

## RESEARCH PAPERS

PRACTICAL REDUCTION OF ORTHOKINETIC FLOCCULATION  
IN PROCESSING OF STERILE MILK CONCENTRATES

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## ABSTRACT

Comparison between sediment deposition rates in milk concentrates sterilized statically and dynamically demonstrated convincingly that orthokinetic flocculation plays as important a part in the coagulation process as does thermal coagulation. Substitution of 2-, 3-, and 4-in. holding tubes for the 5/16-in. Malloy holding-coil furnished conditions for laminar flow and shear gradient variation in processing a large number of concentrates of varying composition. For storage periods up to four months, no measurable amount of sediment was observed in concentrates processed under conditions of reduced velocity gradients, whereas some sediment was usually observed in concentrates processed under conditions of relatively high gradients. A two- to fourfold decrease in sediment deposition was observed in concentrates stored for 8-12 months when conditions favoring low-velocity gradients in the holding period were substituted for those conducive to a high-velocity gradient. Substitution of evaporative cooling for forced convective cooling brought forth further marked decreases in the sediment deposition rate. At the end of 16 months of storage at 70F no sediment was observed in high-fat concentrates (fat to SNF ratio = 1:1.8) sterilized under conditions of reduced gradients.

Sediment deposition during the storage of 3:1 high-short sterilized milk concentrates is a serious problem, the solution of which requires a many-sided approach. Settling-out is a consequence of the destabilization of milk colloids during sterilization of the concentrates. Destabilization is masked in practice, since homogenization employed as a terminal step tends to crush large aggregates to an impalpable size, not small enough, however, to prevent settling-out during storage.

The steps employed to cope with the problem are those commonly used in evaporated milk technology—forewarming and the addition of stabilizing salts. These necessary steps, although exerting a profound effect, fall short of the desired goal.

The question arises: What additional steps may be taken? Coagulation theory suggests as

factors worthy of study—orthokinetic flocculation,<sup>1</sup> the fats-to-solids ratio, and evaporative cooling.

Experiments designed to study these factors will be described and the theory underlying the experimental program will be discussed.

## THEORETICAL PART

The orthokinetic flocculation rate is a sensitive function of the particle size and holding-tube diameter. Moreover, a critical particle size exists for each holding tube diameter above which orthokinetic flocculation, and below which thermal flocculation, dominates the total coagulation. The circumstances under which orthokinetic flocculation assumes importance, although restricted, are precisely those met when forewarmed, unhomogenized 3 to 1 concentrates are sterilized under high-short conditions in plant practice.

The pertinent mathematical relationships were developed by Smoluchowski (6) and validated by others (5,7,8) many years ago. It remains only to apply them to the problem at hand. The ratio  $N_2/N_1$  between the orthokinetically and the thermally induced collision rate in a suspension of  $N_0$  uniform spheres per

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<sup>1</sup>Orthokinetic from ortho, meaning straight, and kinetic, referring to motion, is used in the literature of colloid chemistry to refer to aggregation engendered by relative motion of particles in a straight line. Thus, a distinction is made between orthokinetic coagulation and perikinetic, the latter term referring to round-about or thermal motion.

cubic centimeter is given by the following equation:

$$\frac{N_2}{N_1} = \frac{32r^3GN_0}{\frac{3}{8kTN_0}} = \frac{4r^3G\eta}{kT} \quad [1]$$

The numerator in the term following the first equality sign is equal to the orthokinetic induced rate per particle  $N_2$  and the denominator is equal to the thermally induced rate per particle  $N_1$ .  $r$  is the particle radius,  $G$  is the velocity gradient,<sup>2</sup>  $\eta$  the viscosity,  $T$  the absolute temperature, and ( $k = 1.38 \times 10^{-16}$  ergs degree<sup>-1</sup>) is Boltzman's constant. Of particular interest is the strong dependence of the ratio on particle radius  $r$ . For any given value of  $G$ , the velocity gradient, a critical value of  $r$  exists such that  $N_2 = N_1$ . In a suspension the particles of which have a radius significantly smaller than the critical, the coagulation process is essentially a thermal one. Once the particles have grown to such size that the critical radius is exceeded, orthokinetic coagulation soon becomes the dominant factor.

