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The Viscosity and Surface Tension of Dispersions of Sucrose, Lactose, Skim Milk Powder, and Butterfat

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THE VISCOSITY AND SURFACE TENSION OF DISPERSIONS OF SUCROSE, LACTOSE, SKIM MILK POWDER, AND BUTTERFAT

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INTRODUCTION

Viscosity is considered an important property of ice cream mixes because it is related to the body or texture of ice cream. Viscosity is also believed to be related, in general, to the whipping property of mixes which is important in determining over-run in making ice cream.

Surface tension is important in ice cream since it, also, is related to the whipping property of ice cream mixes. A low surface tension is believed to be related to the ease of incorporating air into the mix.

The object of this research was to study the viscosity and surface tension of water dispersions of certain components of ice cream mixes; namely, sucrose, lactose, butterfat, and milk proteins.

This work was done in single component systems with the exception of the butterfat emulsions where a protein was necessary to stabilize the emulsion. In this manner, it was thought that the behavior of each component could be studied much better than in the complex system represented in milk, cream, or ice cream mixes.

The research was undertaken as a part of a systematic study of the different components of ice cream mixes with the idea that the data obtained could be used in connection with other data in the study of the viscosity and surface tension of ice cream mixes. A portion of this subject has been covered by Percival (33) in his thesis submitted in 1926 for the Master of Science degree.

REVIEW OF LITERATURE

The literature dealing with the viscosity and surface tension of milk and cream is quite brief and, with a few exceptions, the work might be classed as qualitative rather than quantitative. The cause of this is to be found, in part, in the complex system presented in milk and cream. In many cases, however, the apparatus used was not calibrated so that results could be reported in absolute units. A survey of the literature, however, gives a knowledge of the factors causing variations in the viscosity and surface tehsion,which is essential in carrying out experimental work.

Soxhlet (37) in 1876, studied the viscosity of milk at temperatures ranging from 0°C. to 30°C. He found that the viscosity decreased with an increase in temperature.

Babcock (2) showed that the albuminoids in milk had a greater influence on the viscosity than any of the other constituents. Later, Babcock and Russell (3, 4) found that the consistency of milk was due to two factors, one dependent on the characteristics of the dissolved portion and the other on the suspended matter. The consistency of the dissolved portion was believed to be due to the casein which existed in a semi-colloidal condition. The consistency of the suspended portion was due to the fat which had a purely mechanical influence depending on the size, surface tension, and ability of the globules to form aggregates or clots. Centrifuging and pasteurizing cream was found to diminish the consistency. The consistency could be restored, however, by adding lime in the form of the sucrate.

Babcock, Russell, and Woll (5) reported that, in addition to changes in consistency, the whipping qualities of cream were injured by pasteurization. Microscopic study showed that the fat globules were clustered in raw milk, but that after pasteurizing a homogeneous distribution of the globules occurred.

Woll (44) found that pasteurizing cream at ordinary temperatures decreased viscosity while heating to higher temperatures increased the viscosity. Steiner (38) and Taylor (39) confirmed these results but Jensen (26) stated that heating milk above the pasteurizing temperature did not increase the viscosity.

Kobler (27) studied the factors affecting the viscosity of milk and found that the viscosity varied with individual cows. He also reported that a reduction in the fat or protein content of milk lowered the viscosity. The viscosity increases during storage at a low temperature.

Evenson and Ferris (22) showed that holding milk for one week at a temperature of 3°C. increased the viscosity over that of milk held at 3°C. for one hour. The effect was not due to the presence of fat since skim milk showed an increase in viscosity under the same conditions. Pasteurization at ordinary temperatures was found to lower the viscosity slightly but at high temperatures pasteurization caused a slight increase in viscosity due perhaps to the coagulation of the albumin.

Whitaker, Sherman and Sharp (43) determined the viscosity of skim milk. They found that the viscosity decreased with increasing temperature and considered the change due largely to the proteins.

Palmer and Dahle (32) found that re-made milk, in general, had a higher viscosity than natural milk. This they attributed to a change in the proteins in the drying process.

Oertel (30) showed that the viscosity of milk varies with the fat content although not directly. He found that the size of the fat globules influenced the viscosity. The content of casein and albumin was also found to influence the viscosity.

Dumaresq (17) stated that the main factors influencing the viscosity of cream are acidity, temperature and fat content. He considered acidity the most important and found that the viscosity varied little with increasing acidity until a certain critical acidity was reached after which it increased very rapidly. The change at the critical acidity of skim milk was very slight, showing that the critical acidity was related to the fat content. He found also that the viscosity was a quadratic function of the fat content when other factors were constant.

Mortenson (29) studied the viscosity of creams from 12 per cent to 35 per cent butter fat before and after pasteurizing and after aging at 0° to 4°C. for 24, 48 and 72 hours. He found the changes in viscosity due to pasteurization were greater in rich cream than in thin cream. He attributed the greater changes in the viscosity of rich cream on pasteurization to the high viscosity of the rich cream. He also showed that the viscosity of cream increases on aging at low temperatures.

Dahlberg and Hening (15), studying the viscosity of cream, found that the viscosity of milk and cream increased with aging and with increased percentages of fat, but that both effects were variable. The effect of fat was most noticeable at concentrations above 20 per cent. Pasteurization slightly reduced the viscosity of milk and greatly reduced the viscosity of aged cream. The effect of aging was inhibited by pasteurization.

The viscosity of cream could be greatly altered by the condition of the milk-fat at the time of separation. If the fat were in a semi-solid condition at the time of separating, the viscosity both of pasteurized and raw milk was increased. If the milk were separated immediately after milking or pasteurization a low viscosity resulted like that of normal pasteurized milk. This effect as well as that of aging was dependent on the size and grouping of the fat globules.

Bishop and Murphy (11) found that homogenizing cream increased its viscosity so that a 15 per cent cream after homogenization resembled a 25 per cent cream. Pasteurizing homogenized cream destroyed the viscosity imparted by homogenization.

Doan (16) found that when fluid milk or cream, containing sufficient fat, (8 per cent or more), was homogenized at a pressure of 2000 to 3000 pounds per square inch, a peculiar structure of fat was created. The globules were reduced in size and tended to clump together, the clumps arranging themselves so as to occupy the greatest possible space..

