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M. F. MILLER, *Director*

A Study of the Crystallization and Occurrence of Lactose Crystals in Several Milk Products

C. W. DECKER AND W. H. E. REID

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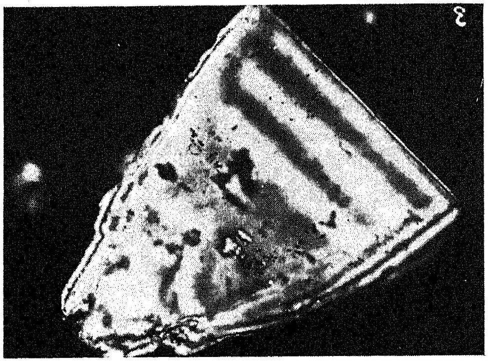

Alpha Lactose
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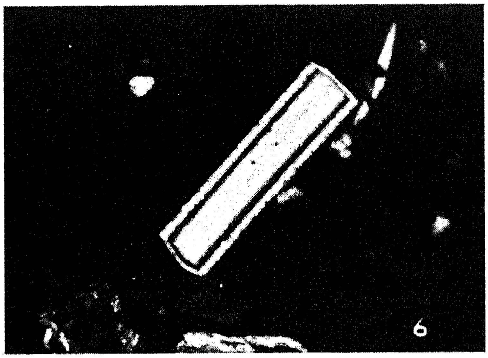

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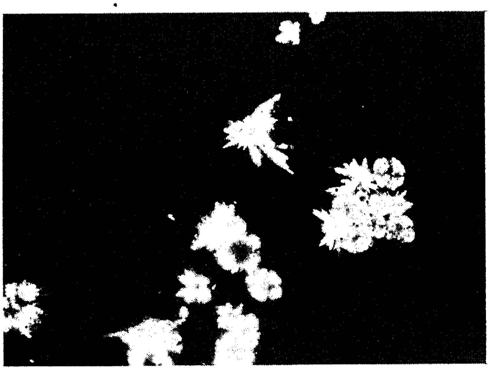


Alpha Lactose, Sandy Sample
70X

Fig. 1.—Alpha lactose, beta lactose, and needle-like alpha lactose crystals removed from "sandy" ice cream and shown in polarized light.

A Study of the Crystallization and Occurrence of Lactose Crystals in Several Milk Products

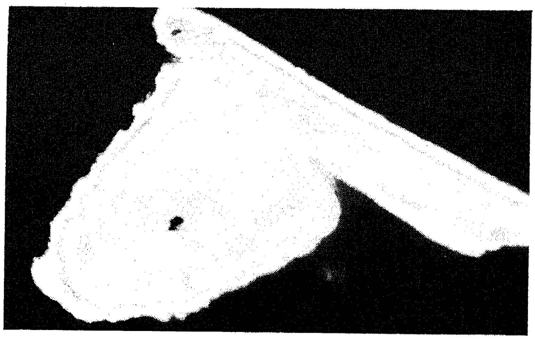
C. W. DECKER AND W. H. E. REID

There are numerous reasons which lead one to believe that lactose should crystallize out of ice cream during the hardening process. Agitation occurs in the freezer, the concentration of lactose in the ice cream is sufficient to be in the labile area, and probable nuclei which initiage crystallization are present; however, the phenomenon of lactose crystallizing out of ice cream during the hardening process has not been reported in the literature.

The investigations of lactose as it occurs in ice cream have been primarily concerned with the study of lactose crystals in a palpable state in which they are detectable in the mouth, which condition is known as "sandiness." The study of lactose as it exists in ice cream before reaching the palpable state has been quite limited. Polarimetric studies have been carried out by Troy and Sharp (1930), who concluded that lactose exists in a supersaturated state. However, this represents an indirect method of study as the ice cream must be melted rapidly, the proteins precipitated with mercuric chloride and alcohol, promptly filtered, and the filtrate examined in the polariscope. In the application of such a procedure it is possible that small amounts of crystalline lactose go into solution and other difficulties may be experienced. It is believed that the microscopic examination of lactose presents a more satisfactory approach to this problem. However, when using plain light as a source of illumination, it is difficult to detect lactose crystals in the initial stages of growth. This suggests the use of polarized light and the petrographic or polarizing microscope for the study of lactose crystals in ice cream.

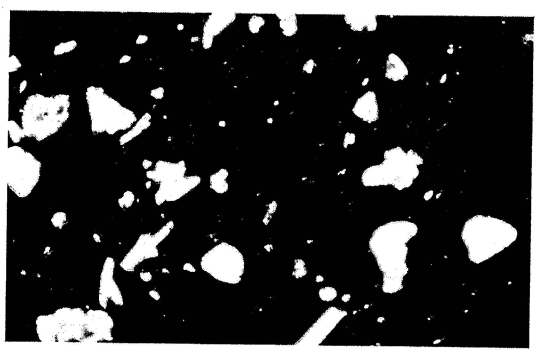
The periodic examination of ice cream for lactose development was suggested by a study made in 1939 by Decker, Arbuckle and Reid on sandy material isolated from aged, heat shocked, sandy ice cream. A tomahawk crystal was identified optically as alpha hydrate lactose and a columnar crystal as beta anhydride lactose. The ice cream studied contained 15.5 per cent serum solids and 42.8 per cent total solids. It was placed in a dispensing cabinet at 6 and 10° F., respectively, for 3 months and subsequently in a hardening room for 5 months.

Fig. 1 shows alpha lactose and beta lactose crystals in the sample of aged experimental ice cream, and a needle-like alpha lactose crystal in the process of development in a sample of sandy commercial ice cream.



Alpha and Beta Lactose
324X

Beta Lactose
144X



Proportion of Alpha and Beta
72X

Fig. 2.—Microscopic fields of alpha and beta lactose crystals removed from "sandy" ice cream and shown in polarized light.

Fig. 2 shows an alpha and beta lactose crystal together, in a field in which beta lactose crystals predominate, and a field showing the relative proportion of alpha and beta lactose crystals in the sandy material.

The great variation in the crystalline habit of the lactose crystals isolated from different samples of sandy ice cream prompted a study of the complete process of development of lactose crystals in ice cream.

REVIEW OF LITERATURE

Lactose

Lactose, also known as milk sugar, is a disaccharide, $C_{12}H_{22}O_{11}$, and is known to occur in the milk of all land mammals. It is of importance in ice cream because it comprises from 5 to 7 per cent of the ice cream mix.

Lactose may exist in three homogeneous forms. When crystallization occurs above 93.5° C. and anhydrous form known as beta anhydride results, whereas when crystallization occurs below this temperature a hydrate form known as alpha hydrate results. A third form may be prepared by drying under vacuum at any convenient temperature above 65° C. and is called alpha anhydride. At a temperature less than 93.5° C. the alpha and beta forms exist in solution in a definite ratio varying slightly with temperature. The two forms differ chemically in that alpha lactose is alpha-glucose-beta-galactoside and beta lactose is beta-glucose-beta-galactoside. A change in the OH and O in the aldehyde group of the glucose end of the molecule is responsible for the rotational differences of the two forms.

Hudson (1908) found the specific rotation for alpha-lactose to be +86° at 25° C. and for beta lactose to be +35.3° when the specific rotation of the equilibrium mixture was taken as +55.3. He considered the equilibrium relation to be:

alpha-anhydride + H_2O hydrated form H_2O + beta-
(fast) (slow)
anhydride.

Gillis (1920) expressed his view of the equilibrium relation as follows:

alpha-anhydride + H_2O alpha-hydrate

beta-anhydride + H_2O beta-hydrate

The hydration takes place almost instantaneously while the change from alpha to beta occurs more slowly and is the basis of the mutarotion phenomena. He postulates the presence of beta-hydrate although such a substance has never been isolated and he attributes this to the fact it is still more soluble than beta-anhydride.

Hunziker and Nissen (1927) furnished the crystallographic features of alpha hydrate lactose. Lactose crystals have trapezoidal side faces and rhombic top and bottom faces, belong to class C, and have one axis of symmetry. The lactose crystal has, in addition, beveled faces at the bottom which may terminate in a sharp edge, giving the crystal a distinct tomahawk appearance.

Wherry (1928) determined the optical data and crystallographic features of alpha lactose, beta lactose and sucrose. He gives the following refractive indices:

	Refractive Indices		
Sugar	Alpha	Beta	Gamma
Alpha lactose	1.517	1.550	1.555
Beta lactose	1.542	1.572	1.585
Sucrose	1.540	1.567	1.572

Crystallographically, he states, beta lactose belongs to the holo-axial-polar (sphenoidal) class of the mono-clinic system.

Lactose Crystallization from Water Solutions

Extensive research has been done on lactose crystallization from water solutions, the factors affecting it, and its application to more complicated solutions such as condensed milk and ice cream. Among the factors of importance studied were time, temperature, solubilities, seeding, mutarotation of equilibrium ratio of alpha to beta lactose and factors affecting it such as acids, bases, salts, and other substances.

When a supersaturated solution of lactose crystallizes, alpha lactose crystallizes equal to the degree of supersaturation, and some beta lactose must then change to alpha lactose to restore the equilibrium ratio between the two forms in solution. This ratio exists as a definite value for a given temperature and varies to some extent with temperature, i. e., the amount of beta in proportion to alpha increases as the temperature decreases. Therefore, the rate of change of beta to alpha has an important influence upon crystallization of lactose.

Solubility of Lactose and Factors Affecting It.—Hudson (1908) gives the initial solubility of lactose at O° C. as five parts of lactose per 100 parts of water and a final solubility of 11.9 parts per 100 parts of water; the solubility of beta lactose as 45.1 parts per 100 parts of water; and solubilities for a wide range of temperatures are also stated.

Rahn and Sharp (1928) obtained from the solubility curves of Hudson (1908) the solubility values for lactose at -10, -20 and -30° C. of 9.41, 7.47, and 5.95 gm., respectively, per 100 gm. of water.

Hunziker and Nissen (1926) report that the presence of sucrose decreased the solubility of lactose and that this decrease is slight in dilute sucrose solutions such as are present in ice cream mixes, but becomes greater as the sucrose concentration increases, as in sweetened condensed milk, where it amounts to approximately 15 per cent of the lactose solubility in aqueous solutions. They state that colloids did not have any material influence on the solubility of lactose.

Herrington (1934c) found that many salts combine with lactose and the existence of such molecular compounds is possibly a factor contributing to the stability of supersaturated solutions of lactose such as are found in ice cream and milk powders. Lactose is more soluble in molar solutions of calcium chloride or of calcium nitrate than in pure water, and an increase in the concentration of salt further increases the solubility of lactose.

Leighton and Peter (1923) froze solid solutions of 10 parts of water without visible appearance of lactose crystals. In one instance they obtained a concentration of over 70 parts in 100 parts of water, with the crystals which appeared fine enough to go through filter paper, and thus they concluded that lactose exhibited a wide labile area.

Mutarotation of Alpha and Beta Lactose and Factors Affecting It.—Hudson (1904) reported the relative changes of beta to alpha lactose in one hour and gave an equation for the determination of the transformation constant which Rahn and Sharp (1928), by inter- and extrapolation determined, the amount changed. Both sets of values are given in the following table:

Temperature °C.	Per Cent Changed in One Hour Hudson	Rahn and Sharp
0	3.4	2.8
—10	..	0.85
—20	..	0.23
—30	..	0.055

Herrington (1934a), in a study of the effect of acids and salts upon the mutarotation velocity of lactose, found that they had an important influence which he attributed to general acid and base catalysis.

Troy and Sharp (1930) studied the influence of pH upon rate of change of beta to alpha lactose and found it to be at a minimum between pH 2 and 7. The rate approached infinity at a pH of O and 9. The effect of pH on the rate of change of alpha to beta lactose influenced the rate of solution of alpha lactose. The effect of pH on the rate of change of alpha to beta lactose influenced the rate of precipitation of alpha lactose. Relatively concentrated sucrose solutions had little effect upon the rate of change of the two forms of lactose into each other.

Whitaker (1933) recommended a rapid attainment of low temperature in hardening the ice cream as the rate of conversion of beta to alpha is decreased under such conditions and crystallization is delayed.

