Polyethylene-propylene oxide had the longest solubility range of the emulsifiers tested.

An increase in particle size or an appearance of two phases in emulsions prepared with emulsifiers which undergo solubility inversion below 85° was found. Emulsions prepared with emulsifiers whose inversion temperatures were above 85° maintained, generally, a low particle size on autoclaving, did not separate into a watery phase and an emulsion phase, and did not form a layer of oil.

Emulsions prepared with two emulsifiers, such that one had some lipophilic characteristics stronger than the other, were found to be stable and maintain a low particle size on autoclaving.

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Dilatometric Properties of Some Butyropalmitins, Butyrostearins, and Acetopalmitins[1]

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CERTAIN DI- AND TRIGLYCERIDES containing both short and long chain fatty acids normally solidify to waxy solids and as such possess a number of potential uses (3, 4). These glycerides are unique because of their ability to exist in waxy polymorphic forms which apparently are seldom thermodynamically stable but usually are stable for all practical purposes.

More than 20 articles and patents describe these unique glycerides, generally acetoglycerides, but only one of these articles is concerned with dilatometric behavior (6). It is a report of the dilatometric properties of 1-aceto-3-stearin and 1,2-diaceto-3-stearin. Two related articles are concerned primarily with other thermal properties and polymorphism of various glycerides (4, 7).

The present investigation was undertaken to obtain dilatometric and related data on six compounds which have heretofore not been described in the literature or for which information is meager. The six compounds, thought to have potential use in some practical applications, are: 1-butyro-3-palmitin, 1,2-dibutyro-3-palmitin, 1-butyro-3-stearin, 1,2-dibutyro-3-stearin, 1-aceto-3-palmitin, and 1,2-diaceto-3-palmitin.

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Materials and Procedures

Materials. Each of the aceto- and butyroglycerides was prepared by the direct esterification of 1-monopalmitin or 1-monostearin with either acetyl or butyryl chloride, using a procedure described previously (6). Briefly, the monoglyceride and acid chloride, each in chloroform solution, were mixed in the presence of pyridine and allowed to react at room temperature. After completion of the reaction the solution was washed successively with dilute acid, dilute alkali, and water. Then the chloroform was removed by stripping at a low temperature. The triglycerides were purified by several recrystallizations from acetone. The diglycerides were purified by cooling a hexane solution to remove the greater part of the uncombined monoglyceride, after which the partially purified diglycerides were recrystallized several times from acetone.

The monoglycerides used in the preparations possessed a purity of about 99%, estimated on the basis of melting points, hydroxyl values, and other analytical determinations. The acid chlorides were used as received from a manufacturer of reagent grade chemicals. The final aceto- and butyroglycerides possessed purities of 96% or better, with the triglycerides tending to be of higher purity than the diglycerides. The

TABLE I

Melting or Transition Points and Other Data for the Aceto- and Butyroglycerides

Compound	Melting or transition point[a] °C.			Density, grams per ml. at 50°C.	Hydroxyl value		Content of monoglyceride,[b] %
	Form						
	I	II	III		Found	Theory	
1-Aceto-3-palmitin	43.6	35.0[c]	−19	0.9483	161	150.6	3.7
1,2-Diaceto-3-palmitin	41.5	21.6[c]	−20.5	0.9522	1.8	0.0	0.0
1-Butyro-3-palmitin	42.4	39.6	29.4[c]	0.9354	146	140.0	2.4
1,2-Dibutyro-3-palmitin	10.5	2.8[c]	−34.5	0.9325	3.4	0.0	0.0
1-Butyro-3-stearin	48.4	42.2	37.5[c]	0.9283	135	130.9	1.3
1,2-Dibutyro-3-stearin	20.7[c]	.....	.....	0.9257	3.0	0.0	0.3

[a] Obtained by the capillary tube method.
[b] Calculated as monostearin or monopalmitin.
[c] Form obtained by cooling melt quickly to 0°C.

main impurities in the latter were small amounts of monoglycerides. Analytical data for the various final products are recorded in Table I.