Many attempts have been made to find a satisfactory expression for the relation between concentration and viscosity in solutions. The linear formula is $\eta = (1 + Ax)\eta_0$ where η_0 is the viscosity of the pure solvent, x is the volume of solute per unit volume of solution, and A is a constant. This expression holds only for dilute solutions. Arrhenius(1) proposed the logarithmic expression $\eta = A^x\eta_0$, but this holds only for dilute solutions.

Green, (23) working with sugar solutions, found that none of the existing formulae held and proposed the equation $\eta = A^{\frac{v}{w}}\eta_0$ where v is the volume of sucrose, and w the weight of water in unit volume of solution. This expression he found to hold over the whole range of concentration.

Powell (34) found that, by substituting the concentration in grams per 100 grams of solvent for the volume concentration in Arrhenius' formula $\eta = A^x\eta_0$, the expression held for sugar concentrations up to 60 grams per 100 grams of water.

Bingham (10) considered that fluidity is the additive property in the viscosity of mixtures. The fluidity concentration curves, plotted from the observations of Bingham and Jackson (9) on the viscosity of sugar solutions, differed widely from the linear.

For two phase systems, Hatschek (24) obtained the equation $\eta_1 = (1 + 4.5f)\eta_0$, where η_1 is the viscosity of the system, η_0 that of the liquid continuous phase and f is the ratio,

$$\frac{\text{total volume of disperse phase}}{\text{total volume of the system}}$$

Einstein (21) also obtained a similar expression. This formula states that the viscosity of a system of deformable particles suspended in a liquid is a linear function of the volume of the disperse phase. This formula holds, in general, for low concentrations and coarse particles only, according to Dunstan and Thole (20).

In determining the viscosity of dispersoids, particularly emulsoids, Weissenberger (42) noted that the viscosity decreased on successive passages of the dispersoid through a capillary tube until finally a constant value was reached which was dependent on the size of the capillary. This effect has been reported with such relatively simple compounds as glycerin. He attributed this effect to the formation of structures of a higher order in the dispersoid.

Leighton and Williams (28) studied this effect in ice cream mixes, and found that the increased viscosity due to aging the mix could be destroyed upon agitation without the incorporation of air. A constant value was obtained for the viscosity which was lower than the apparent viscosity. The constant value they termed the basic viscosity. The relationship between the logarithm of the basic viscosity and the concentration in parts per 100 solvent proved to be a linear function (Arrhenius). This study of the basic viscosity was confirmed by Turnblow and Nielson (40).

In studying the influence of fat and proteins on the surface tension of milk, Kobler (27) found that variations in both fat and proteins caused changes in surface tension. Dahlberg and Hening (15) found, likewise, that an increase in the concentration of fat resulted in a lowering of the surface tension. Behrendt (8) found that reducing the protein content of milk increased the surface tension, but that the fat had little influence on this property.

Burri and Nussbaumer (12) found that milk held at temperatures above 20°C. showed little decrease in surface tension on aging, while milk cooled at 10°C. for half an hour showed a greater depression of the surface tension. On cooling at 0°C. the surface tension increased a little. The surface tension increased upon heating the processed milk to 50°C. for half an hour so that the depression almost disappeared. Bauer (7) confirmed this result. However, Quagliariello (35) found that the decrease in surface tension, due to cooling milk at 0° to 10°C. for half an hour, was permanent and the surface tension could not be restored by heating.

Dahlberg and Hening (15) found that aging generally decreased the surface tension while pasteurization increased it.

Work done by Clarke and Mann (14) and also Butler (13) showed that the surface tension of sugar solutions increases with the concentration. Butler's results showed that the surface tension of sugar solutions was a linear function of the sugar concentration in mols. However, von Lippmann (41) found that the surface tension was decreased by increasing concentrations of sucrose.

THEORETICAL DISCUSSION

Viscosity

Viscosity is the property of liquids which offers resistance to flow. It refers only to forces within the liquid, so it is frequently considered as internal friction. The internal forces are due to mutual attraction between the particles which tends to resist flow or deformation.

Bingham (10) defines viscosity as the ratio between the deforming force and the amount of deformation. This holds only for linear or viscous flow. If the deforming force is large enough the ratio suddenly decreases and turbulent or hydraulic flow occurs.

In viscous flow, the deforming force sets up a shearing of one layer of liquid past another. From the definition of viscous flow, the amount of deformation or the velocity of one layer compared to another layer is proportional to the deforming force. The velocity of one layer compared to another will also be dependent on the distance between the two layers. If F is the shearing force, V the velocity of one layer in respect to another layer, and d the distance between the two layers, the proportionality expression is,

$$V \propto Fd$$

or

$$F \propto \frac{V}{d}$$

Introducing a coefficient, $F = \eta \frac{V}{d}$

Then $\eta = \frac{Fd}{V}$

The coefficient η is designated as the coefficient of viscosity. It may be defined as the tangential force, on a unit area of either of two parallel planes at unit distance apart, required to move one plane at unit velocity in reference to the other plane, the distance between being filled with the viscous substance.

The viscosity coefficient is expressed in dyne seconds per square centimeter. The unit is called the poise. For convenience, a unit one one-hundredth times as large, known as the centipoise, is used. By using the centipoise troublesome decimals are eliminated since the value for water is almost exactly unity at 20°C.

In molecular dispersions the viscosity varies with the temperature, and with the concentration of the solute. In colloidal dispersions, however, there are more variables to consider. For colloidal dispersions of the emulsoid type, Ostwald (31) lists the following variables:

1. Degree of dispersity.
2. Previous thermal treatment.
3. Previous mechanical treatment.
4. Time or aging.
5. Solvate formation.
6. Electric charge.
7. Inoculation with small quantities of more viscous colloids.
8. Addition of electrolytes and non-electrolytes.

Of these factors, the first four listed are of major importance in this work.

In cream, or in butterfat emulsions, the influence of the degree of dispersity, or the size of the fat globules, can be demonstrated by homogenization. In homogenizing cream, the size of the fat flobules is greatly reduced, causing an increase in the volume occupied by the butterfat. An increase in viscosity results from the process.