Crystallization Habit of Lactose and Factors Affecting It.—Herrington (1934b) found that the crystallizing habit of lactose varied greatly under different conditions of crystallization. The principal factor governing the crystalline habit of lactose was the precipitation pressure and the ratio of the actual concentration to the solubility. Sucrose had a precipitating effect upon lactose and influenced its crystallization. He reported that both alpha hydrate and beta anhydride will form needles of crystallization is sufficiently rapid, but they may be distinguished by the fact that alpha hydrate prisms are always straight while those of beta anhydride are curved. Under favorable conditions he was able to grow crystals with 13 faces and occasionally with as many as 16, although crystals with more than 13 faces are extremely rare.

Hunziker and Nissen (1927) described the change in lactose crystal shapes as slight in the presence of 14 per cent sucrose solution. The crystals are shorter and thicker but possess full development resembling short truncated pyramids with flat rhomboid base and apex. Crystallo-graphically the crystals were the same, but the crystalline habit appears to be altered.

Lactose Crystallization in Ice Cream and Factors Affecting It

There are three possibilities to consider in the crystallization of lactose from ice cream. First is that alpha lactose always crystallizes out similar to the crystallization of lactose in sweetened condensed milk, and the size of the lactose crystals determines the existence of a "sandy" condition in the ice cream; second, beta lactose as well as alpha lactose crystallizes out; and third, that lactose exists in a supersaturated condition.

These possibilities were considered by Rahn and Sharp (1928), Troy and Sharp (1930), and a discussion and summarization of their findings by Sommer (1935). These investigators concluded that lactose existed in ice cream in a supersaturated condition. It is of interest to see how they arrived at this conclusion and to discuss the three possibilities of lactose crystallization:

Rahn and Sharp (1928) theorized that the crystallization and development of lactose in ice cream were based upon the first two possibilities, that both alpha and beta lactose crystallized out, and that the growth of alpha hydrate crystals was controlled by the temperature effect upon the rate of change of alpha to beta lactose. That lactose crystals are not found in good ice cream does not preclude their absence. Only alpha lactose produces sandy material since beta lactose, should it crystallize out, would form such small crystals or be so soluble that they could not be detected. At -10° C. the crystal size is much greater than at -24° C. as the rate of change of beta to alpha lactose is much more rapid.

Troy and Sharp (1930) proved to their satisfaction by a simple seeding test, that the theory advanced by Rahn and Sharp was untenable. An ice cream containing alpha lactose crystals would induce copious crystallization in a supersaturated lactose solution. They admitted, however, that "sandy" ice cream did not give as satisfactory results in the seeding tests as did other dairy products. They then made a polarimeter test by melting the ice cream rapidly, precipitating the proteins with mercuric chloride, and alcohol, filtering rapidly, and then examining in a polariscope to identify "sandy" ice cream. If any alpha lactose is present the equilibrium ratio changes and consequently the rotation is altered. In normal ice cream they found no indication of lactose crystallization except in one case which evidenced incipient sandiness.

Sommers (1935) says, "These results point quite definitely to the conclusion that normal ice cream does not contain lactose crystals."

"With a moderate serum solids content the lactose content is low enough so that freezing can progress to a considerable extent before the unfrozen portion reaches a point of spontaneous crystallization of lactose. When that point has been reached the unfrozen portion is then so viscous that it is in the glass state or at least in a condition where crystallization will be exceedingly slow. With a higher serum solids content the point of spontaneous crystallization is reached earlier, the unfrozen portion is less viscous, and crystallization can proceed more rapidly."

"Even in a mix of 6 per cent serum solids and 30 per cent total solids the saturation point must be reached during freezing. In such a mix there would be about 3.24 parts of lactose to 70 parts of water, but when 70 per cent of the ice was frozen there would only be 21 parts of water to hold the lactose equivalent to 15.4 parts of lactose to 100 parts of water at 0° C. This means the problem is not one of keeping the lactose content below the saturation point."

Herrington (1934) stated that original concentration of lactose in ice cream was not so important after freezing had begun because the actual concentration of the unfrozen syrup was then dependent only upon the temperature and not upon the original concentration.

Zoller (1924) found that about 7 per cent of water was still unfrozen in ice cream containing 14 per cent sugar, 10.5 per cent serum solids, and 10 per cent fat, held at -24° C. for 7 days. Rahn and Sharp (1928) calculated this to be 127 parts of lactose to 100 parts of water and equal to saturation at 85° C., or 109° C. greater than saturation value.

Leighton (1923) observed that forewarming of milk causes a precipitation of some of the milk salts which may act as nuclei, and states that if no nuclei are present the lactose concentration in ice cream reaches the labile area.

Homberger and Cole (1933) observed lactose crystals in an ice cream a few hours after manufacturing. This particular ice cream contained 16 per cent serum solids and 41.35 per cent total solids, and was held at -3 to -4° F. and -7 to 11° F., respectively, in a storage cabinet.

Sandiness in Ice Cream

When lactose crystallizes in ice cream and the lactose crystals are large enough to be detected in the mouth, the ice cream is termed "sandy." Bothel (1920) was the first to observe that sandiness in ice cream was due to the presence of lactose crystals

Zoller and Williams (1921) isolated sandy material from ice cream and identified it chemically and optically as alpha lactose.

Homberger and Cole (1933) observed that lactose crystals when first palpable or detectable in the mouth are from 16 to 30 microns in size.

Sommer (1935) stated that the fundamental explanations of the problem of sandiness are inadequate.

Troy and Sharp (1930) explained the formation of "sandy" ice cream as being due to the presence of sufficient non-frozen water to permit the alpha lactose to crystallize. Rapid freezing of the ice cream may concentrate the lactose solution to a region of supersaturation where the crystallization of lactose does not occur. Alpha lactose crystals may form in such ice cream if sufficient ice is melted to dilute the syrup to a range of concentration where crystallization may occur.

Factors Affecting Sandiness

Temperature Fluctuations.—Temperature fluctuations have been shown to be important in the development of sandiness by Dahle (1923, 1929), Hall (1924), Reid (1930), Homberger and Cole (1933), and Whitaker (1933). These temperature fluctuations of the ice cream may occur at any time after freezing but are most likely to occur in the dealer's cabinet.

Serum Solid Content.—Leighton (1923) believed that it would not be possible to use a high concentration of lactose in ice cream without producing sandiness.

Dahle (1923) reported that a lactose content of 8.5 per cent of the water present was capable of producing sandiness and that a water content 6.4 times as high as the serum solids content is required if sandiness is to be avoided. Sommer (1935) found that this value gave too low a serum solids content in the mix as judged by practical experience and the influence of other factors, and developed an equation for calculating the highest serum solids content permissible under different commercial conditions.

To avoid sandiness, Webb and Williams (1934) and Corbett (1939) recommended a low lactose source of serum solids.

Presence of Inert Nuclei and Nut Meats.—Dahle (1929) and Reid (1930) have shown that nut meats tend to induce sandiness. The latter washed the nut meats in warm water and autoclaved them to delay sandiness, while the former soaked the nut meats in a syrup solution to retard sandiness.

Whitaker (1933) accounted for the increased tendency toward sandiness when nut neats were used by the assumption that unsoaked nut meats absorbed water from the ice cream, thereby increasing the lactose concentration and its tendency to crystallize. He also observed that the presence of inert nuclei was an important factor in producing sandiness.

Freezing and Hardening.—Whitaker (1933) maintained that lower drawing temperature in a batch freezer increased the tendency toward sandiness, while Homberg and Cole (1933) took the opposite viewpoint and reported lower drawing temperatures decreased sandiness when using a continuous freezer. These opposing viewpoints are explainable since the freezing process requires from 5 to 8 minutes in a batch freezer and to obtain a lower drawing temperature the ice cream must be left longer in the freezer; while only 15 seconds is required for the freezing process in a continuous freezer and the drawing temperature is independent of the time in the freezer.

Other Ingredients.—Erb (1931) found that dextrose failed to control sandiness.

Anthony and Lund (1931) retarded sandiness with partial replacement of sucrose with dextrose and also decreased the size of lactose crystals formed.

Homberger and Cole (1933) found that 30 per cent replacement of sucrose with dextrose decreased the size of lactose crystals formed, but increased the number of lactose crystals. They concluded that the quantity and size of lactose crystals were both important in producing a "sandy" condition. They also observed that the addition of skim milk powder at the freezer, thereby "seeding" the ice cream, tended to delay sandiness.

Whitaker (1933) varied the homogenizing pressures and produced fat clumping, thereby decreasing the tendency to form sandiness, which he attributed to the increased viscosity. He also increased the viscosity by standardizing the acidity with divalent salts and obtained the same results.

Types of Lactose Crystals Found in Sandy Ice Cream

Zoller and Williams (1921) centrifuged tomahawk shaped crystals of lactose from ice cream and identified them as alpha lactose.

Williams and Peter (1930) reported the finding of a new diamond-shaped crystal which they identified as alpha lactose.

Homberger and Cole (1933) described a tomahawk crystal, a diamond-shaped crystal, and needle-like crystals radiating out from the faces in the early stages of crystallization which disappeared in a few days and were replaced by a shorter maize-shaped crystal.

Decker, Arbuckle and Reid (1939) identified optically a tomahawk crystal as alpha lactose and a columnar shaped crystal as beta lactose.

Arbuckle (1940) prepared a series of mixes and was able to isolate alpha lactose crystals of several different crystalline habits. Those found included a fully developed lactose crystal, thin triangular crystals which developed into the saw-tooth lactose crystals, and thin diamond-shaped crystals which developed into short truncated crystals.

PROCEDURE

The ice creams included in this study were made over a 3 year period. Commercial practices of manufacture were applied with laboratory control. The ingredients used in the different mixes included sweet cream, whole milk, skim milk, sucrose, dried skim milk and gelatin.

Method of Processing

The method of processing the mixes was as follows: (1) pasteurization at 155° F. (68.3° C.) for 30 minutes; homogenization at the pasteurization temperature at pressures of 1500-1000 pounds on the first and second stages, respectively; (3) immediate cooling to 40° F. (4.4° C.); and aging for variable periods of time, usually 2 to 20 hours.

Method of Freezing and Types of Freezers Used

When using both a direct expansion batch freezer of 40 quart capacity and a Vogt continuous freezer with a capacity of 85 gallons per hour, the ice creams were drawn at 24° F. (-4.45° C.) with an overrun of 100 per cent.

Method of Seeding the Mixes

During the freezing process skim milk powder was added gradually to the mix at the rate of 2.5 pounds to each 5 gallons of ice cream mix containing 10.5 per cent serum solids.

Microscopic Examination and X-ray Analysis of Skim Milk Powder for Crystalline Lactose

Several samples of spray process dry skim milk of a grade suitable for use in ice cream manufacture were examined with the petrographic microscope using polarized light and a magnification of 900 X. The dry skim milk was placed in an immersion oil of known refractive index and a cover slip placed on top. Examination was made under polarized light for birefringent material as well as for other optical characteristics, and representative sample was photomicrographed.

An x-ray analysis on two samples of dry skim milk was made by the Physics Department of the University of Missouri, with crystal reflected radiation of 0.71×10^{-8} cm., recorded on a cylindrical camera of 12 cm. radius, and an intensity frequency curve made from the negative.

Microscopic Examination of Ice Cream

A petrographic microscope utilizing polarized light was used to study the crystallization and development of lactose in ice cream, as it provides an excellent means for the detection of birefringent or crystalline material. Between crossed nicols the background appears dark because the nicols polarize light in two planes at right angles to each other, which are then extinguished. The crystalline material shows bright against the dark background of crossed nicols by virtue of the retardation of light rays to produce visible light.

The ice creams were placed in a hardening room having an average temperature range from -8 to -12° F. (-22.2 to -24.4° C.) and examined at definite time intervals, as hourly intervals of 0, 4, 6, 8, 12 and 24 hours, and daily intervals of 7, 28 and 56 days. In a few instances other time intervals were used.

The ice cream samples were prepared for microscopic examination by a procedure involving two techniques of preparation. Fig. 3 shows the laboratory equipment used in the microscopic examination of ice creams for the presence of lactose crystals.