The equation requires that the velocity gradient should be known or calculable. In the processing of high-short sterile concentrates, it is the practice to pass the concentrate at sterilization temperature and at high velocities through small-bore holding coils. The mean velocity gradient under these circumstances is hardly calculable; hence, it becomes necessary to idealize the situation. The tube is assumed to be straight, the fluid, Newtonian, and the flow laminar. The mean velocity gradient then is given by the following expression, thus:

$$\bar{G} = \frac{16Q}{\pi d^3} \quad [2]$$

$d$  is the tube diameter, and  $Q$  is the volumetric flow rate. Substituting for  $G$  in Equation [1], one obtains the following equation, thus:

$$\frac{N_2}{N_1} = \frac{64r^3\eta Q}{\pi d^3 kT} \quad [3]$$

If for  $T$ , the sterilizing temperature, 410 K is

<sup>2</sup>The velocity gradient should not be confused with the velocity. It denotes the difference in velocity divided by the distance between adjacent layers in a moving fluid. Because of the velocity difference the particles in one layer under suitable circumstances can collide with the particles in the adjacent layer. The ratio  $N_2/N_1$  in Equation [1] would be unaffected by the introduction in both the numerator and denominator of a probability factor denoting the chance of a successful collision, i.e., one leading to aggregation.

chosen, and if  $\eta$  and  $Q$  are assigned the values of 0.02 poise and 100 cc per second, respectively, Equation [3] reduces to the following:

$$\frac{N_2}{N_1} = 7.2 \times 10^{14} \left(\frac{r}{d}\right)^3 \quad [4]$$

Thus, the ratio is not only a sensitive function of  $r$ , the particle radius, but also of  $d$ , the tube diameter. Setting  $\frac{N_2}{N_1} = 1$ , one may calculate

the tube diameter,  $d$ , corresponding to any given value of the critical particle radius  $r$  from the following equation:

$$d(\text{critical}) = 9 \times 10^4 r(\text{critical}) \quad [5]$$

*Ratio between encounter rates in skimmilk.* Since Equation [5] is derived for model suspensions, and since milk concentrates do not conform exactly, an idealized situation will once again be assumed to exist, one in which the protein particles number  $3 \times 10^{13}$  per milliliter each 100  $\mu$  in diameter. Skimmilk would be expected to contain approximately this number of particles. Application of Equation [5] yields the interesting result that when  $r(\text{critical}) \leq 5 \times 10^{-5}$  cm,  $d(\text{critical}) < 0.8$  cm. Hence, if the specified milk is passed through a holding tube 0.8 cm in diameter, the particles resisting thermal aggregation will also resist orthokinetic flocculation. However, milk concentrates are not completely heat-stable, and the micelles therein may be expected to grow thermally. If  $r$  increases beyond  $10^{-5}$  cm, Equation [5] shows that  $d$  must exceed 0.8 cm if orthokinetic flocculation is to be avoided; hence, under the stipulated conditions, orthokinetic flocculation may be expected to assume a dominant role once the particle radius exceeds  $10^{-5}$  cm. Under its impetus, the particles will agglomerate autocatalytically to the point at which they will settle out rapidly. To subordinate orthokinetic flocculation during sterilization, one may increase the diameter of the holding tube. Equation [5] provides a rationale for holding-tube design if the reasonable assumption is made that the caseinate complex particle radius in 3 to 1 concentrate (stabilized by means of forewarming and additive) will not increase in size because of thermal effects alone beyond  $2.5 \times 10^{-5}$  cm.<sup>3</sup> Under these conditions, the critical holding-tube diameter is equal to 2.3 cm. Allowing a factor of safety of 100%, one obtains approximately a value of 5 cm, or 2 in., for

<sup>3</sup>If settling follows Stokes law, particles  $2.5 \times 10^{-5}$  cm in radius should settle out at 20 C at a rate roughly of 1 cm per 100 days.

the tube diameter. Employment of tubes of this diameter or larger should effectively rule out the danger of orthokinetic flocculation, provided that the diameter of a large majority of protein particles through thermal effects alone does not increase during the holding period much beyond 500  $\mu$  (see Appendix).