Ordinarily in hydrophilic dispersions, the effect of time or of aging is considered to be due to hydration of the dispersed phase. While there is a slight increase in the viscosity of skim milk upon aging at a low temperature, the effect of aging on creams is due rather to a change in the condition of the fat globules. Raw creams, containing a high percentage of butterfat, show a very great increase in viscosity on aging. Furthermore, examination of aged cream by the mixroscope shows that the fat globules are clustered to form aggregates. Bancroft (6) states that the increased viscosity is due to this agglomeration of the fat globules. The aggregates decrease the amount of free water in the system and thus increase the concentration of fat causing an increased viscosity.

The effect of thermal treatment on the viscosity of cream is due to the breaking up of the clusters of fat globules resulting in a homogeneous distribution of the globules.

The effect of the mechanical treatment on the viscosity of cream may be due to two causes. First, the mechanical treatment may result in breaking up the aggregates of the fat globules and thus decrease the viscosity. Second, by homogenization the size of the fat globules may be reduced so as to increase the viscosity.

Surface Tension

At the surface of liquids there is a force which tends to contract that surface. The force is seen in falling drops which assume a spherical shape thus bringing the ratio of surface to volume to a minimum value. This force is known as surface tension. The surface tension is measured in terms of the force per unit length and generally is reported in dynes per centimeter.

In colloidal dispersions surface tension is important since the surface area is very great, due to the small size of the particles. In emulsions where one liquid is dispersed in another liquid, the two being immiscible, the surface area at the interface is tremendous in comparison with the amount of the emulsion. So, in milk and cream, where the fat exists in small golbules dispersed in the liquid, the surface tension is an important physical property.

The addition of substances to water may cause an increase or a decrease in surface tension. From a study of the energy relationships in solutions, the theory has been advanced that the concentration of a substance increasing the surface tension is lower at the surface than in the rest of the system, while the concentration of a substance decreasing the surface tension is greater at the surface than in the rest of the system. Thus the addition of substances increasing the surface tension increases the surface tension very little, comparatively, because the concentration of the substance is lower at the surface. This theory also explains why the addition of small amounts of substances lowering the surface tension gives very marked results. The concentration of a substance lowering the surface tension is greater at the surface than in the rest of the system, so the addition of a small amount of the substance may cause a marked lowering of the surface tension.

EXPERIMENTAL RESULTS

Viscosity

All solutions used were made up on the basis of percentage by weight. Later, for theoretical reasons it became necessary to calculate the concentrations used in the viscosity determinations on the basis of grams per 100 grams of water.

The experimental work was done at a temperature of 25°C. Whenever possible the determinations were made with the apparatus immersed in a Freas thermostat, running at 25°C ± .02°. This temperature was selected as the lowest temperature practical for work during the warmer part of the year.

For determining viscosity, four Ostwald tubes were used having the water constants 94.4, 90.6, 92.2 and 94.9 respectively. The absolute viscosity was calculated from the equation,

$$\eta = \frac{d \times t}{d_0 \times t_0} \; n_0$$

where η = viscosity of the system, η_0 = viscosity of water at 25°C. t = time of flow of system; t_0 = time of flow of water; d_0 = density of water at 25°C; d = density of system. The viscosity of water was taken as .008937 poises at 25°C.

In determining the viscosity of butterfat emulsions it was found impossible to obtain concordant results with Ostwald tubes due to the emulsion sticking to the walls of the viscosity tube. An attempt was made to find a viscosity pipette similar to the standard pipette used in determining the viscosity of glues and gelatins. However, these pipettes were found to be unsuited for the range of viscosity found in milk and cream.

A coaxial type of viscometer was tried, but it was not suited to the purpose as it was designed for systems of high viscosity.

The viscometer finally used for the butterfat emulsions was a Redwood instrument, No. 1288. This instrument was not ideal for the purpose, but it was the only viscometer available which was suitable for determining the viscosity of such a liquid.

The viscometer was calibrated in the manner described by Herschel (25) and by Sheely (36). The viscosity is calculated from the equation,

$$\frac{n}{d} = At - \frac{B}{t}$$

where n = viscosity, d = density, t = the time of flow, and A and B are constants. Since A and B vary for different viscometers, it was necessary to calibrate the instrument in order to obtain absolute values.

The value of the constant A was fixed by determining the time of flow of a sample of castor oil of known viscosity and density obtained from the Bureau of Standards. The term, $\frac{B}{t}$, or the kinetic energy correction, may be disregarded. The value of A, therefore, is obtained from the equation $\frac{n}{d} = At$. The value of A for this particular viscometer was found to be .00249.

Having fixed the value of the constant A, the value of B was then found from the viscosity equation by determining the time of flow of solutions of known viscosity and density, having a value of t small enough so that the term $\frac{B}{t}$ was significant. For this purpose solutions of glycerin, sugar, and alcohol were used. The average value of B was found to be 1.84. Any error in the value of B is minimized since it is necessary that the time of flow be over 30 seconds in order that the flow be viscous. Thus any error in the value of B is divided by 30 at least.

A stop watch, graduated to read tenths of a second, was used to determine the time of flow. The watch was wound and allowed to run for five minutes before making a set of determinations.

The surface tension measurements were made by the DuNouy (18) method, using a model 10400 instrument. In making the surface tension measurements it was necessary to calibrate the apparatus since it did not give direct readings. The dial was calibrated by the use of weights, and the calibration checked for every set of determinations.

The sucrose and lactose used in this investigation were Pfanstiehl, C.P., special reagents having a specific rotation of +66.5° and 52.5° respectively. The butter - oil used in making the emulsions was obtained from the Land O'Lakes Creameries, and contained 99.5 per cent butterfat and .5 per cent water. The skim milk powder used was all taken from a single, carefully mixed sample. A moisture determination run at approximately 65°C. in a vacuum oven showed 6.16 per cent moisture.

The water used in making up the solutions was redistilled from potassium permanganate using a block tin condenser.

The viscosities of sugar solutions, ranging from 10 to 27 per cent sucrose, were determined by use of Ostwald tubes. The results are given in Table I.