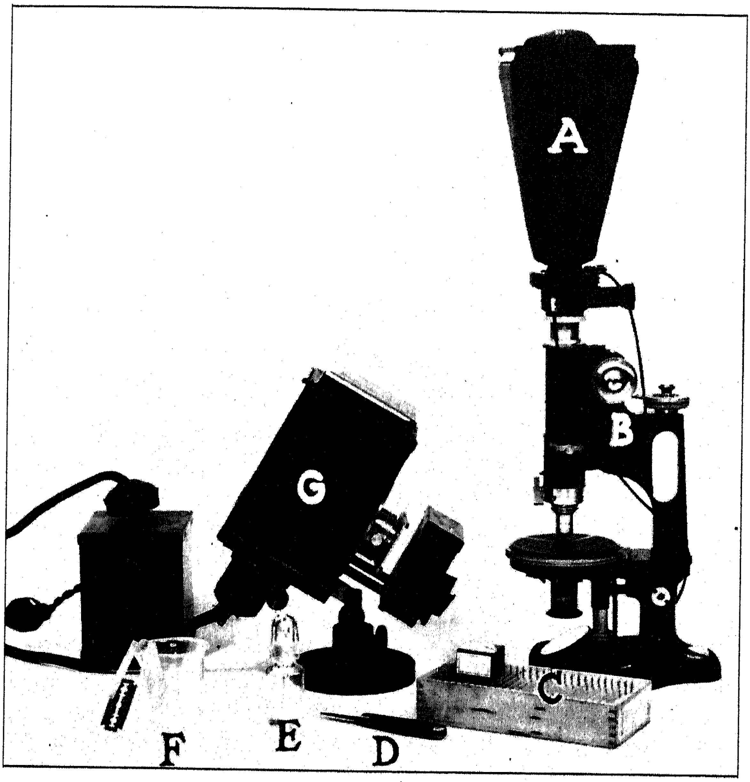


Fig. 3.—Laboratory equipment used in studying lactose crystallization and development in ice cream.

A. Microscope camera
B. Polarizing microscope
C. Slides and cover slips
D. Knife
E. Immersion oil
F. Razor blade knife
G. Microscope lamp

Thin Section Technique.—When it was desirable to examine ice creams in their natural state in the hardening room without alteration of changes in any way, the ice creams were prepared by the method of Keller (1934) and Reid and Hales (1934). The ice creams were hardened at -10 to -12° F. (-23.3 to -24.5° C.) and examined at the latter temperature. All equipment used was pre-cooled to this temperature, including the petrographic microscope, lamp, cover slips, slides, camera and razor blade knife.

A thin section of approximately 10 microns in thickness was cut with the razor blade knife, immersed in an immersion oil of known refractive index and covered gently with a cover slip. Care was taken not to disturb or distribute the ice cream material in any way so as to avoid breaking or shattering the lactose crystals which, in the early stages of crystallization, are extremely fragile. The immersion oil must be selected with discretion as ice cream is soluble in many instances. Ethyl acetate of refractive index of 1.379 at 20° C. is an excellent immersion medium for ice cream.

The section of ice cream was then photomicrographed with a microscope camera using a magnification of approximately 100 X.

Melted Drop Technique.—To avoid the health hazard of working in such low temperature as are essential when using this section technique, a second method of studying lactose at room temperature was developed.

A portion of ice cream about the size of a grain of wheat was removed from the sample, placed on a slide, and a cover slip placed over it. The weight and warmth of the cover slip were sufficient to melt the ice cream, which was immediately examined under the microscope. Care was taken not to distribute the ice cream or rock the cover slip. Lactose crystals in the sample distributed themselves in the center or near the edge of the melted drop, leaving the remainder of the drop nearly free of lactose crystals except in the case of sandy ice cream, where it was distributed more widely.

Range of Samples of Ice Cream Examined and a Study of Factors Influencing Lactose Development

In using these two techniques and methods of preparing ice cream samples for microscopic study of the lactose development, several series of ice cream samples were prepared. Ice cream mixes containing 10.5 and 16.0 per cent serum solids were prepared, representing low and high lactose concentrations, respectively. A portion of each of these mixes was frozen in the direct expansion batch freezer and another portion in the Vogt continuous freezer. "Seeding" was performed as previously mentioned.

In addition to the serum solids content, age, type of freezer used and "seeding," other factors influencing the development of lactose in ice cream were studied, including aging of mix, type and size of container, location within a container, and the effect of the freezing and hardening processes upon the concentration of lactose and their influence on crystallization of lactose.

Lactose Crystals in Samples of Aged Sandy Ice Cream, Condensed Milk and Processed Cheese

Several samples of aged sandy ice cream were examined by the drop method for the presence and type of lactose crystals. The effect of careful distribution and rocking of the cover slip on the shattering of lactose was also observed.

A sample of 30 per cent condensed skim milk was made "sandy" by further condensing, and the type of lactose development noted.

"Sandy" material isolated from a sample of process cheese was received for identification after having been identified by two other laboratories and another invéstigator as calcium tartrate. It was examined under the microscope and identified optically.

Photomicrographs were taken of all sandy samples and in addition a macroscopic picture taken of the large sandy crystals removed from the process cheese.

EXPERIMENTAL

Ice creams were prepared to include a low and a high serum solids content. In addition several other factors were studied in conjunction with compositional changes. All compositions were repeated from eight to twelve times to obtain checks, and only where the results could be repeated in all trials were they recorded.

TABLE 1. COMPOSITION OF ICE CREAM MIXES STUDIED

Mix No.	Serum solids	Fat	Sugar	Gelatin	Total solids	Lactose	Lactose per 100 gm. water
	%	%	%	%	%	%	gm.
1	10.5	12.0	15.0	0.30	37.8	5.7	9.1
2	16.0	12.0	15.0	0.30	43.3	8.7	15.3
3	16.0	"Seeded" by adding 2.5 lbs. dry milk solids to 5 gal. of 10.5 per cent serum solids ice cream mix. (Mix No. 1)					

Table 1 shows the composition of the mixes studied. It will be noted that only the serum solids content was varied and the percentages of butterfat, sugar and gelatin were held constant. A factor of 54.5 per cent of the serum solids as lactose was used to calculate a lactose content of 5.7 per cent in an ice cream mix containing 10.5 per cent serum solids, and a lactose content of 8.7 per cent in an ice cream mix containing 16.0 per cent serum solids. Ice cream mix containing 10.5 per cent serum solids, a lactose content of 5.7 per cent, and a total solids content of 37.8 per cent has 9.1 gm. of lactose per 100 gm. of water. Similarly, an ice cream mix containing 16.0 per cent serum solids and 43.3 per cent total solids has 15.3 gm. of lactose per 100 gm. of water. The figures were obtained by the application of the equation.

$$\frac{100}{\text{Percentage water in mix}} \text{ X per cent lactose.}$$

The percentage of water in a mix was determined by subtracting the percentage of total solids from 100.

Microscopic Examination of Materials Entering the Ice Cream Mix for the Presence of Lactose Crystals

A petrographic or polarizing microscope, utilizing both ordinary and polarized light, was used for all microscopic work.

The fluid portions of the ice cream mixes, including cream, skim milk and whole milk, were examined for the presence of lactose crystals. All portions were observed to be free of lactose crystals.

Spray processed skim milk powder was examined for the presence of crystalline material and lactose in particular.

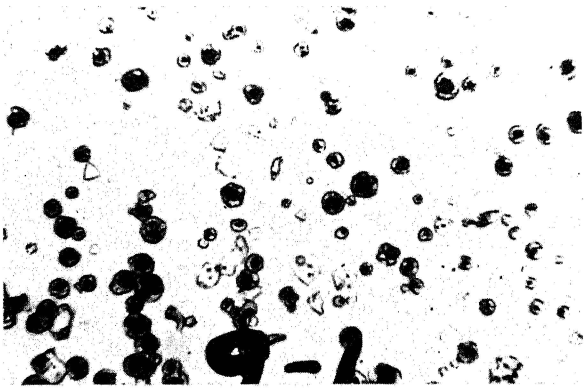


Fig. 4.—Photomicrograph of dry skim milk powder (original magnification 100 X).

Fig. 4 shows a photomicrograph of skim milk powder using plain light. The microscopic characteristic of skim milk powder particles is a dark center with a clear border. The index of refraction of the clear border was determined in several samples and found to be quite constant at 1.542.

The dark center represents a trapped air bubble.

An examination of the skim milk powder with the use of polarized light and a magnification of 900 X was made and it was found that dry skim milk particles are almost completely void of crystalline material. Birefringent material, indicative of crystalline material, was slightly in evidence in the dark center but may have been due to bubble birefringence or strain. Microscopic evidence pointed to the conclusion that skim milk powder is amorphous or non-crystalline in nature. In order to have more conclusive evidence as to the physical state in which the lactose exists, an X-ray analysis was made.

X-ray Analysis of Dry Skim Milk Powder

Two samples of spray process skim milk powder were analyzed by X-ray. Crystal radiations of 0.71×10^{-8} cm. were used and recordings were made on a cylindrical camera of 12 cm. radius. An intensity frequency curve recording was made of the film negative and showed a typical amorphous pattern. The presence of crystalline material can be detected when 2 per cent or more of the material present is crystalline, and an X-ray analysis would show that at least 98 per cent of the dry skim milk powder is in the amorphous state.

Microscopic Analysis of Ice Cream for the Development of Lactose Crystallization at Time Intervals

Regular Process Ice Cream Frozen in a Batch Freezer and Containing 10.5 Per Cent Serum Solids.—Fig. 5 shows photomicrographs of melted drops illustrating the development of lactose crystallization at different time intervals after being placed in the hardening room at an average temperature range of -8 to -12° F. (-22.2 to -24.4°. C.).

These ice creams are not considered to be "sandy" and the complete process of lactose crystallization which takes place in ice creams containing 10.5 per cent serum solids may be summarized in the following manner: The first evidence of crystalline material, as shown by birefringent material, occur between 3 and 4 hours after being placed in the hardening room. The number and size of lactose crystals increases quite rapidly between 6 and 8 hours, and thereafter to 56 days there is a gradual decrease in the number of crystals and a slow process of recrystallization or molecular orientation and packing taking place.

These photomicrographs show the lactose present in a melted drop of a representative sample of ice cream containing 10.5 per cent serum solids.

Regular Process Ice Creams Frozen in a Batch Freezer and Containing 16.0 Per Cent Serum Solids.—Fig. 6 shows the development of lactose at intervals of time in melted drops of ice creams containing 16 per cent serum solids. The time intervals of examination are the same as those of the ice creams containing 10.5 per cent serum

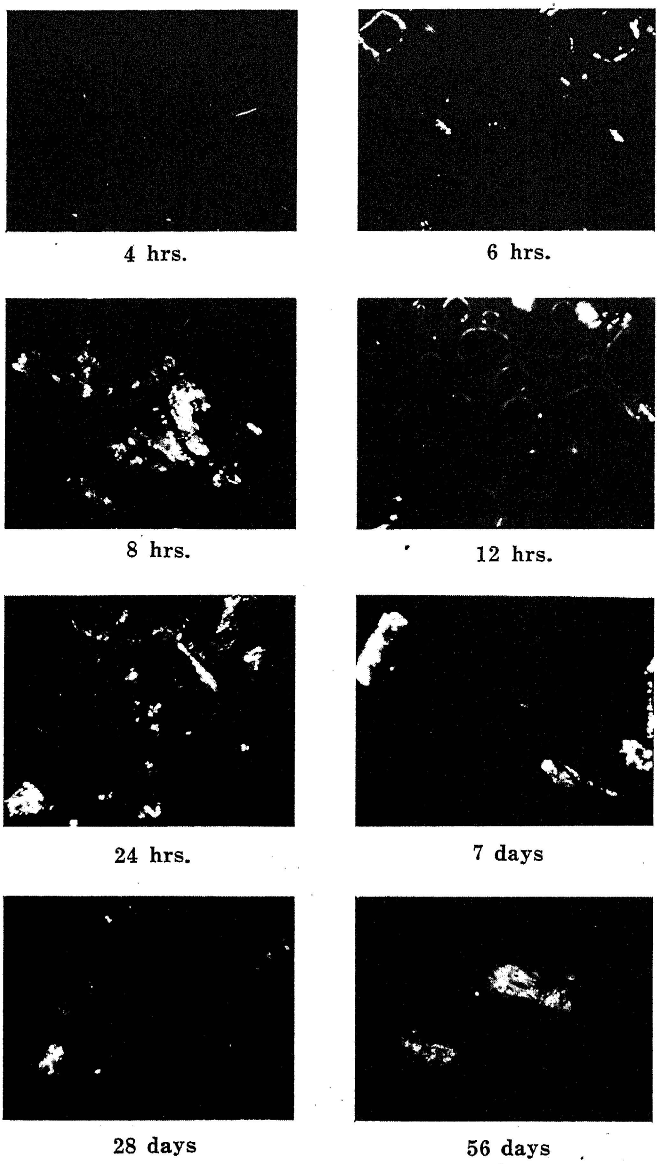
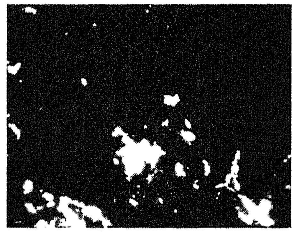
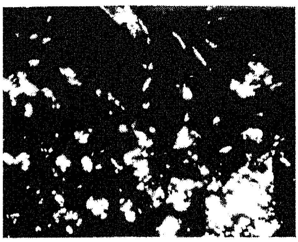


Fig. 5.—Photomicrographs of melted drops of ice cream containing 10.5 per cent serum solids and showing lactose crystallization in polarized light at time intervals (original magnification 100 X).