Procedures. All dilatometric examinations were carried out in dilatometers of the gravimetric type, with mercury as the confining liquid. The construction of these dilatometers, their method of use, and methods employed in correcting dilatometer readings for the expansion of mercury and glass and making various other calculations were, with one exception, in accordance with previously published descriptions (1, 2).

The procedure of using the dilatometers was modified in that the precautions exercised by Bailey and Singleton (2) to reduce the effect of vacuole formation as far as possible were not taken. Vacuoles are most likely formed when a liquid fat is solidified in one polymorphic form and then transformed into another form without melting. In the present investigation a large proportion of the measurements were made on the polymorphic forms in which the compounds originally solidified. In the remaining measurements vacuole formation did not appear to be a significant factor because behavior characteristic of the presence of vacuoles was generally not observed and because dilatometric data could be duplicated under somewhat different conditions of chilling and tempering. It is probably significant that all of the glycerides when chilled rapidly or at a moderate rate first solidified to relatively soft, waxy solids which deform quite readily.

In preliminary experiments an attempt was made to obtain dilatometric curves by using fine crystals of the high-melting forms of the acetopalmitins, which were obtained by fractional crystallization from solvent. An apparently large shrinkage in volume occurred at the point where melting of the crystals in the dilatometer commenced. It is presumed that the mercury, which does not wet the crystals, was prevented by its high surface-tension from filling all of the crevices in the crystal faces. As the total surface area of the crystals was decreased by melting, the amount of voids between the crystal faces and the surface of the mercury decreased and an apparent shrinkage of the glycerides resulted.

In most of the dilatometric determinations the glyceride samples in the dilatometers were chilled to about −60°C. in a dry ice-acetone bath, after which the dilatometer was warmed to the melting point of mercury (−38.7°C.) and dilations with increasing temperature were recorded. In the determinations the conditions of chilling and tempering and the time intervals at successive temperatures were varied as required to obtain data on the various polymorphic forms.

As a guide to the dilatometric treatment of each glyceride and as an aid in the interpretation of the dilatometric data, melting and transition points of each of the glycerides were obtained by the capillary tube method. A sample of each glyceride was sealed in a capillary tube, after which the sample was melted and heated to about 100°C. and then chilled quickly in a dry ice-acetone bath. Then each sample was warmed slowly, and the temperatures at which melting or phase transformations occurred were noted. The phase transformations generally were detected by abrupt changes in translucence and other visual appearance. Each sample was further examined over short temperature intervals by the "thrust-in" technique, that is, each sample was solidified and tempered in a number of ways and then thrust into a constant temperature bath maintained just above or below the temperature where melting or a phase transformation was thought to occur. The recorded points of melting or phase transformation are in every case the average of the lowest temperature where a change