*Encounter rates in suspensions of fat and proteins.* It will now be shown that with respect to collisions involving protein particles, the fat globules in unhomogenized stabilized milk concentrates may be expected to behave as an inert phase. To take into account the two size groups, the equation for the thermal collision rate per cubic centimeter is modified, thus:

$$N_{(\text{thermal})} = \frac{2kT(R+r)^2 N_r N_r}{3rR\eta} \quad [6]$$

Orthokinetic flocculation, it is assumed, has been controlled.  $R$  and  $r$  are the radii, respectively, of the fat and protein particles, and  $N_r$  and  $N_r$ , their numbers per milliliter.

Equation [6] may now be applied to an idealized suspension of average-sized protein micelles and fat globules containing initially per milliliter  $3 \times 10^{13}$  micelles,  $5 \times 10^{-6}$  cm in radius and  $8 \times 10^9$  globules,  $1.5 \times 10^{-4}$  cm in radius.

The initial collision rate per cubic centimeter with respect to the micelles in the absence and presence of fat, respectively, is  $3,500 \times 10^{12}$  and  $3,515 \times 10^{12}$ . Thus, initially the protein system behaves essentially as if the fat were not present. Moreover, since more than 20,000 micelles would be required to associate with each fat globule to overcome buoyancy, a sedimenting fat-protein agglomerate can hardly be realized in the early stages of coagulation. In this stage the protein micelles develop in size essentially independent of the fat. As the micelles grow, and the radius of the micelles approaches that of the globules, conditions become more favorable for formation of mixed sedimenting agglomerates.

If orthokinetic flocculation can be controlled within tolerable limits, the radius of the micelles will not reach this large value, but rather an intermediate one, such that the number of encounters in which protein participates will remain largely unaffected by the fat. In mixed aggregates, the fat can be expected to hinder settling, and the protein, creaming.

Superior homogenization may be expected to bring the number and size range of the fat globules much closer to those of the protein particles. Hence, homogenization can have disastrous consequences on heat stability unless the fat globules are properly surfaced to re-

sist attachment following encounters with protein.

Calculations based on theory lead one to expect a significant reduction in the initial thermal protein-protein encounter rate without compensating increases in the protein-fat collision rate, if in unhomogenized forewarmed 3 to 1 milk concentrates the ratio between fat and SNF is increased. A further reduction is to be expected if evaporative is substituted for forced convective cooling.

#### EXPERIMENTAL PROCEDURE

The pilot plant arrangement for producing high-temperature-short-time sterile milk concentrates was essentially the same as described in a previous publication (2). To substitute for the 5/16-in. Mallory holding coil, three holding tubes were constructed out of 2-, 3-, and 4-in. stainless steel standard pipe. The respective inside diameters of these were 1.94, 2.90, and 3.83 in., and the lengths were adjusted so that the capacity of each was 1,630 cc after allowances were made for baffle plates 1/16-in. thick and 1/2-in. smaller in diameter than the holding pipe to which it was adapted. Flanged bolted ends were used together with asbestos sheet packing to secure against leakage, and the baffle plates were secured to the bottom flanges by means of machine screws passing through three 1/2-in. long spacers. The holders were mounted in a vertical position on a suitable stand. Fluid passing through each of the tubes following deflection by the baffles moved without the slightest sign of turbulent mixing or channeling. To connect the holding tubes to the bringing-up coil at one end and the cooling coil at the other, 129 in. of 5/16-in. tubing were required. Corresponding to a volumetric flow rate of 107 cc per second, 15.2 and 1.6 sec., respectively, were required to fill the holding and connecting tubes.