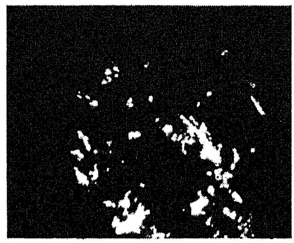
4 hrs.



6 hrs.

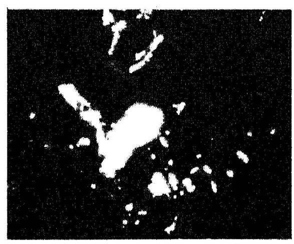


8 hrs.



12 hrs.

24 hrs.



7 days

28 days

56 days

Fig. 6.—Photomicrographs of melted drops of ice cream containing 16 per cent serum solids and showing lactose crystallization in polarized light at time intervals (original magnification 100 X).

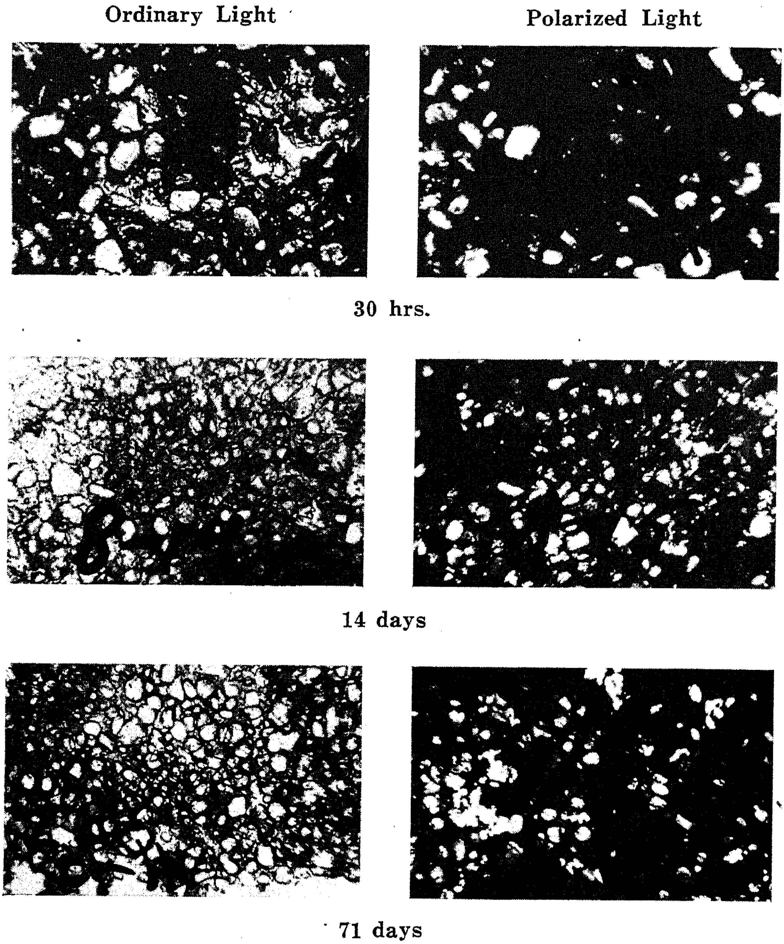


Fig. 7.—Photomicrographs of thin sections of ice cream containing 16 per cent serum solids and showing lactose crystallization in ordinary and polarized light at time intervals (original magnification 100 X).

solids and all processing and manufacturing treatments were identical for the ice creams.

In general the process of lactose crystallization in ice cream containing 16 per cent serum solids seems to proceed quite rapidly from 4 to 12 hours, then more slowly up to 24 hours, and thereafter up to 56 days there is a gradual decrease in the size of the lactose crystals and perhaps some decrease in number. These ice creams were not "sandy" during the period in which they were studied.

In comparing the lactose crystallization in ice creams containing 10.5 per cent and 16 per cent serum solids, respectively, they appear to be very similar. With the higher serum solids content, the lactose crystals are more abundant and persisted for a longer period of time, being still fairly abundant in number, although considerably decreased in size, at 28 and 56 days.

Fig. 7 shows the development of lactose crystallization in ice creams containing 16 per cent serum solids as illustrated by the application of the thin section technique in both ordinary and polarized light, at time intervals of 30 hours, 14 days and 71 days.

The photomicrographs were originally taken from Kodachrome slides where it was much easier to distinguish between the ice crystals, whose outline is more or less regular, and the lactose crystals, which are more irregular in shape.

Regular Process Ice Creams Frozen in a Continuous Freezer and Containing 10.5 and 16.0 Per Cent Serum Solids.—Fig. 8 shows the development of lactose crystallization of ice cream containing 10.5 per cent serum solids, frozen in a continuous freezer, and examined at time intervals.

After 7 days aging, two variations of lactose crystals are shown. In Field 1, the large crystals show that the process of recrystallization is taking place and the loosely knit form has changed to a closer knit, thicker crystal. In Field 2, a pin point type of crystal is shown, illustrating the finer type of crystal which is also present in ice cream frozen in the continuous freezer. However, the presence of the small crystals may be partially due to the technique used.

Fig. 9 illustrates the development of lactose crystallization in ice cream containing 16 per cent serum solids, frozen in a continuous freezer, and examined in polarized light at time intervals.

After 7 days the large, loosely bound crystals appear to have been replaced by a much greater number of smaller crystals as illustrated in Fields 1 and 2. The process of recrystallization is very evident and the crystals, although smaller, have become thicker.

At 14 days the appearance is much the same as at 7 days with but little difference in the size or quantity of lactose crystals. The ice cream gives the appearance of incipient sandiness and a further increase in the size, quantity and perhaps firmness of lactose crystals may cause the ice cream to be sandy.

In a comparison of ice creams of 10.5 and 16.0 per cent serum solids contents, there are both similarities and differences in the process of lactose crystallization. The ice creams containing 10.5 per cent serum solids took longer to develop lactose crystals than the ice cream containing 16.0 per cent serum solids and lactose was present in the latter in greater amounts especially at 7 days. The former has fewer crystals formed, but they are as large if not larger in size and thickness.

In a comparison of ice creams frozen in the batch and continuous freezers and containing 10.5 and 16.0 per cent serum solids, some

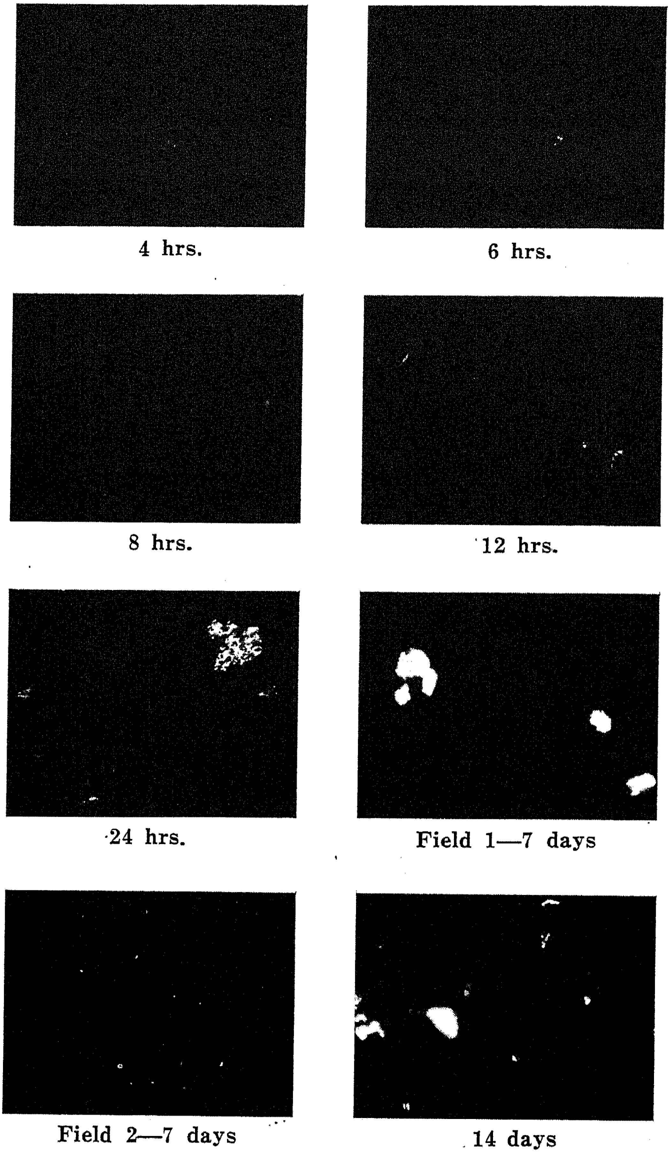
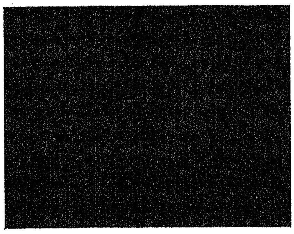
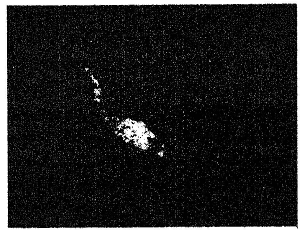


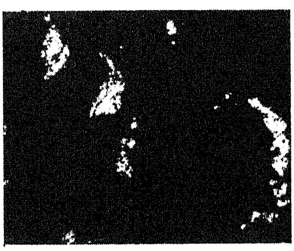
Fig. 8.—Photomicrographs of melted drops of ice cream containing 10.5 per cent serum solids, frozen in the continuous freezer and showing lactose crystallization in polarized light at time intervals (original magnification 100 X).

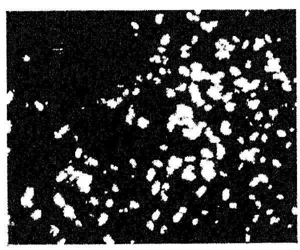

4 hrs.

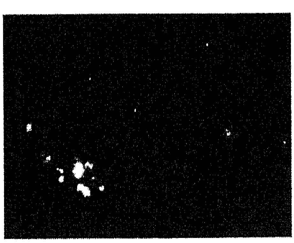

6 hrs.


8 hrs.


12 hrs.


24 hrs.


Field 1—7 days


Field 2—7 days

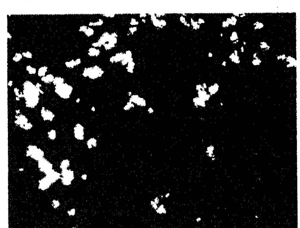

14 days

Fig. 9.—Photomicrographs of melted drops of ice cream containing 16 per cent serum solids, frozen in the continuous freezer, and showing lactose crystallization in polarized light at time intervals (original magnification 100 X).

interesting observations may be made. Ice creams frozen in the continuous freezer were definitely slower to develop lactose; whereas in most instances the ice creams from the batch freezer gave a greater abundance of lactose.