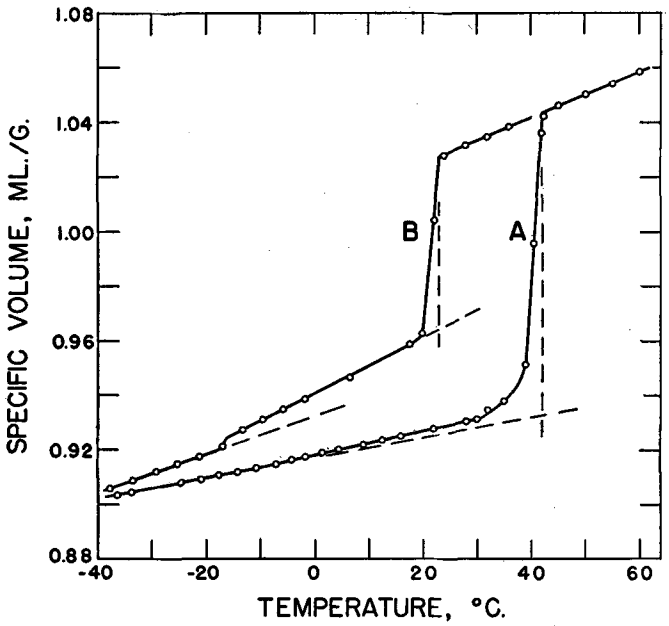

FIG. 1. Specific volumes of the polymorphic forms of 1,2-diaceto-3-palmitin as a function of temperature: *(A)* higher melting form and *(B)* lowest melting form obtained on quick chilling of melt (first portion of curve, −40 to −17°C.) and the intermediate melting form (portion of curve from about −17 to 23°C.).

TABLE II

Expansibility and Melting Dilation of the Aceto- and Butyroglycerides

Compound	Temperature interval, °C.	Polymorphic form	Coefficient of expansion, ml./gm./°C.	Melting dilation, ml./gm.	Dilatometric melting point, °C.
1-Aceto-3-palmitin	−38 to −10	I	0.000310	0.1081	41.5
	−38 to −21	II	0.000541	0.0821	34.8
	41.5 to 60	Liquid	0.000920	.....	.....
1,2-Diaceto-3-palmitin	−38 to −10	I	0.000383	0.1083	42.2
	−16 to 7	II	0.000976	0.0634	23.0
	−38 to −18	III	0.000674	.....	.....
	42.2 to 60	Liquid	0.000828	.....	.....
1-Butyro-3-palmitin	−38 to −14	I	0.000243	0.1199	42.0
	−38 to 2	III	0.000323	.....	.....
	42.0 to 60	Liquid	0.000903	.....	.....
1,2-Dibutyro-3-palmitin	−10 to 2	I	0.000640	0.0889	11.0
	.....	II	.....	0.0512	2.3
	11.0 to 60	Liquid	0.000857	.....	.....
1-Butyro-3-stearin	−38 to −14	I	0.000256	0.1283	48.8
	−38 to 18	III	0.000271	.....	.....
	48.8 to 60	Liquid	0.000877	.....	.....
1,2-Dibutyro-3-stearin	−38 to −20	I	0.000462	0.0858	20.2
	20.2 to 60	Liquid	0.000867	.....	.....

definitely was observed and the highest temperature where the change was not observed.

X-ray diffraction patterns were obtained for the most stable or highest melting form (Form I, Table I) of each of the glycerides which existed in the solid form at room temperature. The X-ray diffraction patterns were obtained with a General Electric Diffraction Unit, Model XRD,[3] using CuK_α radiation with a nickel filter (0.0007-in. thick). The long spacings were calculated from patterns obtained by photographing a small amount of sample pressed on a thin piece of glass and oscillated 15° in the X-ray beam. A plate distance of 10 cm. was used. The short spacings were calculated from patterns obtained by photographing finely powdered samples in sealed capillary tubes, using the XRD powder camera.

Results and Discussion

Dilatometric Data. The plots of volumetric expansion data for 1,2-diaceto-3-palmitin, Figure 1, are illustrative of the plots used to obtain the coefficients of expansion in the solid and liquid states, the melting dilations, and the dilatometric melting points recorded in Table II. Returning to Figure 1, the coefficient of expansion of the highest melting or most stable form (Form I) was determined from the slope of the lower, nearly horizontal portion of curve *A*, representing the tempered form of 1,2-diaceto-3-palmitin. This slope was measured at a temperature as low as possible in order to reduce the possibility of any premelting affecting the result. In the same manner the coefficient of expansion for Forms II and III, which were obtained by quick chilling of the melt, curve *B*, was determined at the lowest possible temperature.

The discontinuity or hump in curve *B* at the temperature of —17°C. represents the transformation of Form III to Form II without melting. This transformation occurs with a volume change of about 0.0035 ml./g. The change in the dilatometer occurred at about —17°C. and not at the temperature of —20.5°C. found by the capillary tube method. The shift in temperatures can be attributed, of course, to the slower heating and consequently greater tempering of the sample in the dilatometer.