In one experiment, in which it was desired to cool by evaporation rather than by forced convection, a small Rogers vacuum pan designed for carrying out evaporation aseptically was already available, equipped with an automatic vacuum pressure controller.

Sterilization in situ on a micro-scale was carried out as previously described (3). To measure sediment in reconstituted milks derived from milk concentrates sterilized in situ, the sample, approximately 0.01 ml, was forced out of its capillary on to a water-repellant surface, then drawn into a thin-walled capillary tube, where it was mixed with the required amount of water to yield a reconstructed milk. To effect dilution and mixing following the

introduction of sample and prior to the introduction of water, an air cell was introduced into the capillary tube to act as a barrier against premature mixing of the concentrate and water. Centrifuging was carried out in a clinical centrifuge operating at 2,900 rpm for 10 min. The settling-out index was taken as the ratio between the length of the sediment thread and that of the sample thread.

Settling out in larger samples (15 ml) of reconstituted milks was determined in lusteroid centrifuge tubes. The supernatant suspension remaining after centrifuging was decanted carefully, the adhering material on the wall of the tube washed away with a stream of water, the tube with its mouth down allowed to drain 2 min, after which milk residues adhering to the walls to within approximately 1/16-in. of the sediment deposit were removed with the aid of a snug-fitting test-tube brush. The walls of the tube were dried with towelling, and the weight of deposit determined by difference. Fat in the deposit was determined on suitable aliquots by means of the Babcock method carried out with skim milk test bottles, and total solids in the sediment was determined by the Mojonnier method (4). Nitrogen was determined by the Kjeldahl method on samples of the original milk and supernatant, and these data combined with the other analytical data permitted calculation of the quantity of protein in the sediment.

Viscosity measurements were carried out with

a transpiration-type viscometer in the manner described previously (3).

#### EXPERIMENTAL RESULTS

Experimental data contained in succeeding tables support in a qualitative sense theoretical conclusion.

*Sterilization in situ compared with sterilization in the plant.* The data in Table 1 refer to an experiment in which sterilization of forewarmed 3 to 1 concentrates in one instance was carried out statically in thin-walled capillary tubes and in another under the influence of high-velocity gradients such as those encountered in plant practice. Sample 1, containing polyphosphate and sterilized in situ at 137.8 C for 30 sec, experienced no significant viscosity change, and in a sediment deposition test showed no well-defined evidence of phase separation. Sample 2, also sterilized in situ but containing no additive, increased tremendously in viscosity (manifestly an indication of structure formation) on sterilization, yet the reconstituted milk prepared from it showed no clear-cut signs of creaming or settling-out. Sample 3 sterilized in the plant for only 17 secs with the holding coil 5/16-in. in diameter in the line suffered serious damage, reflected not so much by the increase in viscosity as by the tendency of a substantial part of the protein to settle out. Sample 4 was also sterilized in the plant; but now with a 3-in. diameter holding in the line in place of 5/16-inch

TABLE 1  
High-short sterilization in situ compared with sterilization under dynamic conditions showing influence of velocity gradients during sterilization on sediment deposition in sedimentation tests

Sample	Viscosity before sterilization (centipoises)	Viscosity after sterilization (centipoises)	Sediment <sup>a</sup> %	Cream layer %	Remarks
1	25.8	26.4	See remarks	See remarks	No sharp division. Diffuse cream, merges with diffuse protein layer.
2	12.9	665.0	See remarks	See remarks	No sharp division. Diffuse cream, and protein layers merge.
3	25.8	39.6	15.5	2.8	Sharp lines of demarcation between cream, serum, and sediment layers.
4	25.8	26.8	See remarks	See remarks	No sharp division. Diffuse cream, and protein layers merge.

Samples 1-4 contain 36% solids; concentrates were prepared from milk forewarmed at 137.8 C for 16.8 sec. Samples 1, 3, and 4 contain 0.55 lb hexametaphosphate per 100 lb milk solids. Samples 1 and 2 were sterilized in thin-walled capillary tubes at 137.8 C for 30 sec. Sample 3 was sterilized in Mallory at 137.8 C for 16.8 sec. Sample 4 was sterilized in modified Mallory containing 3-in. holding tube in place of 5/16-in. coil.