"Seeded" Ice Cream Containing 16.0 Per Cent Serum Solids and Frozen in a Batch Freezer.—Fig. 10 shows the development of lactose in ice creams seeded at the freezer by the addition of dry skim milk powder, and examined by the thin section method in ordinary and polarized light at 30 hours and 14 and 115 days. In ordinary light there are no lactose crystals visible, but in polarized light the lactose

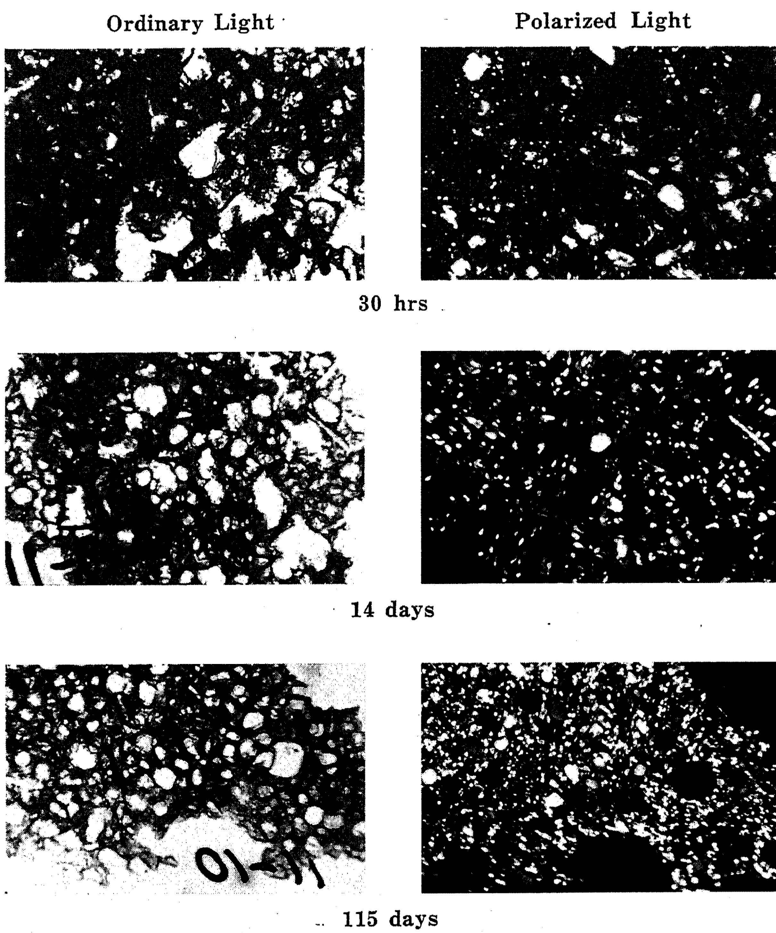


Fig. 10.—Photomicrographs of thin sections of "seeded" ice cream containing 16 per cent solids showing lactose crystallization in ordinary and polarized light at time intervals (original magnification 100 X).

crystals show up as pin points of light. During the period of aging of 30 hours to 14 days there is a slight increase in number and proportionately a greater increase in size of lactose crystals. At 115 days there is quite a large increase in the number but little, if any, increase in the size of crystals.

The process of "seeding" with dry skim milk powder produces crystal centers which initiate lactose crystallization in the form of a myriad of literally thousands of small crystals. These crystals vary in size from 1 to 2 microns in the smaller crystals first formed up to 5 to 7 microns in more fully developed crystals. It seems that so many lactose crystals form that none of them develop to a palpable size and produce true sandiness.

Fig. 11 illustrates the development of lactose in "seeded" ice creams examined in polarized light by the melted drop method at intervals

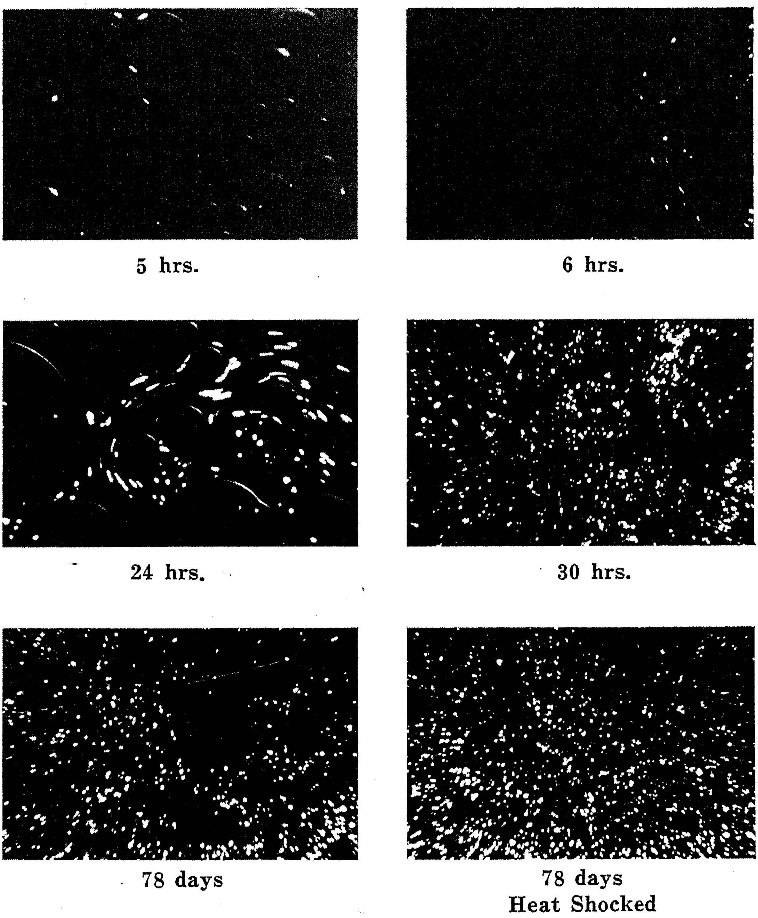


Fig. 11.—Photomicrographs of melted drops of "seeded" ice cream containing 16 per cent serum solids and showing lactose crystallization in polarized light at time intervals (original magnification 100 X).

of 5, 6, 24 and 30 hours and at 78 days, the latter before and after heat shocking at 12 to 16° F. (-11.1 to -8.9° C.) for 2 days and then rehardening.

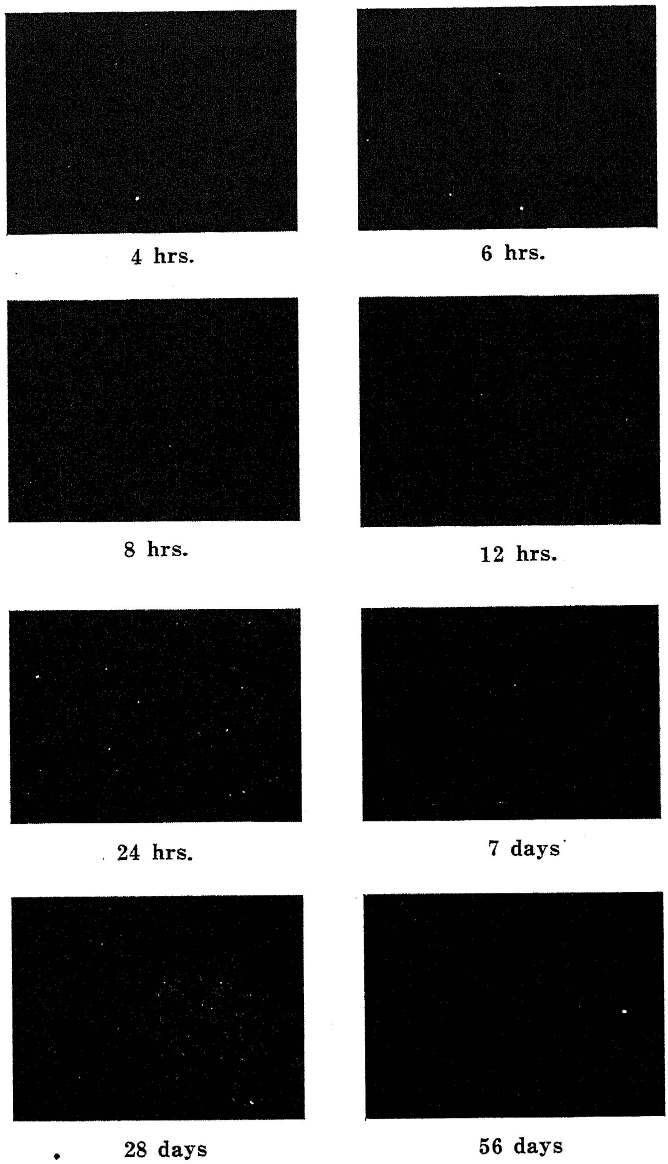


Fig. 12.—Photomicrographs of melted drops of "seeded" ice cream containing 16 per cent serum solids and showing lactose crystallization in polarized light at regular time intervals (original magnification 100 X).

The amount and size of lactose crystals increase slowly when the ice cream is aged from 5 to 24 hours. Some flow is present in the photomicrograph at 24 hours, leaving star tracks and large air bubbles visible in the background. The number of lactose crystals has increased quite markedly after aging from 24 to 30 hours. Thereafter up to 78 days there is practically no change in the type or number of lactose crystals. When the ice creams were aged for 78 days and then heat shocked the crystals increased in number without any apparent increase in size.

Fig. 12 shows the development of lactose crystallization in "seeded" ice cream at the same intervals of time as studied in the regular process ice cream. The first pin point lactose crystals appeared after 3 and 4 hours of aging and thereafter up to 24 hours there was a slow increase in the number of lactose crystals. From 24 hours to 56 days there was but little increase in the number of lactose crystals.

The lactose crystals were much finer than those found in previous trials with "seeded" ice cream and some difficulty was experienced in redissolving of the crystals.

Influence of Freezing and Hardening Processes Upon the Development of Lactose Crystallization

It has been shown in the previous section, "Microscopic Analysis of Ice Cream for the Development of Lactose Crystallization at Time Intervals," that lactose crystals appear in all ice creams after aging from 3 to 4 hours in the hardening room. This is true when the ice cream is made by the regular process or "seeded," when 10.5 or 16.0 per cent serum solids is used, or whether frozen in a batch or continuous freezer.

In order to determine why and how lactose crystal aggregates crystallize from ice cream during the hardening process, certain facts must be known and certain assumptions and deductions made from them. This includes range of temperature through which ice cream passes in the hardening process as it influences the amount of ice frozen, the lactose concentration and crystallization, the equilibrium ratio of beta to alpha lactose, the percentage change of beta to alpha lactose in 1 hour, and a theoretical calculation of the amount of lactose crystallizing from ice cream.

The Effect of the Amount of Ice Frozen During the Freezing and Hardening Process Upon the Concentration of Lactose as It Affects Lactose Crystallization.—If the amount of ice frozen in ice creams of definite composition is determined, as shown in Table 4, and the temperature of the ice cream when lactose crystals first appear is known, as given in Table 5, the concentration of lactose in parts per 100 parts of water can be calculated. These concentrations can be compared with the solubilities given in Table 3 and the degree of supersaturation estimated. Comparisons can also be made with the theoretical amounts of lactose crystallizing out of the ice cream as given in Table 2.

TABLE 2* AMOUNT OF LACTOSE CRYSTALLIZING OUT OF ICE CREAM

	-10° C.		-24° C.			
			When beta crystallizes		When alpha crystallizes	
Storage period	Of the insoluble lactose	Of the total lactose	Of the insoluble lactose	Of the total lactose	Of the insoluble lactose	Of the total lactose
hrs.	%	%	%	%	%	%
0	37	26	36	35	37	35
10	43	29	37	35	38	36
50	59	41	37	55	42	39
100	73	50	38	36	46	44
200	88	61	39	37	54	52
1000	100	68	48	46	86	82
	100	68	100	95	100	95
	In Percentage of Ice Cream Weight					
0	1.5		2.0		2.0	
10	1.7		2.0		2.1	
50	2.4		2.0		2.3	
100	2.9		2.1		2.6	
200	3.5		2.1		3.0	
1000	3.9		2.7		4.8	
	3.9		5.5		5.5	

*From Table VI, p. 159, Rahn and Sharp, 1928.

Table 2 (Table VI, p. 159, Rahn and Sharp, 1928) shows the amount of lactose theoretically crystallizing out of ice cream at -10 and -24° C. at time intervals. They consider two possibilities—first, that only alpha lactose crystallizes out, and second, that both alpha and beta lactose crystallize out.

Table 3 (Tables I and II, pp. 149, 150, Rahn and Sharp, 1928) includes the solubilities and equilibrium ratios taken from Table I and the percentage of beta changing to alpha lactose in 1 hour from Table II at temperatures of 0, -10, -20 and -30° C. The values in Table II are determined from the equation given by Hudson (1904),

$$\log k_2 = 12{,}016 - \frac{3800}{T}$$

where k_2 is the reaction constant for the change of beta to alpha and T is the absolute temperature.