The logarithm of the viscosity in centipoises was plotted against the concentration in grams per 100 grams of water for the results given in Table I. The graph shows the logarithm of the viscosity to be a linear function of the concentration. (Fig. I).

The type equation for this curve is

$$\log \eta = \log A + x \log B$$

where η = viscosity

x = concentration

A & B = constants.

The values of A and B were calculated from the data and found to be 0.8825 and 1.027 respectively. This equation may also be given in the form $\eta = ae^{kx}$ where η = viscosity

x = concentration

a & k = constants.

e = 2.718 (Natural base)

The values of a and k were found to be 0.8825 and .02655 respectively. The viscosity of a sugar solution at 25°C. can thus be calculated from the equation,

$$\log \eta = 0.05428 + 0.01153\,x \quad \text{or}$$

$$\eta = 0.8825e^{.0266x}$$

These equations will hold for concentrations up to 40 per cent or 66 grams per 100 grams of water.

TABLE I

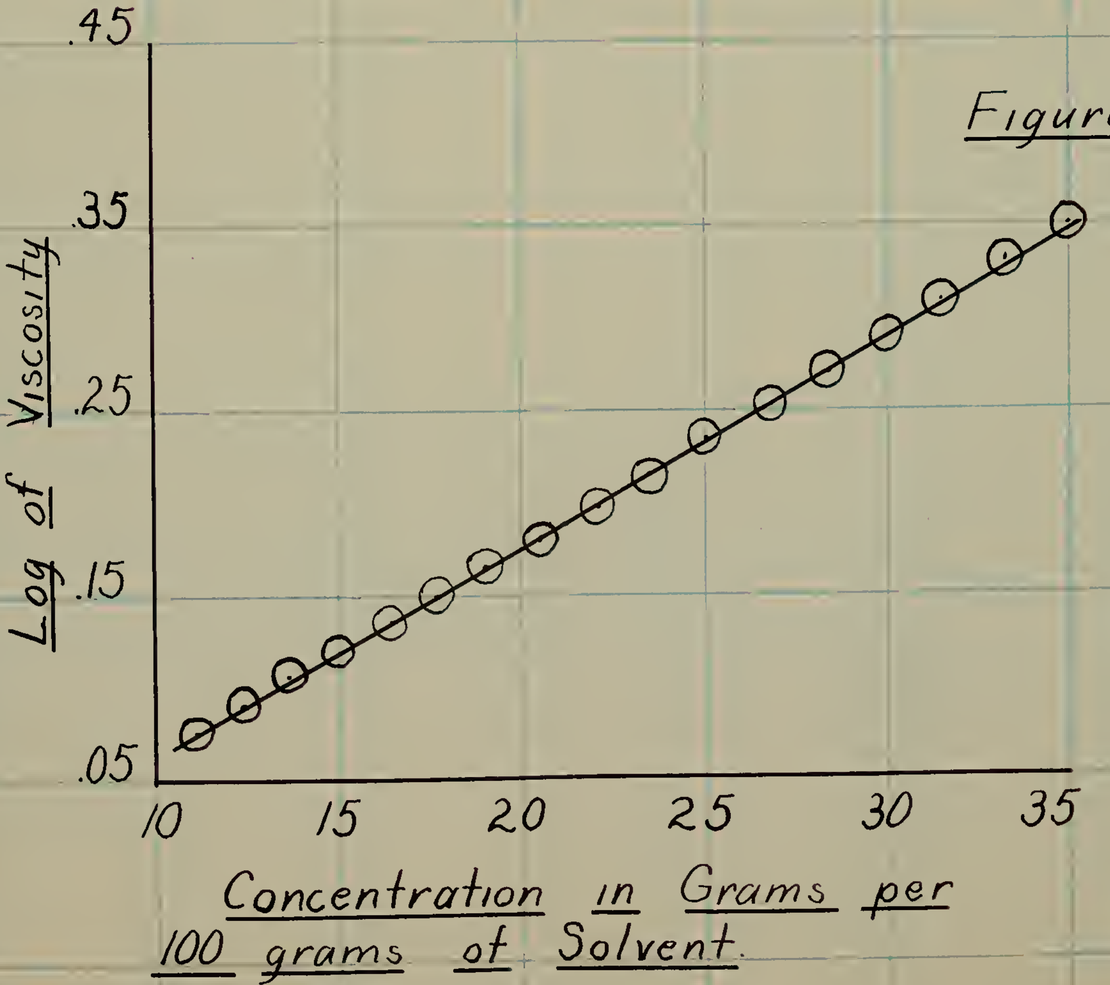
Viscosities of Sucrose Solutions from 10 to 27 per cent Sucrose.

Per cent	Grams per 100 gr.H_2O	Density	η in c.p.	Average η	Log η
10	11.11	1.037	1.182 1.187 1.186 1.184	1.185	.07372
11	12.36	1.041	1.225 1.232 1.236 1.228	1.231	.09026
12	13.63	1.046	1.268 1.277 1.275 1.275	1.273	.10483
13	14.94	1.049	1.310 1.314 1.314 1.307	1.311	.11760
14	16.28	1.054	1.360 1.363 1.362 1.359	1.361	..13386
15	17.65	1.058	1.406 1.419 1.421 1.407	1.413	.15014
16	19.05	1.063	1.465 1.464 1.469 1.469	1.467	.16643
17	20.48	1.067	1.514 1.522 1.523 1.512	1.518	.18127
18	21.95	1.072	1.573 1.586 1.579 1.573	1.578	.19811

TABLE I (Con't)

Per cent	Grams per 100 gr.H_2O	Density	η in c. p.	Average η	Log η
19	23.46	1.075	1.636	1.638	.21431
			1.638		
			1.643		
			1.634		
20	25.00	1.081	1.712	1.714	.23401
			1.714		
			1.717		
			1.712		
21	26.58	1.086	1.777	1.783	.25115
			1.790		
			1.784		
			1.782		
22	28.20	1.090	1.862	1.861	.26975
			1.866		
			1.862		
			1.855		
23	29.87	1.093	1.940	1.942	.28825
			1.952		
			1.949		
			1.928		
24	31.58	1.099	2.030	2.035	.30856
			2.044		
			2.025		
			2.040		
25	33.33	1.105	2.131	2.138	.33001
			2.155		
			2.134		
			2.134		
26	35.13	1.109	2.233	2.237	.34967
			2.250		
			2.234		
			2.232		
27	36.99	1.113	2.357	2.356	.37218
			2.373		
			2.353		
			2.341		

Figure I



Log η as a function of the Concentration of Sucrose.