The coefficient of expansion for the liquid 1,2-diaceto-3-palmitin was determined, of course, from the slope of the upper, nearly horizontal portions of curves *A* and *B*, Figure 1.

The dilatometric melting points for Forms I and II of 1,2-diaceto-3-palmitin were regarded as being determined by the intercepts of the middle portions of curves *A* and *B* with their respective upper horizontal portions, that is, the dilatometric melting points are the lowest temperatures at which complete melting occurred. These dilatometric melting points are considered to be more accurate than those obtained by the capillary tube method, mainly because the translucent nature of the compounds makes difficult the visual detection of the exact point of complete melting.

Melting dilations for the 1,2-diaceto-3-palmitin were determined from the lengths of the intercepts drawn perpendicular to the temperature axis at the dilatometric melting points and through the nearly horizontal portions of the curves or their extensions.

Comparison of Tables I and II reveals that dilatometric data were not obtained for a number of the melting or transition points determined by the capillary tube method. In general, changes at the points omitted from Table II were too rapid for the dilatometric technique. When 1-butyro-3-palmitin and 1-butyro-3-stearin were quickly chilled in the dilatometers and the temperature then was increased gradually, the lowest melting form of each compound transformed through the intermediate form and into the highest melting form far below the capillary melting and transition points for these forms. Conversion of 1-butyro-3-palmitin to the highest melting form apparently commenced at a temperature as low as 7°C. At room temperature both compounds transformed rapidly to the highest melting form.

The behavior of 1-butyro-3-palmitin and 1-butyro-3-stearin on examination by the capillary tube method provides additional information on the rapidity with which polymorphic changes occur. In capillary tubes the low-melting form of the compounds converts to the intermediate form in about 1 second at the melting point of the low-melting form. The meniscus of fat in the capillary definitely melts and resolidifies in

[3] This instrument is named as part of the exact experimental conditions. Naming it does not constitute a recommendation or endorsement of this instrument over that of any other manufacturer.

this period of time. The intermediate form of these diglycerides is somewhat more stable; at the transition temperature it changes to the stable form in the course of 15 to 30 min.

Some evidence was obtained of a fourth melting or transition point for 1-butyro-3-stearin at —6°C. though this was not definitely established. Unfortunately facilities for obtaining X-ray diffraction patterns at this temperature were not available.

In contrast to the behavior of the diglycerides containing butyric acid, the low-melting crystal forms of the diglyceride 1-aceto-3-palmitin appear to be more stable. The expansibility and melting dilation of the intermediate form of the latter compound could readily be obtained dilatometrically when examination was begun a few degrees below the melting point of the intermediate form.

As a group, the lower-melting polymorphic forms of the triglycerides which were examined were far more stable than those of the diglycerides. The most stable of the lower and intermediate forms encountered in the investigation was Form II of 1,2-dibutyro-3-palmitin. Tempering for several days at about 2°C. (just below the melting point of Form II) only started the transformation to the highest melting form. The transformation then was completed by tempering an additional two or three days at 5°C., which is between the melting points for Forms I and II. In the examination of the triglyceride 1,2-dibutyro-3-stearin some evidence of a polymorphic transformation at —21°C. was obtained, but, because this transformation point is doubtful, it was not recorded in Table I. Dilatometric data for Form III of 1,2-dibutyro-3-palmitin could not be obtained because the polymorphic changes occurred at about the melting point of the mercury used in the dilatometers.

Several of the melting or transition points recorded for the triglycerides, Table I, have heretofore not been reported. In an earlier investigation of these same triglycerides Jackson and Lutton (4) failed to observe some of the points at which polymorphic transformations occur. Because the finding of melting and transition points involves some element of chance, it is possible that points other than those recorded in Table I do exist.