<sup>a</sup> Sediment in reconstituted milk. Milk in thin-walled capillary tube centrifuged 10 min at 2,900 rpm in clinical centrifuge.

TABLE 2

Results of sediment deposition tests on 37% high-short sterile concentrates differing with respect to the velocity gradient during sterilization

No.	Sample	Wet sediment per 100 ml milk <sup>a</sup>	Solids in sediment	Solids-not-fat in sediment		Homogenization index <sup>b</sup>
				(g)	(%)	
1	Sterilized unhomogenized concentrate held in 5/16 in. coil 16.8 sec during sterilization.	15.0	4.7	1.5	3.2	
2	Sterilized unhomogenized concentrate held in 3 in. tube 15.3 sec and in 5/16 in. coil 1.5 sec	2.0	0.4	0.05	0.35	
3	Same as (1) but homogenized	3.0	0.8	0.10	0.7	85
4	Same as (2) but homogenized	1.1	0.3	0.03	0.27	88.3

Forewarming temp and time — 129.4 C for 16.8 sec.

Sterilization temp and time — 137.8 C for 16.8 sec.

Homogenization temp and pressure — 71.1 C, 7,500 psi first, and 500 psi second stage.

<sup>a</sup> Concentrates reconstituted to 12.5% solids; 15 ml centrifuged at 2,900 rpm for 10 min in clinical centrifuge. Sediment weighed and analyzed.

<sup>b</sup> Per cent transmission at 1,020 m $\mu$  of milk diluted with 0.1 N NH<sub>3</sub> to contain 0.014% fat (1). Measurement with Model B, Beckman spectrophotometer, 1-cm cell.

coil, the sterilized product with respect to viscosity and settling-out characteristics resembled the concentrates sterilized in situ.

*Effect of substituting 3-in. holding tube for 5/16-in. coil.* Additional data were obtained in a succeeding experiment (see Table 2). Settling-out tests were carried out in centrifuge tubes under standardized conditions. The amount of wet sediment settling out from milks derived from concentrates exposed to high-velocity gradients was 8 to 12 times greater than the amount deposited from milks derived from concentrates processed with the 3-in. holding tube.

The proportion of fat, in agreement with theory, was considerably greater in the sedi-

ment belonging to the more destabilized concentrate. Superior homogenization followed by second-stage rehomogenization, although it served to reduce grain size, was insufficient to bring about parity between the aggregates in the two sets of concentrates.

Results of storage tests (Tables 3 to 5) on a number of concentrates varying in composition confirmed the results of settling-out tests on reconstituted milks. The data in Table 5 pertain to 28% skimmilk concentrates. The effect of substituting the 3-in. holding section for the Mallory coil was to decrease two- to threefold the amount of sediment depositing in the concentrates over a period of 56 wks at 21.1 C. In the processing of the concentrates

TABLE 3

Sediment deposition in sterile 28% skimmilk concentrates as influenced by the velocity gradient during sterilization

Concentrate	Type of holding section	Storage time — weeks at 21.1 C			
		33		56	
		Viscosity (centipoises)	Sediment (% [in.])	Viscosity (centipoises)	Sediment (% [in.])
Control	3-in. pipe	gel	.....	gel	.....
Additive	3-in. pipe	6.8	1.8 (0.04)	6.8	4.0 (0.08)
Additive	5/16-in. Mallory coil	7.0	4.0 (0.08)	7.0	9.2 (0.18)

Forewarming at 137.8 C for 15 sec. Concentrate sterilized at 137.8 C for 15 sec, and homogenized at 71.1 C and at 7,500 psi first, and 500 psi second stage. Additive — 0.75 lb polyphosphate (14.5 P atoms per chain) per 100 lb S.N.F. Shear gradient varied in holding section of sterilizer.