The viscosities of lactose solutions from 1 to 15 per cent lactose were then determined. It was first planned to cover the range from 1 to 10 per cent, but the viscosity of 15 per cent solution was afterwards determined so that the viscosity of a lactose solution could be compared to that of a sucrose solution of the same strength. It was found that the 10 and 15 per cent lactose solutions had a viscosity slightly greater than the sucrose solutions of the same strength. The results are given in Table II.

The logarithms of the viscosities were plotted against the concentrations in grams per 100 grams of water. The function was found to be linear as in the case of sucrose. The equation for this curve was found to be:

$$\log \eta = -0.0488 + .0114x$$

or in form $\eta = ae^{kx}$ the equation was found to be:

$$\eta = 0.894e^{.0262x}$$

(Fig. II).

TABLE II

Viscosities of Lactose Solutions

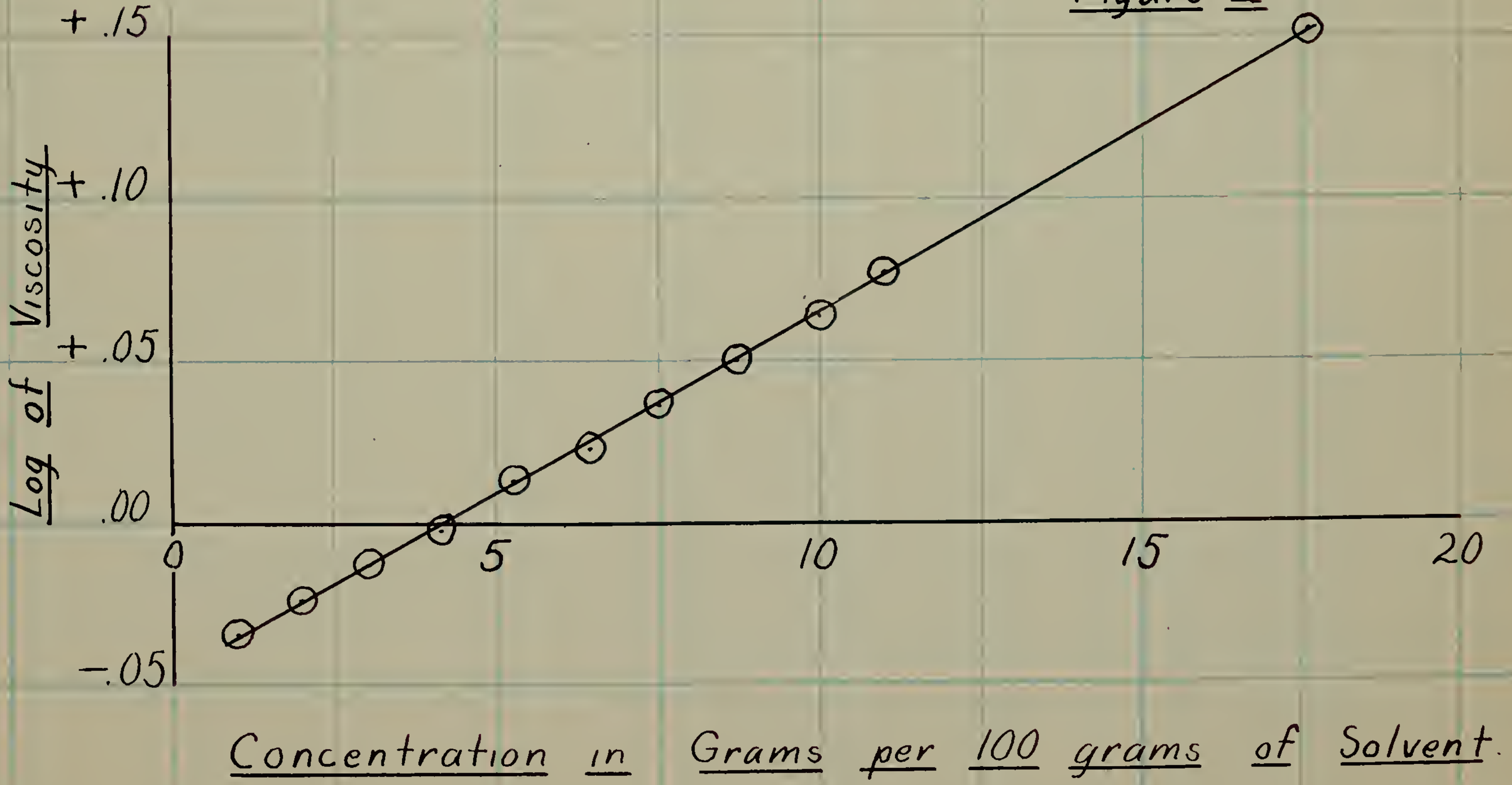
from 1 to 15 per cent Lactose.

Per cent	Grams per 100 gr. H_2O	Density	η in c.p.	Average η	Log η
1	1.010	1.002	0.9226 0.9215 0.9231 0.9223	0.9224	-0.03508
2	2.041	1.006	0.9481 0.9462 0.9453 0.9421	0.9454	-0.02438
3	3.093	1.008	0.9706 0.9674 0.9722 0.9711	0.9703	-0.01309
4	4.166	1.014	0.9974 0.9961 0.9926 0.9997	0.9965	-0.00152
5	5.263	1.017	1.025 1.026 1.029 1.025	1.026	0.01115
6	6.383	1.022	1.057 1.056 1.055 1.061	1.057	0.02407
7	7.527	1.025	1.086 1.089 1.088 1.085	1.087	0.03623
8	8.70	1.029	1.126 1.125 1.125 1.124	1.125	0.05115

TABLE II (Con't)

Per cent	Grams per 100 gr. H_2O	Density	η in c.p.	Average η	Log η
9	9.89	1.033	1.153 1.155 1.161 1.151	1.155	0.06258
10	11.11	1.036	1.199 1.199 1.196 1.189	1.196	0.0777
15	17.65	1.057	1.412 1.423 1.417 1.414	1.417	0.15137

Figure II



Log η as a function of the Concentration of Lactose.