Table 4 (Tables 2 and 3, Cole, 1938) shows the percentage of water frozen and the percentage of ice in three samples of ice cream mix

TABLE 3* SOLUBILITY OF LACTOSE AT DIFFERENT TEMPERATURES

Temperature °C.	Beta to alpha (Gillis) Ratio	Beta to alpha Per cent changed in 1 hour	(Hudson) Solubility in per cent Total	A-form	B-form	Solubility in gm. per 100 gm. water Total	A-form	B-form
-30	1.73	0.055	5.62	2.06	3.56	5.95	2.18	3.77
-20	1.71	0.230	6.95	2.56	4.39	7.47	2.76	4.71
-10	1.68	0.850	8.60	3.20	5.40	9.41	3.51	5.90
0	1.65	2.800	10.60	4.00	6.60	11.90	4.49	7.41

*Given as Table I, p. 149, Rahn and Sharp (1928) with the Per cent change beta to alpha lactose in 1 hour from Table II, p. 150.

TABLE 4*. RELATION OF TEMPERATURE TO THE AMOUNT OF ICE FROZEN IN THREE SAMPLES OF ICE CREAM MIX CONTAINING APPROXIMATELY 12 PER CENT FAT AND 38 PER CENT TOTAL SOLIDS

Temperature °C.	Percentage Water Frozen M-2	M-7	M-8	Average	Percentage Ice in Sample M-2	M-7	M-8	Average
-3.0	13.1	13.1	18.25	14.75	8.1	8.1	11.30	9.2
-3.5	24.5	23.1	28.25	25.30	15.2	14.3	17.50	15.7
-4.0	33.2	31.4	35.90	33.50	20.6	19.4	22.25	20.8
-5.0	45.2	45.2	47.80	46.10	28.0	28.0	29.65	28.6
-6.0	61.4	61.0	55.40	59.30	38.0	37.7	34.45	36.7
-7.5	70.8	69.8	63.40	68.00	43.8	43.2	39.30	42.1
-10.0	76.3	75.4	71.95	74.60	47.3	46.7	44.65	46.2
-12.5	80.0	79.0	77.35	78.80	49.5	48.9	48.00	48.8
-15.0	85.0	84.3	81.05	83.50	52.7	52.2	50.25	51.7
-17.5	--	--	84.05	--	--	--	52.15	--
-20.0	85.0	84.3	86.40	85.20	52.7	52.2	53.55	52.8
-25.0	88.5	87.8	89.90	88.70	54.8	54.3	55.80	55.0

*From Tables 2 and 3, Cole (1938), p. 149, with changes made of averaging two trials of M-8 and the addition of column of averages of the three samples.

containing approximately 12 per cent fat and 38 per cent total solids. The determinations were made by using the dilatometer and the original freezing point for the three ice cream mix samples, M-2, M-7 and M-8, which were -2.58, -2.55 and -2.47° C., respectively. It will be noted that in Table 4 several changes have been made from the original Tables 2 and 3 of Cole (1938) in that date for sample M-8 which covered two trials, have been averaged, and also that two columns of averages for the three samples have been added.

Table 5 gives the history of the freezing and hardening processes of ice cream containing 10.5 per cent serum solids and "seeded" at the freezer during the freezing process so as to increase the serum

TABLE 5. EFFECT OF AGE, SERUM SOLIDS CONTENT, PROCESS AND TYPE OF CONTAINER UPON THE DEVELOPMENT OF LACTOSE DURING THE HARDENING PROCESS*

			"Seeded" Ice Cream					
			Pan		Pint		Gallon	
Age	Hardening room temperature		Temperature	Lactose concentration and crystallization	Temperature	Lactose concentration and crystallization	Temperature	Lactose concentration and crystallization
Hrs.	°F	°C.			°C.		°C.	
0	-5.0	-20.6	-5.0	None	-5.0	None	-5.0	None
1.0	-6.0	-21.1	-6.0	None	-5.3	None	-5.3	None
2.0	-5.0	-20.6	-8.5	None	-7.5	None	-5.5	None
2.5	-5.0	-20.6	-11.5	--	-7.5	None	-5.5	None
3.0	-7.0	-21.7	-14.5	43.5 parts-100 parts water; few fine crystals	-9.3	29 parts-100 parts water; very few fine crystals	-6.3	21.75 parts-100 parts water; very few fine crystals
4.0	-5.0	-20.6	-19.0	Several fine crystals	-12.0	Several fine crystals	-6.3	Most abundant of all containers
6.0	-5.5	-21.0	-21.0	Slight increase	-15.0	Slight increase	-6.6	Very abundant
24.0	--	--	-15.5	Many medium crystals; fine crystals redissolve easily	-15.0	Same as pan except somewhat coarser and larger	-13.0	Coarsest and most abundant; needles and tomahawks

*All ice creams frozen in a batch freezer.
Ice cream "seeded" by adding 2.5 lbs. dry skim milk powder per 5 gallons of mix at the freezer at a uniform rate during the freezing process to ice cream containing 10.5 per cent serum solids.

solids content to 16 per cent. Those factors studied include the effect of the hardening room temperature, method of freezing, age and temperature of ice cream, serum solids content, and type of container used as they influence the growth and development of lactose crystallization in ice cream during the hardening process.

After 3 hours of aging, lactose crystals were present in all containers, at which time the temperature of the ice cream in the pan was -14.5° C., the closed pint -9.3° C., and the gallon container -6.3° C. In the instance of the ice cream in the shallow pan, it is possible that lactose crystals may have developed as early as 2.5 hours after it was placed in the hardening room.

The lactose concentration may be determined by reference to Table 4, which shows the amount of water frozen at the temperatures when lactose crystallization first occurred. Knowing the amount of water frozen, the amount of unfrozen water can be determined by difference. Since crystallization of lactose first occurs after approximately 3 hours of aging, an average figure is used. It is assumed that 60 per cent of the water is frozen in the gallon container of ice cream at -6.3° C., 70 per cent in the pint at -9.3° C. and 80 per cent in the pan sample at -14.5° C. The percentages of 60, 70 and 80 frozen water may be taken from 100, and the values of 40, 30 and 20 per cent unfrozen water obtained. Using the value of 8.7 per cent lactose in the mix and the above values in the equation

$$\frac{100}{\text{Per cent unfrozen water in mix}} \times 8.7,$$

the concentration of lactose, expressed in parts of lactose per 100 parts of water, is obtained. The result is 21.75 parts of lactose per 100 parts of water for 60 per cent of the water frozen in the gallon, 29 parts of lactose per 100 parts of water for 70 per cent of the water frozen in the closed pint and 43.5 parts of lactose per 100 parts of water for 80 per cent of the water frozen in the shallow pan.

Therefore, from these calculations it is apparent that the lactose concentration is in the labile area when lactose crystallization first occurs, being two, three and five times the saturation value in the three different containers.

Certain difficulties are encountered in arriving at the concentration of lactose. Prior to "seeding," the ice cream contained 10.5 per cent serum solids and 37.8 per cent total solids, and after "seeding" contained 16 per cent serum solids and about 43 per cent total solids. Although the dry skim milk powder was added at the freezer at a uniform rate after freezing of the ice cream mix had begun, it is difficult to determine the amount of powder that goes into solution and how much remains as "seeding" material. However, the concentration of lactose in the ice cream mix which was formerly 5.7 per cent was potentially 8.7 per cent after "seeding." The amount of dry skim milk powder going into solution lowers the freezing point of the ice cream and consequently less ice will be frozen at any given temperature.

To compare the rate of freezing and temperature of regular process ice creams during the hardening process with "seeded" ice creams, samples of ice creams processed in the regular manner containing 10.5 and 16.0 per cent serum solids were manufactured and the temperatures observed during the hardening process. These data are shown in Table 6. The three containers used for the ice creams containing 10.5 per cent serum solids were identical with those used for the "seeded" ice creams. Only pint samples of the ice creams containing 16 per cent serum solids were used and all ice creams were frozen in a batch freezer.

TABLE 6. COMPARATIVE TEMPERATURES DURING THE HARDENING OF REGULAR PROCESS ICE CREAMS CONTAINING 10.5 and 16.0 PER CENT SERUM SOLIDS

Age	Hardening room temperature		10.5% serum solids			16% serum solids
			Pan	Gallon	Pint	Pint
			Temperature	Temperature	Temperature	Temperature
hrs.	°F.	°C.	°C.	°C.	°C.	°C.
0	-2	-19	-4.2	-4.2	-4.2	-4.5
2	-6	-21	-9.5	-5.5	-8.0	-7.0
3	-4	-20	-13.5	-5.7	-11.5	-10.3

The temperatures of the pint samples of ice cream containing 16 per cent serum solids followed very closely those of the "seeded" ice cream. The temperature, during the hardening process, of the three containers of ice cream containing 10.5 per cent serum solids also followed quite closely the temperature of both the regular process ice cream containing 16 per cent serum solids and the "seeded" ice creams. The temperatures of the hardening room were slightly different during the two trials.

The location of section of the container from which the portion of ice cream to be examined was to be taken should also be considered, as the center of a container cools more slowly than the outside, thereby allowing greater opportunity for lactose crystallization. The influence of this factor is illustrated by the relative abundance of lactose crystals in the ice cream samples removed from the containers of different sizes. The differential rate of cooling between the center and the outer portions of the ice creams in the pint container is not sufficient to produce a significant difference in the type and abundance of lactose crystallization. However, in the case of the gallon container, a greater abundance of lactose crystallization is found in the center. It is to be expected that in a 5 gallon container, because of the increased volume of ice cream, that a greater difference in lactose crystallization would be observed.

It may be concluded that the rapidity with which the ice cream in these different containers was hardened and the temperature lowered was dependent upon the volume of ice cream and the amount of surface exposed. The rate of freezing and the amount of ice frozen also determined the lactose concentration (Table 5).

In the shallow pan the rate of freezing proceeded quite rapidly and the temperature of the ice cream reached equilibrium with the temperature of the hardening room in a period of 6 hours. The appearance of lactose crystals in the ice cream was questionable at 2.5 hours but definite after 3 hours of aging. The lactose crystals were slightly more abundant in the pan at this time but thereafter up to 6 hours developed very slowly until at 24 hours of age many medium crystals were present.

The appearance and development of lactose crystals in the pint sample of ice cream paralleled very closely that of the shallow pan. The rate of cooling was slower as the temperature of the ice cream had not reached equilibrium with the temperature of the hardening room after 6 hours of aging, but by 24 hours of aging equilibrium had been reached.

The rate of cooling and freezing was much slower in the gallon container. The development of lactose crystals was slower during the initial period, but after 4 hours aging was accelerated to a much greater degree than in the ice cream in the two other containers. Equilibrium temperature of the ice cream with that of the hardening room was not reached even after 24 hours of aging. The lactose crystals present in the ice cream after this period of aging were coarse and abundant with needles and tomahawks predominating.

These observations indicate that the ice cream in the shallow pan and the pint container show much the same type and number of lactose crystals. The pint sample loses its latent heat or hardens fairly rapidly but at a somewhat slower rate than the pan sample of ice cream, whereas the gallon container hardens much more slowly. If the lactose concentration is at the transition point or sufficiently advanced in the labile area to produce spontaneous crystallization in the ice cream in the gallon container, the opportunity of producing larger crystals of greater abundance exists because the temperature (-6.3° C.) is higher and the process of cooling is relatively slow, allowing time for crystallization to occur. In view of this fact, more rapid hardening would reduce lactose crystallization.

There is no method of determining the amount of lactose which actually crystallizes out of ice cream and that can be compared with the theoretical amount calculated in Table 2. A microscopic examination is qualitative and not quantitative. It is doubtful if lactose crystallizes out of ice cream to any great extent at -24° C. because of the excessive viscosity and the small amount of unfrozen water which is so highly concentrated as to prevent any molecular movement and crystallization. Some investigators believe it is so highly concentrated at low temperatures that the lactose exists in an amorphous (glass) state or at least in a highly supersaturated condition in the ice cream.