TABLE 4  
Sediment deposition in additive containing 36% sterile concentrates as influenced by the velocity gradient during sterilization

Type of holding section	Storage time — weeks at 21.1 C			
	32		55	
	Viscosity (centipoises)	Sediment (% [in.])	Viscosity (centipoises)	Sediment (% [in.])
2-in. pipe	18.2	3.0 (0.06)	16.6	5.5 (0.11)
4-in. pipe	17.0	2.5 (0.05)	16.4	4.2 (0.08)
5/16-in. Mallory coil	16.2	9.0 (0.18)	16.1	12.7 (0.25)

Fat-to-solids ratio — 1:3.3. Forewarming at 137.8 C for 15 sec. Concentrate sterilized at 137.8 C for 15 sec, homogenized 71.1 C, and at 7,500 psi first, and 500 psi second stage; 0.75 lb polyphosphate (14.5 P atoms per chain) added per 100 lb SNF.

superior homogenization was employed as a terminal step.

The advantage gained in reducing the magnitude of the mean velocity gradient in the processing of whole milk 3 to 1 concentrates of normal composition was likewise quite marked (see Table 4). Some additional advantage was obtained in using the 4-in., in preference to the 2-in. holding section. The reduction in the amount of sediment depositing over a period of 55 wks at a storage temperature of 21.1 C was realized without any adverse effect on the resistance of the concentrates to gelation.

*Effect of increasing the ratio between fat and solids.* The substituting of fat for solids-not-fat, thereby increasing the ratio between them from 1:2.3 to 1:1.8, increased the resistance to settling-out (see Table 5). Thus, high-fat concentrates processed with the 4-in. holding tube in the line showed no measurable amounts of sediment deposition even after 67 wks of storage at 21.1 C.

*Substitution of evaporative in lieu of forced convective cooling.* Theory dictates prudence in avoiding high-velocity gradients even in the

cooling of sterilized concentrates. This is best accomplished by substituting evaporative for forced convective cooling. In this procedure an additional advantage is gained; for, with the use of evaporative cooling it becomes possible to sterilize concentrates of much lower solids concentrations. Evaporative cooling may be relied upon to effect evaporation and to increase the solids concentration from a lower to the desired level. The relationship between the change in concentration effected by evaporative cooling as a function of sterilizing and product temperature is shown in Table 6. The change in concentration is very nearly proportional to the difference between sterilizing temperature and finished product temperature. Under extreme conditions the change may amount theoretically to as much as 6.7%. Actually, the increase in concentration will be less by an amount depending on heat losses caused by radiation.

In Table 7 results of an experiment are shown in which evaporative cooling was combined with relatively quiescent sterilization to yield a product which, after 54 wk of storage

TABLE 5  
Sediment deposition in high-fat 36% sterile concentrates as influenced by the velocity gradient during sterilization

Concentrate <sup>a</sup>	Type of holding section	Storage time — weeks at 21.1 C			
		30		67	
		Viscosity (centipoises)	Sediment (% [in.])	Viscosity (centipoises)	Sediment (% [in.])
Control	4-in. pipe	loose gel	.....	gel	.....
Additive	4-in. pipe	18.5	none	18.4	none
Additive	Mallory coil				
	5/16-in. diam	22.0	3.8 (0.08)	20.4	<sup>b</sup> 6.5 (0.13)

<sup>a</sup> Fat-to-SNF ratio = 1:1.8. Forewarming at 37.8 C for 15 sec. Concentrate sterilized at 137.8 C for 15 sec, homogenized at 71.1 C, and at 7,500 psi first, and 500 psi second stage. Additive — 0.75 lb polyphosphate (14.5 P atoms per chain) per 100 lb SNF. Velocity gradient was varied in holding section of sterilization unit.

<sup>b</sup> Sediment quite loose and easily dispersible.