The melting dilations recorded in Table II for the various crystal forms of the aceto- and butyroglycerides are, as a group, smaller than those for tristearin and tripalmitin. The melting dilation for tristearin ranges from 0.1190 to 0.1674 ml./g., the exact value depending on the crystal form. The melting dilation for the highest melting form of tripalmitin is 0.1619 ml./g. By contrast the melting dilations for the aceto- and butyroglycerides range between 0.0512 to 0.1283 ml./g.

The aceto- and butyroglycerides in the liquid state possess coefficients of expansion approximating those of tristearin and tripalmitin. However there is no such agreement between the coefficients of expansion of the various crystal forms. For tristearin the coefficents range from 0.000227 to 0.000324 ml./g./°C., depending upon the crystal form. Some of the coefficients for the aceto- and butyroglycerides are three to four times as great. There is no obvious relationship between the coefficients for the different aceto- and butyroglycerides and their polymorphic form number or chemical structure. Among the different compounds a similar polymorphic form number is, of course, no indication of a similarity in crystal structure, that is, a term like "Form I" is not indicative of a certain type of crystal arrangement.

X-Ray Diffraction Data. To define to some extent the highest melting forms of the aceto- and butyroglycerides in terms of the generally accepted nomenclature of polymorphism based on X-ray diffraction patterns (5), the Form I of those compounds solid at room temperature was examined by the X-ray technique. The calculated long spacings and the principal short spacings associated with each compound are recorded in Table III.

TABLE III

Interplanar Spacings of Highest Melting Polymorphic Forms of Some Aceto- and Butyroglycerides[a]

Compound	Long spacing, Å	Principal short spacings, Å	Polymorphic form
1,2-Diaceto-3-palmitin	31.84	4.77 S 4.58 VS 3.76 VS	Beta
1-Aceto-3-palmitin	50.46	4.38 S 4.14 VS 3.54 MS	Beta prime
1-Butyro-3-palmitin	65.46	4.59 VS 3.85 VS 3.76 VS	Beta
1-Butyro-3-stearin	71.15	4.56 VS 3.84 VS 3.75 VS	Beta

[a] Highest melting polymorphic forms are designated as "Form I" in Tables I and II.

The polymorphic designations ordinarily associated with the short spacings recorded in the third column of the table are listed in the fourth column. The finding that the most stable forms of 1-aceto-3-palmitin and 1,2-diaceto-3-palmitin are the beta prime and beta forms, respectively, is in accord with earlier evidence. Jackson and Lutton (4) found the thermodynamically stable form of 1,2-diaceto-3-palmitin and 1,2-diaceto-3-stearin to be beta. Also Vicknair *et al.* (6) found the stable forms of 1-aceto-3-stearin and 1,2-diaceto-3-stearin to be the beta prime and beta forms, respectively. The 1-butyro-3-palmitin and 1-butyro-3-stearin differ from their aceto homologs in that the beta form is the most stable.

The long spacings recorded in Table III, which are characteristic of crystal structure in the long chain direction, cannot in all instances be related to a given chain length multiplicity. The long spacing of 31.84 Å for 1,2-diaceto-3-palmitin agrees with that found by Jackson and Lutton (4) and indicates the triple chain length structure shown in Figure 2 A. The long spacing of 50.46 Å for 1-aceto-3-palmitin agrees with a triple chain length structure of a somewhat different form, shown as Figure 2 B. The long spacings of

FIG. 2. Postulated triple chain length structure of *(A)* the beta form of 1,2-diaceto-3-palmitin and *(B)* the beta prime form of 1-aceto-3-palmitin.

65.46 and 71.15 Å for the 1-butyro-3-palmitin and 1-butyro-3-stearin, respectively, can not at this time be associated with a given structure.

Summary

1. Six aceto- and butyroglycerides, including di- and triglycerides, were prepared and purified, and a number of their physical properties were determined.