The Effect of the Temperature Upon the Linear Crystallization Velocity, Equilibrium Ratio and Rate of Change of Beta to Alpha Lactose and Viscosity as They Influence the Time and Rate of Lactose Crystallization in Ice Cream During the Freezing and Hardening Processes.—It is apparent that there are other factors of importance in addition to lactose concentration that influence the time when lactose crystallizes out of ice cream as well as the subsequent type and rate of lactose crystallization. This is demonstrated by the fact that lactose crystallized out of ice cream after being aged for 3 and 4 hours in the different containers, although the lactose concentrations varied from 21.75 to 43.5 parts of lactose per 100 parts of water or two to five times the saturation value at temperatures from -6.3 to -14.5° C. It is therefore evident that time is required for lactose crystallization to develop and that a concentration of 20 or more parts of lactose per 100 parts of water is sufficient to initiate crystallization if sufficient time has elapsed.

Temperature influences several factors involved in lactose crystallization in ice cream, such as the amount of ice frozen, lactose concentration in the remaining unfrozen water, viscosity, linear crystallization velocity, rate of change of beta to alpha lactose and the equilibrium ratio of the two forms.

The influence of the amount of ice frozen upon the lactose concentration in the three different containers and when lactose crystals first appear has been shown.

The influence of temperature upon the linear crystallization velocity has been summarized by Leighton and Peter (1923).

In Table 3, columns 2 and 3 show the equilibrium ratio and rate of change of beta to alpha lactose in 1 hour at temperatures of 0, -10, -20 and -30° C. At these temperatures the ratio is respectively 1.65, 1.68, 1.71 and 1.73. The percentage change in 1 hour is respectively 2.8, 0.85, 0.23 and 0.055. A decrease in the temperature results in an increase in the proportion of beta to alpha lactose in solution while the percentage change in 1 hour decreases more markedly as the temperature decreases.

The effect of these factors upon the initial and subsequent lactose crystallization is illustrated by comparing the samples of ice cream from the shallow pan and the pint with that from the gallon container (Table 5).

The Development of Lactose Crystallization to Sandiness

Fig. 13 shows photomicrographs of melted drops of ice cream containing 16 per cent serum solids illustrating, in polarized light, the development of lactose crystallization to a state of sandiness when examined at time intervals of 6, 24, and 48 hours and 7, 49 and 84 days.

After aging 6, 24, and 48 hours the fields shown exhibit only several crystals of lactose. After 7 days of aging, however, the amount of lactose has become quite abundant.

When the ice creams had been aged 49 days they were slightly sandy and at 84 days of age were mildly sandy. The reason for these samples of ice cream turning sandy is not known as they were subjected to the same treatment as ice creams in other lots which did not go sandy. However, on the whole, ice creams containing 16 per cent serum solids may be kept in the hardening room at -10° F. (-23.3° C.) for periods of time up to 56 days without becoming sandy. It is to be expected that at higher temperatures, for example, those existing in dealer's cabinets, sandiness would develop in ice cream containing 16 per cent serum solids in very much less time. Temperature is not the most important factor affecting lactose crystallization and in the control of sandiness in ice creams.

Development of Sandiness

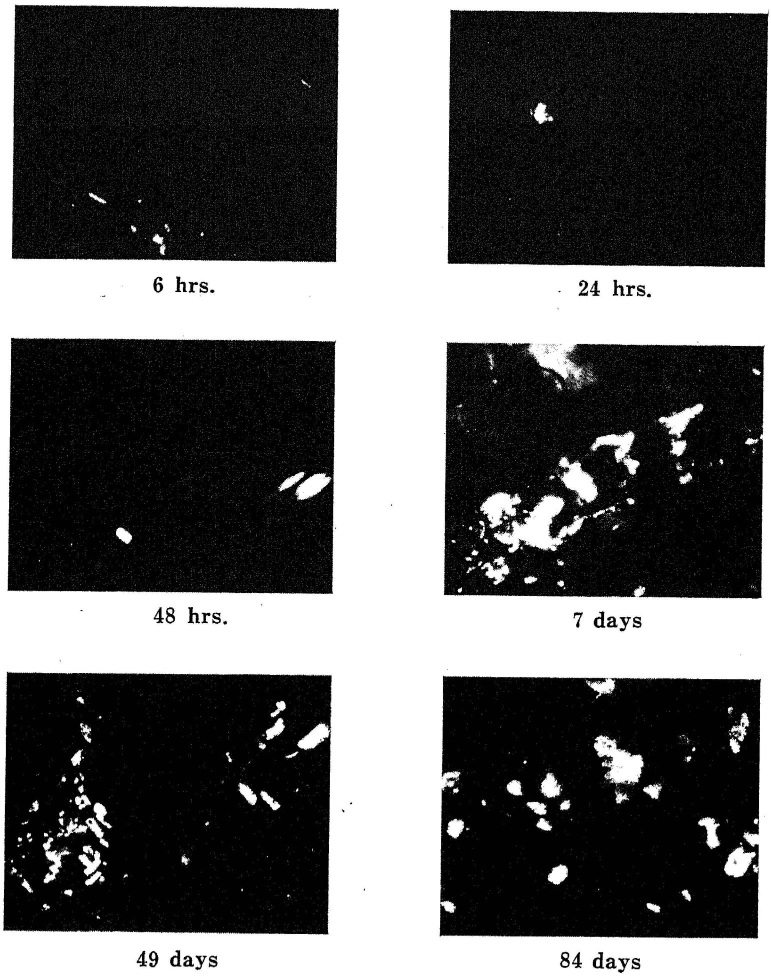


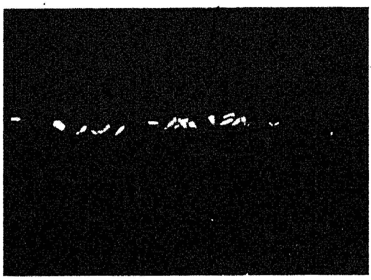
Fig. 13.—Photomicrographs of melted drops of ice cream containing 16 per cent serum solids showing the development of lactose in polarized light during the formation of sandiness (original magnification 100 X).

Laboratory Aging of Slide and Age of Ice Cream—Its Influence Upon the Development of Lactose Crystals

Fig. 14 shows photomicrographs, in polarized light, of ice creams containing 16 per cent serum solids and illustrates the effect of age of ice cream and age of laboratory slide upon the development of lactose crystals.

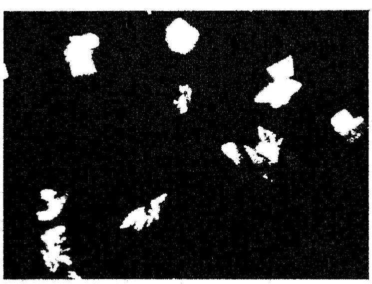
Age of Ice Cream

Laboratory Age of Slide



6 hrs.—Aged 1 hr.

6 hrs.—Aged 6 hrs.



8 hrs.—Aged 16 hrs.

12 hrs.—Aged 12 hrs.

Fig. 14.—Photomicrographs of melted drops of ice cream containing 16 per cent serum solids showing the effect of age and aging of prepared slide in laboratory upon the lactose development in polarized light (original magnification 100 X).

Slides were made of ice cream that had aged 6, 8 and 12 hours in the hardening room. These slides were then allowed to age in the laboratory at room temperature (26° C.) for intervals of time. The subsequent evaporation of the material on the slides increases the lactose concentration and permits crystallization.

A slide made of ice cream aged for 6 hours was held in the laboratory for 1 and 6 hours, respectively. After 1 hour needle-like crystals slightly tapering toward one end to give promise of a triangular shape were identified. After holding the slide for 6 hours in the laboratory, the crystals had grown and thickened to more rectangular shapes, although they still showed promise of having a triangular shape blunted at both ends.

A slide made of ice cream 8 hours of age and aged in the laboratory for 16 hours showed a very large growth in size and thickness of the crystals. Some crystals illustrate a feathery type of crystallization indicative of a rapid growth while others show a disposition to form blunt end triangles. The latter show a thickened center with a crust forming upon the sides.

A slide made from ice cream aged 12 hours and the slide subsequently aged 12 hours in the laboratory gave a splendid illustration of feathery lactose crystals which in time assume a triangular or tomahawk shape.

Lactose crystals which are initially small fail to grow very much. Crystals which have grown to some extent before being aged in the laboratory show primarily a feathery crystalline growth which if given sufficient time, 2 or 3 days, will assume a modified triangular shape with blunted ends.

Sandy Lactose Crystals in Aged Sandy Samples of Ice Cream

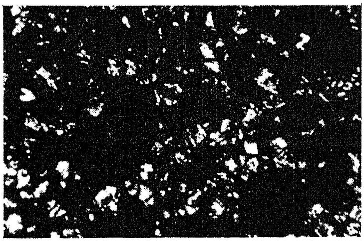
Fig. 15 shows sandy lactose crystals as they existed in several aged sandy samples of ice cream. The top set of photomicrographs taken in polarized light shows the type of sandy lactose crystals found in ice creams the acidity of which was standardized to 0.08 and 0.24 per cent, respectively. The cover slip of the latter sample was rocked to produce a finer type of crystal. The middle set of photomicrographs taken in polarized light shows a commercial sample, "A," before and after rocking the cover slip. Several large, well developed lactose crystals are shown in the sample, as well as the effect of rocking the cover slip, which caused them to disintegrate. However, it will be noted that these crystals are well developed and resist the breaking up action to a marked degree. Sample "B" of commercial ice cream in ordinary and polarized light illustrates how polarized light differentiates the birefringent "sandy" lactose material.

Lactose Crystals Observed in Skim Condensed Milk and Process Cheese

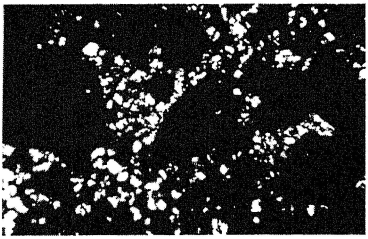
The crystallization of lactose as sandiness from skim condensed milk has not presented as serious a problem as in the instance of sweetened condensed milk.

Hunziker (1935) reported that lactose for some time has been isolated and identified chemically and microscopically as the cause of sandiness in sweetened condensed milk. He stated that the lactose crystals should be less than 10 microns in length and that the size may be controlled by "seeding" at the rate of approximately 4 ounces of powdered milk sugar to a 1000 pound batch of sweetened condensed

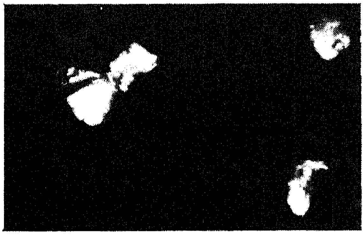
Aged Sandy Samples of Ice Cream



0.08% Acidity



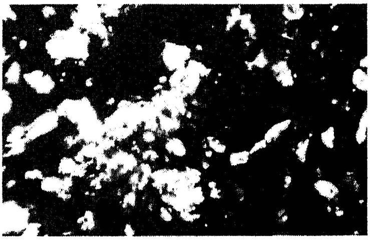
0.24% Acidity
Cover Slip Rocked



Commercial Sample "A"

Cover Slip Rocked

Commercial Sample "B"
Ordinary Light



Polarized Light

Fig. 15.—"Sandy" lactose crystals prepared from several aged sandy samples of ice cream (original magnification 100 X).

milk. This process produces about 300,000 crystals per cubic millimeter and limits the crystals to 10 microns in size.

Sato (1923) found, in addition to lactose crystals, other crystalline materials in sweetened condensed milk such as tricalcium tartrate, tyrosine, tricalcium phosphate and magnesium phosphate in an amorphous condition, and crystals of leucine and cystin.

The presence of crystalline materials in cheese has primarily been concerned with specks in cheese.