Since it was planned to use a solution of skim milk powder in making the butterfat emulsions, it was thought advisable to study the effect of varying concentrations of skim milk powder on the viscosity. A large sample of skim milk powder was secured and mixed carefully to insure uniformity. In making up the dispersions, they were warmed to 60°C. and allowed to stand over night in a cool place before determining the viscosity. Care was taken to treat each dispersion in the same manner so that all conditions, such as aging, would be equal.

The viscosities of skim milk powder dispersions from 2.5 to 15 per cent skim milk powder were determined by the use of Ostwald viscosity tubes. The results are given in Table III. After the determinations were made it was found necessary to correct for the moisture in the sample. The moisture content was found to be 6.16 per cent. In the column, grams per 100 grams H_2O, in Table III, the concentrations are calculated oh a moisture free basis.

The logarithms of the viscosities were plotted against the concentrations in grams per 100 grams of water. The function proved to be linear although the points did not agree as well as in the case of the sugars. The dispersion of skim milk powder is not a true solution, and variations in the size of the particles would cause a variation in the viscosity. (Fig. III).

The equation for the best line drawn through the points is $\log \eta = 0.0225x - 0.0480$, or using natural logarithms, $\eta = 0.895e^{0.052x}$.

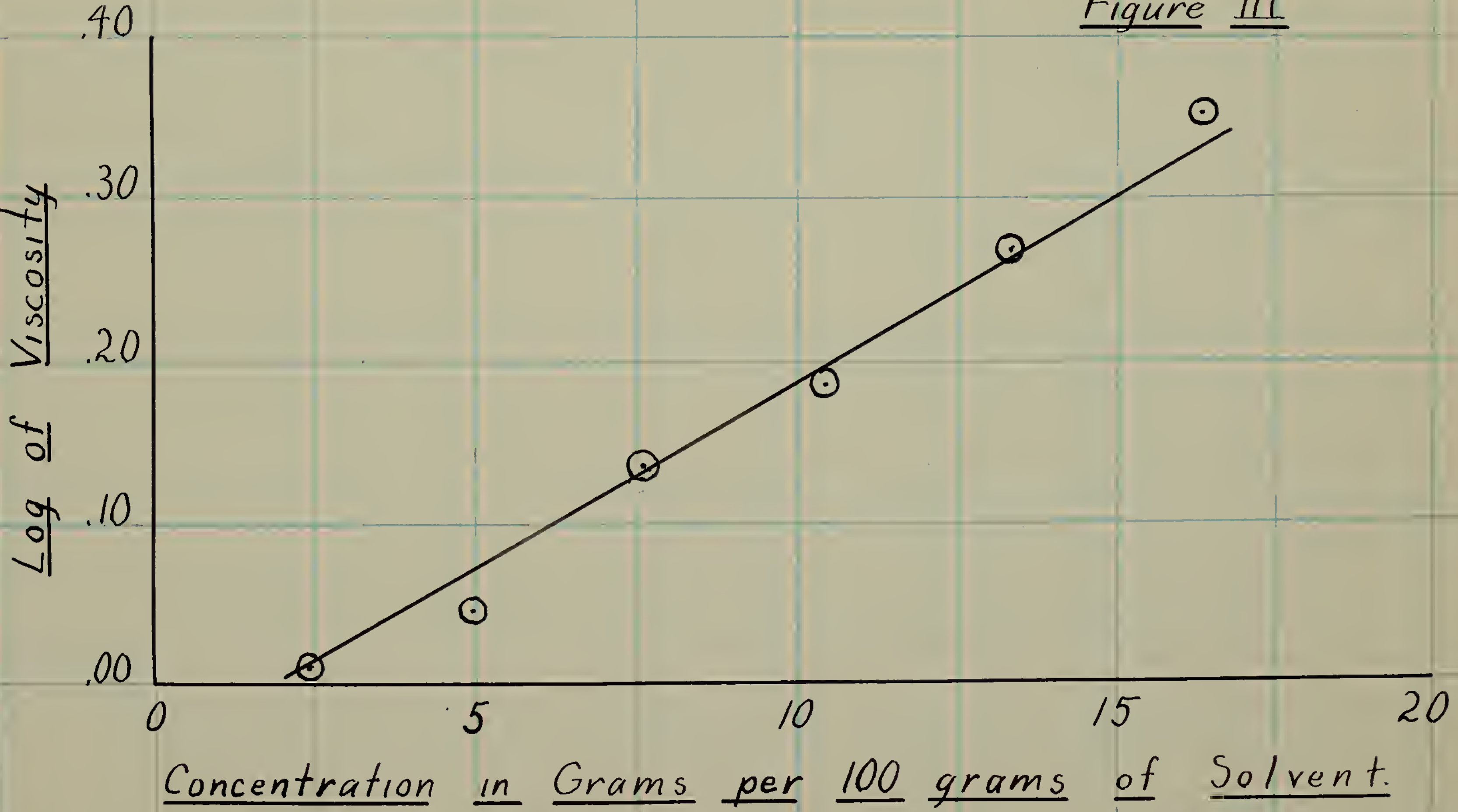
The curve shows a large increase in the logarithm of the viscosity with increasing concentration. The curve is steeper than the curve for sugar, which is considered to have a rapid increase in viscosity with incréasing concentration. The sharp increase in the viscosity in the case of skim milk powder is to be expected since proteins are known to have a marked effect in increasing viscosity.

TABLE III

Viscosities of Solutions of Skim Milk Powder from 2.5 to 15 per cent Skim Milk Powder.

Per cent	Grams per 100 grs. H_2O	Density	η in c.p.	Average η	Log η
2.5	2.41	1.006	1.022 1.025 1.023 1.023	1.023	0.00988
5.0	4.92	1.016	1.166 1.166 1.164 1.167	1.166	0.04766
7.5	7.57	1.026	1.367 1.383 1.370 1.390	1.378	0.13925
10.0	10.35	1.033	1.521 1.538 1.542 1.535	1.534	0.18583
12.5	13.29	1.042	1.851 1.868 1.861 1.848	1.857	0.26881
15.0	16.39	1.052	2.223 2.264 2.280	2.256	0.35334

Figure III



Log η as a function of the Concentration of Skim Milk Powder.