2. Melting and transition points for each of the glycerides were determined by experiments with small portions of the samples in capillary tubes; the "thrust-in" technique was employed to detect some polymorphic transitions occurring in intervals as short as one second.

3. Using the data on melting and transition points, dilatometric data were obtained, insofar as possible, for the various crystalline modifications of the compounds as well as for the liquid state. From the dilatometric data calculations were made for expansibilities in the liquid and various solid states, and melting dilations for several solid states. In one instance the volume change on phase transition, without melting, was calculated.

4. From X-ray diffraction patterns the long and short crystal spacings for the highest melting forms of several of the glycerides were determined. On the basis of the short spacings, polymorphic designations commonly used for fats and oils were assigned. The long spacings obtained indicated a triple chain length structure for some of the compounds and as yet an undetermined structure for the other compounds.

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Letter to the Editor

March 12, 1956.

In the communication, "The Influence of Dietary Fat on the Glyceride Structure of Animal Fats," Reiser and Dieckert (1) state: "Kartha's concept is that all lipases have equal affinity for all fatty acids and *vice versa*" and "although Kartha has assumed that lipases are unselective in their action, such is certainly not the case." These statements make it appear that perhaps Kartha's views should be stated more clearly and completely. The explanation of glyceride structure of natural fats suggested by Kartha (2, 3) was based on the following evidence found in the literature: a) plant and animal lipases are non-specific towards the alpha and beta hydroxyls of the glycerol (4); b) plant lipases are inactive on acids of lower chain-length than C_7 and act non-specifically on all fatty acids above this (5); c) adipose tissue lipases (same properties as pancreas lipase) are inactive on acetic acid but with other saturated acids show increased rates of action as chain-length decreases, and with triglyceride of unsaturated acids from C_{16}–C_{22} reactivity at lower temperatures increases with the number of double bonds (6); d) all lipases act reversibly (7); e) in animal depots as also in maturing seeds, fats are always in presence of active lipase (8).

The reversibility of lipatic reaction leads to dynamic equilibrium, which eliminates the influence of specificities of action of lipases as regards the ultimate distribution of the acids among the glycerol molecules. In such a system there will naturally be continuous interchange of acid radicals between different positions of the same or different glycerol molecules and all initially formed products will be rearranged gradually in the direction of probability requirements wherein the final structure will depend entirely on the proportions of the different fatty acids. In such systems deviation from simple chance distribution is possible only on the basis of limits in the formation of some of the triglycerides required according to chance.

Reiser *et al.* (1) further quote Kartha as saying that "in adipose tissues, as well as mammary glands of animals, fat can be deposited from ingested foods without affecting the normal glyceride type of distribution." The conditions under which ingested fats can be deposited without disturbing the normal glyceride structure pattern have been described elsewhere (9); under other conditions distribution patterns can change within certain limits (9).

The evidence of Reiser *et al.* that endogenous animal fats may contain GS_3 in proportions measurably higher than those required according to random distribution (1) would, if confirmed, raise serious doubt of the validity of the restricted random distribution theory, at least when applied to animal fats. However the isotope dilution method used by them for the GS_3 determinations is based on the premise, which has yet to be established, that added labelled GS_3 does not accumulate in the precipitates or the mother liquors in the course of several crystallizations. If the isolated GS_3 contains less than the representative quantity of labelled tripalmitin, the result of the determination of GS_3 will be too high.

The component acids of the GS_3 of both chicken and rat fats are predominantly palmitic, but both fats contain significant and variable quantities of stearic acid (10). It is therefore possible that some of the values for GS_3 found by R and D are too high because the labelled tripalmitin in the recrystallized fractions was not representative of the whole of the GS_3.

Furthermore, because of the relatively high proportions of palmitic acid in the GS_3, the component acids of the other fractions being predominantly C_{18} or higher, the results reported by R and D in terms of weight percentage are misleading. Recalculation of the results in terms of mol percentage on the as-