TABLE 6

Increase (calculated) in solids concentration of HT-ST sterilized milk concentrates brought about by evaporative cooling

R	0						0.3					
T <sub>s</sub> (C)	138						150					
T <sub>e</sub> (C)	20	30	60	20	30	60	20	30	60	20	30	60
ΔC <sup>a</sup> (%)	4.9	4.5	3.3	5.4	5.0	3.8	6.0	5.6	4.1	6.7	6.3	4.9

R = ratio between fat and total solids; T<sub>s</sub> = sterilizing temp. T<sub>e</sub> = evaporating temperature; ΔC = change in concentration on evaporative cooling.

$$^a \text{ — Calculated from formula: } \Delta C = \frac{C_p [S_f R + S_{nf} (1-R) - S_w] + 100 S_w}{100 L + [S_f R + S_{nf} (1-R) - S_w]} \cdot \frac{C_p \Delta T}{C_p \Delta T}$$

C<sub>p</sub> = Per cent solids in product (28% for R = 0, and 36% for R = 0.3). S<sub>f</sub>, S<sub>nf</sub>, and S<sub>w</sub> = specific heats, respectively, of fat, solids-not fat, and water (0.50, 0.32, and 1.02 cal/g). L = Latent heat of vaporization (585, 580, and 563 cal/g, respectively, at 20, 30, and 60 C). ΔT = T<sub>s</sub> - T<sub>e</sub>.

at 21.1 C, contained between one-third and one-half the sediment present in the corresponding concentrate cooled by forced convective cooling, and approximately one-seventh the sediment present in the concentrate sterilized with the 5/16-in. diameter holding tube in the line. In this experiment, the sterilizing temperature was 137.8 C; the incoming product concentration was 32.5%. The finished product temperature was 62.8 C, the vacuum 24 in., and the change in concentration achieved was 3.9%.

## DISCUSSION

Theory predicts the existence of a critical value of the particle radius for any given value of the velocity gradient. This must not be exceeded if orthokinetic flocculation is to be kept within reasonable bounds. The value of the critical radius varies inversely as the cube root of the velocity gradient if Equation [1]

is given credence. For relatively small values of the gradient, the particles must grow thermally to an objectionably large size before the influence of the orthokinetic effect makes itself felt. The thermal stability of forewarmed unhomogenized 3 to 1 concentrates of normal composition and pH precludes such marked growth; hence, these concentrates are stable when small gradients are employed, and undergo augmented growth only under the influence of very large gradients.

Unforewarmed 3 to 1 concentrates inherently lack thermal stability. Hence, the particles in such concentrates will grow to an objectionable size even in the presence of small gradients.

The absence of orthokinetic effects, therefore, does not guarantee against heat coagulation; it merely insures that aggregation of heat-stable systems will not reach objectionable proportions.

TABLE 7

Sediment deposition in additive containing sterile 36% concentrate as influenced by the shear gradient during sterilization and by evaporative cooling

Type of holding section	Type of cooling	Storage time — weeks at 21.1 C			
		32		54	
		Viscosity (centipoises)	Sediment (% [in.])	Viscosity (centipoises)	Sediment (% [in.])
4-in. pipe	Heat exchange 5/16-in. Mallory coil	15.1	2.7 (0.05)	16.7	6.5 (0.13)
4-in. pipe	Evaporative <sup>a</sup>	16.6	1.5 (0.03)	15.8	2.5 (0.05)
5/16-in. Mallory coil	Heat exchange 5/16-in. Mallory coil	14.2	11.0 (0.22)	13.0	17.3 (0.35)

Ratio fat to SNF = 1: 2.3. Forewarming at 137.8 C for 15 sec. Concentrates sterilized at 137.8 C for 15 sec., homogenized at 62.8 C, and at 7,500 psi first, and 500 psi second stage. Additive — 0.75 lb polyphosphate (14.5 P atoms per chain) per 100 lb SNF.

<sup>a</sup> 32.5% concentrate at 280 F cooled by evaporation to 62.8 C under 24-in. vacuum and concentrated thereby to 36.4%.