In studying the effect of butterfat on the viscosity, an attempt was made to disperse the butterfat in water by running the mixture through an homogenizer. The resulting emulsion was so unstable that it was found impossible to determine the viscosity accurately. Next, an attempt was made to emulsify butterfat by using small amounts of an emulsifying agent. The emulsifying agents used were gelatin and casein. However, after a few experiments it was evident that a great deal of experimental work would have to be done to determine the amount of emulsifying agent necessary to give the best results for the viscosity determinations. It was decided, therefore, to use a dispersion of skim milk powder in which to disperse the butterfat.

An emulsion of butterfat was made, containing 20 per cent butterfat, 5 per cent skim milk powder, and 75 per cent water. This was homogenized at a pressure of approximately 2500 pounds per square inch. From this emulsion, containing 20 per cent butterfat, emulsions containing 8, 10, 12, 14, 16, and 18 per cent fat were made by using a 5 per cent skim milk powder dispersion to dilute the 20 per cent emulsion. The viscosities of butterfat emulsions from 8 to 20 per cent butterfat were then determined by use of the Redwood viscometer.

The 8 per cent butterfat emulsions had a time of flow of less than 30 seconds, so a correction was applied to the viscosity equation by using a lower value of B than that obtained by the calibration. The corrected value of B was obtained from a determination of the time of flow for a liquid of known viscosity having a similar time flow. The value used for constant B was 1.74 instead of 1.84.

The results of the viscosity determinations are given in Table IV.

The logarithms of the viscosities plotted against the concentrations gave a straight line for concentrations of 10 per cent or above. The intercept of the curve at zero concentration is far below the logarithm of the viscosity of water, showing the slope of the curve is not the same for concentrations between 0 and 10. (Fig. IV).

The equation for the curve was found to be, $\log \eta = 0.079x - 0.619$, or $\eta = 0.240e^{0.182x}$.

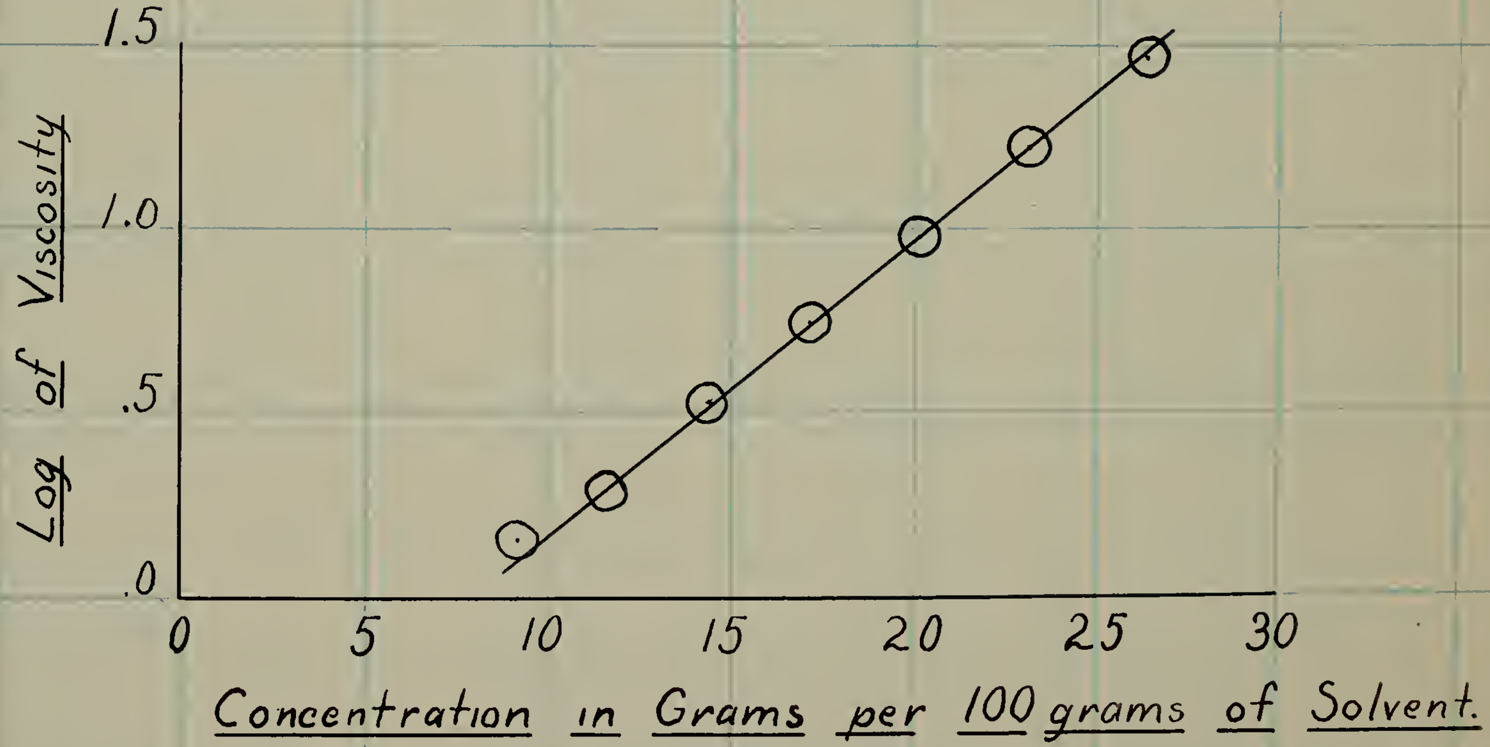
The equation will hold only for concentrations of 10 per cent or above, and its use above 20 per cent is, of course, open to question. Furthermore, the constants will probably differ if the conditions used in making the butterfat emulsion are not the same. However, the range from 10 to 20 per cent shows a logarithmic relationship quite definitely.