Lactose in Cheese and Condensed Milk

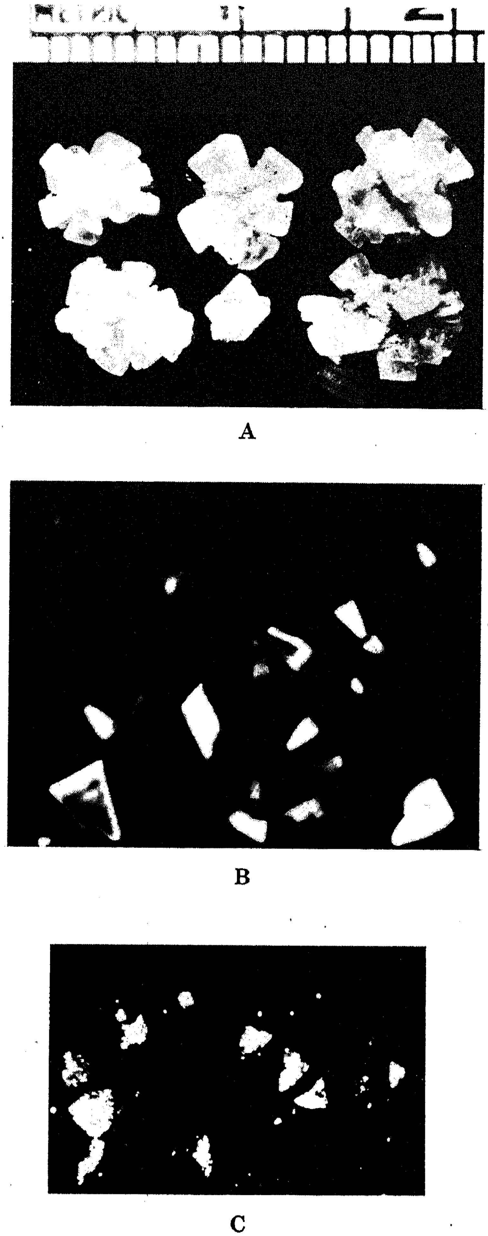


Fig. 16.—A Macroscopic picture of alpha lactose crystals removed from process cheese.

B. Photomicrographs of alpha lactose crystals removed from process cheese.

C. Photomicrographs of alpha lactose crystals in skim condensed milk.

Van Slyke and Publow (1911) detected the presence of white particles in cheddar cheese and concluded they were calcium soaps in which calcium was combined with some of the higher fatty acids.

Dox (1911) reported the presence of white particles in Roquefort cheese and identified them chemically and microscopically as largely tyrosine.

Dorn and Dahlberg (1942) also found tyrosine as white specks in ripening cheddar cheese and identified it by chemical and optical means.

Tuckey, Ruehe and Clark (1938), by X-ray analysis, showed white specks from well ripened cheddar cheese to be calcium lactate.

McDowall and McDowell (1939) also chemically identified white specks to be calcium lactate. However, true sandiness was found in process cheese and a sample of the sandy material was sent to this laboratory for identification after having been identified by two other laboratories and another investigator as calcium tartrate. It was identified optically as fully developed alpha lactose crystals.

Fig. 16 shows sandy material from process cheese and condensed milk. Part A is a macroscopic picture of the sandy material removed from process cheese. The clusters of fully developed tomahawk alpha lactose crystals are shown with the blunt, broad end protruding out and the narrower blade end extending toward the center of the cluster. The metric scale placed at the top of the pictures shows that the dimensions of the broad end of some of the crystals are greater than 1 x 2 mm. Part B represents a photomicrograph in polarized light of the same sandy lactose cheese crystals after being reduced to a finer state. The distinct tomahawk shapes are evident and a few saw tooth crystals are also shown.

Part C shows a photomicrograph in polarized light of sandy material present in skim condensed milk which was further condensed to produced sandiness. The crystals are quite large and triangular in shape, indicating the presence of alpha lactose crystals.

DISCUSSION

An attempt was made to show the manner in which lactose crystallizes out of ice cream during the hardening and storage period, as influenced by several factors, and then to associate certain causes with this phenomena.

The problem of studying the crystallization of lactose in ice cream raises several questions: What is the physical state of the lactose and other constituents present in the dry skim milk powder, and how does this affect their use as "seeding" material? The process of manufacture of skim milk powder shows that in the usual spray process method (Hunziker, 1935), preheated skim milk is atomized or sprayed out under high pressures (2500 to 3000 pounds) through the fine orifice of a spray nozzle into a drying chamber. A thin film is produced to form the walls of a fine bubble and a blast of hot air is admitted at a temperature of about 133° C. (266° F.), accomplishing the drying in a matter of seconds. Because of this rapid evaporation the thin film is dried in the form of an amorphous clear border with a dark center indicating the trapped air bubble. This amorphous clear border includes not only the lactose but all the other materials of the skim milk such as the proteins, minerals or ash and a small amount of butterfat.

Since the lactose is in an amorphous or glass state along with other materials in the clear border material, two theories may be advanced as to the manner in which the dry skim milk powder acts as "seeding" material.

The first theory is based upon the fact that when the dry skim milk powder is added at the freezer it is distributed throughout the ice cream and perhaps broken up, to a certain extent, while a portion goes into solution and a portion remains to act as "seeding" material. The portion going into solution undergoes a change from the amorphous to the crystalline state in the presence of the unfrozen water and crystallizes out on the undissolved "seeding" portion. Because the dry skim milk particles are much larger than the impalpable lactose crystals which crystallize out, this theory would require that the dry skim milk particles be broken up to a fine state in the freezer and a part of the particles be dissolved away and a part remain as a center.

If, however, all the amorphous lactose must go into solution before it can crystallize out, it may subsequently form what might be called "pockets" of supersaturation from which lactose may crystallize as impalpable lactose crystals in the ice cream after 3 to 4 hours of aging in the hardening room. This forms the basis of a second theory.

Two important factors must be considered in regard to this problem. First, a visual and taste examination of the "seeded" ice cream after leaving the freezer reveals that much of the skim milk powder has not gone into solution. However, some of the powder goes into solution during the first few hours in the hardening room. Second, lactose crystallizes out of all ice cream between 3 and 4 hours after being placed in the hardening room regardless of differences in serum solids content, type of freezer used, and whether it is "seeded" or regular process ice cream. This would seem to indicate that the change from the amorphous to the crystalline state requires about the same amount of time to crystallize out as lactose that is in true solution.

The original concentration of lactose in the ice cream mix is not important after the ice cream has started to freeze since the concentration is dependent only upon the amount of unfrozen water or syrup present. This thought was advanced by Herrington (1934) and in the light of the findings herein set forth, is true except that a mix with a higher serum solids content gave a greater abundance of lactose crystals during the hardening process.

The second question is: Why does lactose crystallize out of all ice creams studied after being placed in the hardening room for 3 and 4 hours irrespective of the variations in rate of freezing, type of freezer used, serum solids content, or whether it is "seeded" or frozen in the regular manner? There seems to be a time and temperature relationship involved which may affect crystallization. Time is required so that as the freezing and hardening process progresses and ice separates out, the lactose is continually concentrated until it reaches the boundary of the labile area and spontaneous crystallization occurs.

The classical work of Miers and Isaac (1906), although concerned with supersaturated salt solutions, suggested several contributions to the solution of this problem. They studied cooling solutions at rest, stirred and rapidly cooling solutions and solutions in sealed tubes both with and without friction from an added isomorphous particle.

The question which presents itself is: Which of these conditions exist during the freezing and hardening of ice creams? In the freezer three processes occur—agitation at a high rate of speed, freezing out of ice, and incorporation of air.

Assuming that 45 per cent of the water is frozen at the time the ice cream is drawn from the freezer at -5° C. (Cole, 1938), the lactose concentration in the unfrozen portion of the cream may then be determined. In a mix of low serum solids content (10.5 per cent), the lactose concentration very closely approaches saturation or the metastable area. In a high serum solids mix (16 per cent), the saturation value of lactose is greatly exceeded and may be considered to be definitely in the metastable area. Therefore, when ice creams are drawn from the freezer, the lactose concentration may be considered to be in or closely approaching the metastable area. Should it be true, as Miers and Isaac (1906) and Kucharenko (1925) postulate, that germ centers or crystal centers are present in the air, then conditions may be favorable for the formation of lactose crystals in ice cream when the labile area is reached. Thus it is probable that in a viscous ice cream little lactose crystallization will occur during the hardening process until the labile area is reached.

The process of lactose crystallization and the visible appearance of lactose crystals in ice cream after 3 or 4 hours of aging may be affected by several factors which can be summarized as follows: (1) The amount of the unfrozen water constantly decreases as the temperature is lowered and the ice freezes out, thereby concentrating the unfrozen portion; (2) the concentration of lactose passes through the metastable to the labile area as the ice freezes out and spontaneous crystallization occurs; (3) the rate of chemical reaction is slow at these low temperatures resulting from the reduced molecular activity; (4) the solubility of lactose decreases with the fall in temperature.

Following the initial appearance of lactose crystals in ice cream after 3 to 4 hours of aging in the hardening room, the lactose continues to crystallize out, reaching a maximum in some samples of ice cream at 8 hours of age and in other samples from 12 to 24 hours. During this period, temperature and concentration are favorable for lactose crystallization in ice cream. The amount of lactose crystallizing out during this period is dependent upon the linear crystallization velocity, the lactose concentration, the rate of change of beta to alpha, the amount of unfrozen water, and degree of concentration of the unfrozen syrup. These factors are dependent upon the temperature as it influences the physico-chemical properties of lactose and the amount of ice frozen out.

Following the period of maximum lactose crystallization in ice cream during the hardening process, there is a period when little or no lactose crystallization occurs and the large amount of ice that freezes out has concentrated the small amount of unfrozen water to such a high degree that lactose crystallization is prevented. This is a period of dormant lactose development in ice cream and takes place after a low temperature has been attained during the hardening process and continues during the storage period. There was a gradual decrease in the size and to a lesser extent in the number of lactose crystals in normal ice creams processed in the regular manner when held in the hardening room at -10° F. (-23.3° C.) for a period of time up to 56 days.

The exact temperature of the ice cream during the hardening process below which lactose crystallization is dormant is difficult to predict and would require a detailed study to be determined accurately. It would be affected by such factors as mix composition, type of freezer used, temperature of hardening room during storage period and the size of the ice cream container.

CONCLUSIONS

1. A petrographic or polarizing microscope is an excellent instrument for studying the development of lactose crystals in ice cream.

2. The melted drop of ice cream is the most convenient technique of preparing ice cream for microscopic examination of lactose development.

3. The microscopic characteristics of skim milk powder particles were a dark center representing a trapped air bubble and a clear amorphous border having a refractive index of 1.542.

4. The process of lactose crystallization in regular process normal ice creams of which the additional source of serum solids is made up with skim milk powder can be divided into three periods during the hardening and storage processes, i. e.:

A. The first period (the period during which initial lactose crystallization makes its appearance) requires between 3 and 4 hours after the ice cream is placed in the hardening room and represents the passage through the metastable area to the labile area of lactose concentration.

B. The second period (the period of favorable conditions of temperature and concentration for lactose crystallization) occurs after the initial appearance of lactose crystals, continues between 12 and 24 hours, and corresponds to the attainment of the labile area of lactose concentration. The lactose crystallizes out with more abundance during the first half of the period than during the latter half.

C. The third period (the period of dormant lactose crystal development) takes place after the ice cream has been in the hardening room between 12 and 24 hours, continued during storage at low temperatures, and with passage of time there is a gradual decrease in the number and, in some instances, the size of lactose crystals.

5. The concentration of lactose in the unfrozen portion and the temperature of the ice creams will vary during the hardening process with the size of the container.

6. The second period or zone favorable for lactose crystallization must be avoided if sandiness is to be prevented.

7. The actual amount of lactose which crystallizes out of ice cream under average hardening room conditions during the second or period favorable for lactose crystallization is small in comparison to the total amount of lactose present in solution.

8. The quicker freezing of the ice cream mix in the continuous freezer appeared to delay the development of lactose crystallization during the initial phase of the second period of favorable conditions for lactose development; however, lactose crystals developed quite abundantly in the later stages of the period.

9. The lactose in ice cream of which the additional source of serum solids is made up with skimmilk powder processed and frozen in the regular manner crystallizes out as rather large, loosely knit, thin crystals which are palpable in size, and very fragile. Its few numbers and fragility, however, prevent it from being palpable in the sense of being detectible in the mouth.

10. The lactose crystals in the "seeded" ice creams are of an impalpable form (undetectible in the mouth) and crystallize out in the form of an immense number of small crystals from 2 to 7 or 10 microns in size.

11. Lactose crystals were present in more abundance during the hardening and storage period in ice cream containing 16 per cent serum solids than in ice cream containing 10.5 per cent.

12. Ice cream, to be sandy, must fulfill three requirements: First, there must be a sufficient number of crystals present; second, they must have reached a size that is palpable or detectible in the mouth; and third, they must be of a sufficient hardness or firmness to resist the breaking up action in tasting.

13. Clusters of large sand crystals removed from process cheese to which the whey solids had been added back were identified by optical means as alpha lactose.

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