The theories of thermal and orthokinetic coagulation assume that the process of aggregation is irreversible. This certainly is not the case when orthokinetic aggregation is shear-induced. Under such circumstances, disaggregation effects come into play. The degree of disaggregation depends on shearing stresses acting on the surface of the particle and, hence, increases both with the magnitude of the stress and the size of the aggregate. It is also dependent on the cohesiveness of the aggregate. Hence, loose aggregates tend to fall apart with greater ease than hard, compact aggregates. The data in Table 2 on the composition of the aggregates and on the effect of homogenization on aggregate composition may be construed to support the view that the fat globules in a mixed aggregate tend to reduce the cohesiveness of the aggregate.

Even under the extreme condition of plant practice, velocity gradients encountered in the holding step are not strong enough to promote extensive disaggregation of relatively small aggregates. Only when the aggregates have grown to a palpable size, thus giving rise to graininess, does the disaggregating influence of high-velocity gradients in the holding step manifest itself. Thus, in the processing of thermally unstable unforwarmed 3 to 1 concentrates, the aggregates grow thermally to a point at which they are apparent macroscopically, and under these circumstances the grain size in concentrates processed with the Mallory holding coil in the line will be significantly smaller than the grain size in concentrates processed with the large holding tubes. On the other hand, it has been observed in many instances that the aggregates in thermally stable forwarmed concentrates exposed to only mild stresses during the holding step will, when homogenized, resist disintegration; consequently, the viscosity and the settling-out characteristics of such concentrates will not change appreciably as a consequence of homogenization.

The reasonable value obtained for the critical particle radius may be construed to mean that the assumptions on which the calculations are based are reasonable, that the actual values of the mean velocity gradient are not far removed from the assumed value, and that the encounter rates under initial conditions of polydispersity are not far removed from those calculated on the basis that the suspensions are initially monodisperse.

Although in terms of volume concentration, the ratio between fat and protein in milk concentrates does not differ by much, in terms of

number concentration, the ratio is extremely small; hence, the number of thermal fat-fat encounters will be insignificant compared to the number of protein-protein encounters.

Shear-induced encounters between fat globules can reach significant proportions in the presence of large-velocity gradients, inasmuch as the encounter rate varies as the cube of the particle radius. At elevated temperatures the increase in coagulation rate is no doubt offset by the disaggregating effects of the gradient, and is more than offset when terminal homogenization is employed to achieve, in addition to the disintegration of clusters, the breaking up of the primary fat globules.

At this point, it is worth re-emphasizing that superior homogenization as a terminal step falls short of bringing about the desired degree of disaggregation, the more so the greater the degree of aggregation. Moreover, as the data in Table 2 show, homogenization efficiency is significantly reduced as a consequence of marked aggregation.

#### CONCLUSIONS

By operating under conditions of low-velocity gradients during the holding period, by using evaporative in lieu of forced convective cooling, and by increasing the fat at the expense of solids-not-fat in the processing of 3 to 1 concentrates, one may realize a margin of safety against settling-out, without which forewarming and superior homogenization following sterilization taken by themselves will not insure against the settling-out defect reaching objectionable proportions.

#### APPENDIX

The derivation of the equation (see Equation [1]) for orthokinetic flocculation assumes that any chosen particle will follow a straight path toward a target particle. Actually, the particle center follows a stream-line which in the vicinity of the target particle tends to veer away. Thus, the number of collisions predicted by the equation is too large. Müller's (5) theory for sedimenting particles considers the hydrodynamic factor. The collision ratio between two sedimenting particles with radii  $r$  and  $R$  is

$$\text{given by the expression, } \frac{\sin C}{C}$$

$$C = \frac{v}{D_r + D_r} \left\{ r + R \left[ \frac{r(2R+r)}{4(R+r)^2} - \frac{3}{2} \ln \frac{R+r}{R} \right] \right\}$$

Applying Müller's equation to a situation in which the relative velocity  $v$  derives from the presence of a velocity gradient, we have found

that the average value of  $v$  can be calculated from the following relationship:

$$N_r \pi (R+r)^2 v = \frac{4}{3} (R+r)^3 G N_r$$

therefore:

$$v = \frac{4}{3\pi} (R+r) G$$

Whether Müller's or Smoluchowski's formulation is employed, our conclusions remain the same.

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