TABLE IV

Viscosities of Butterfat Emulsions from 8 to 20 per cent Butterfat.

Per cent	Grams per 100 grs. H_2O	Density	η in c.p.	Log η
8	9.2	1.007	1.46	.16435
10	11.7	1.005	1.88	.27416
12	14.4	1.003	3.30	.51851
14	17.1	1.002	5.51	.74115
16	20.1	1.000	9.88	.99476
18	23.1	.997	16.1	1.20683
20	26.3	.995	29.2	1.46538

Figure IV



Log η as a function of the Concentration of Butterfat in an Emulsion.

In order to compare the viscosities of the butterfat emulsions of varying concentration with cream of the same concentrations, the viscosities of creams covering the same range of concentration of butterfat were determined. A sample of pasteurized cream containing 34 per cent butterfat was standardized by use of skim milk containing 9.60 per cent solids to give creams containing 8, 10, 12, 14, 16, 18, and 20 per cent butterfat.

The viscosities of these creams were determined by use of the Redwood viscometer. The viscosity equation was changed as mentioned before to calculate the viscosity of the 8 per cent cream since the time of flow was less than 30 seconds. The results are given in Table V.

The plot of the logarithm of the viscosity against the concentration in grams per 100 grams of solvent shows that the function is logarithmic over the range from 8 to 20 per cent buttefat. (Fig. V).

The equation for the line was found to be

$$\log \eta = 0.020x + .023 \quad \text{or} \quad \eta = 1.05e^{0.046x}.$$

The viscosity of the 34 per cent cream was determined. The viscosity found experimentally differs considerably from that found from the curve. The value obtained experimentally was 12.76 c.p. while the calculated value is 16.93 c.p. Above 20 per cent butterfat, therefore, the equation does not hold in this case. A plot of the results for cream reported by Dahlberg and Hening (15), using concentrations in grams per 100 grams of water, showed the curve to be a straight line up to 34 per cent butterfat.

TABLE V

Viscosities of Cream from

8 to 34 per cent Butterfat.

Per cent	Grams per 100 grs. H_2O	η in c.p.	Log η
8	9.7	1.45	.16137
10	12.44	1.68	.22531
12	15.31	2.13	.32838
14	18.32	2.30	.36173
16	21.51	2.72	.43457
18	24.86	3.14	.49693
20	28.41	3.76	.57519
34	60.29	12.76	1.10585

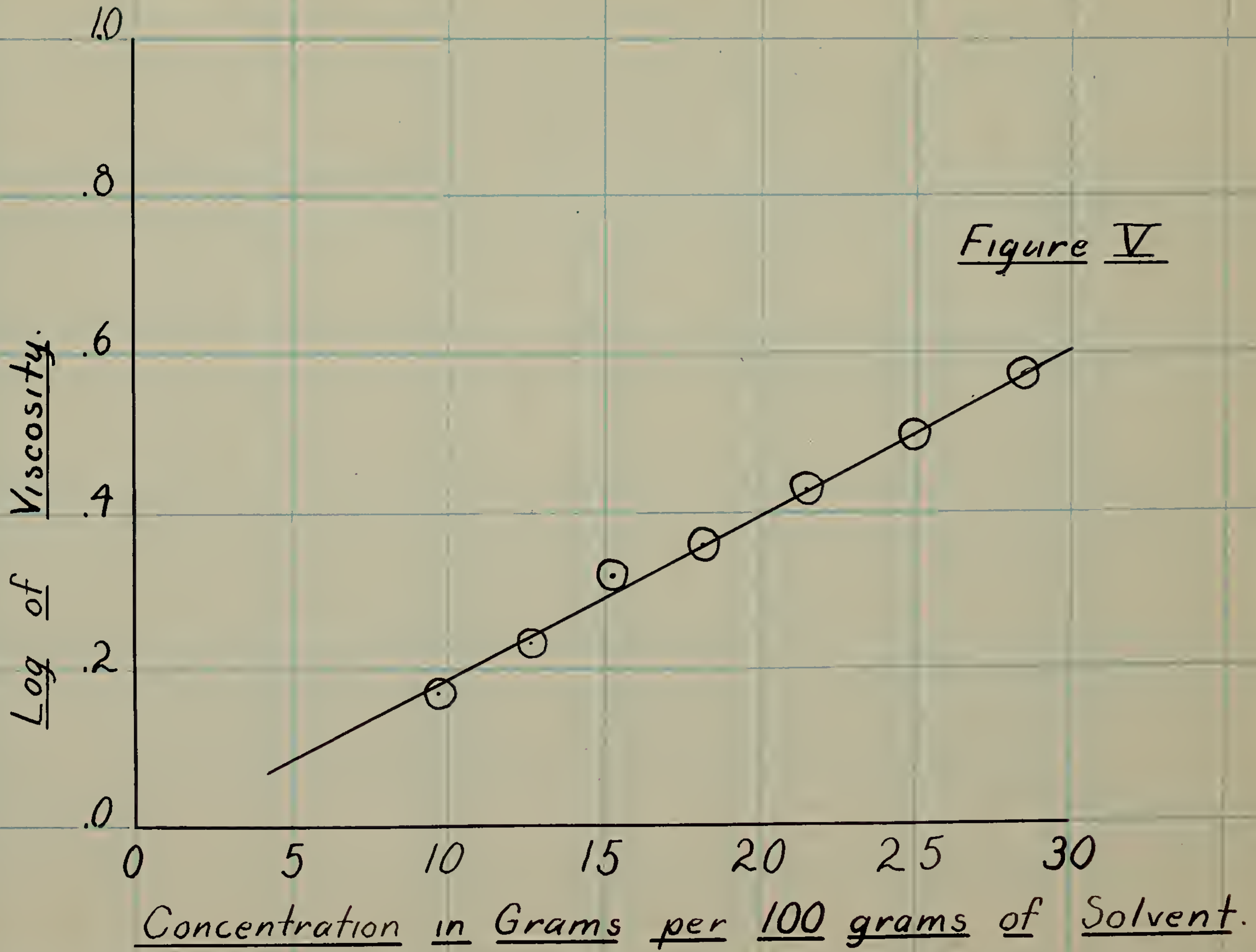


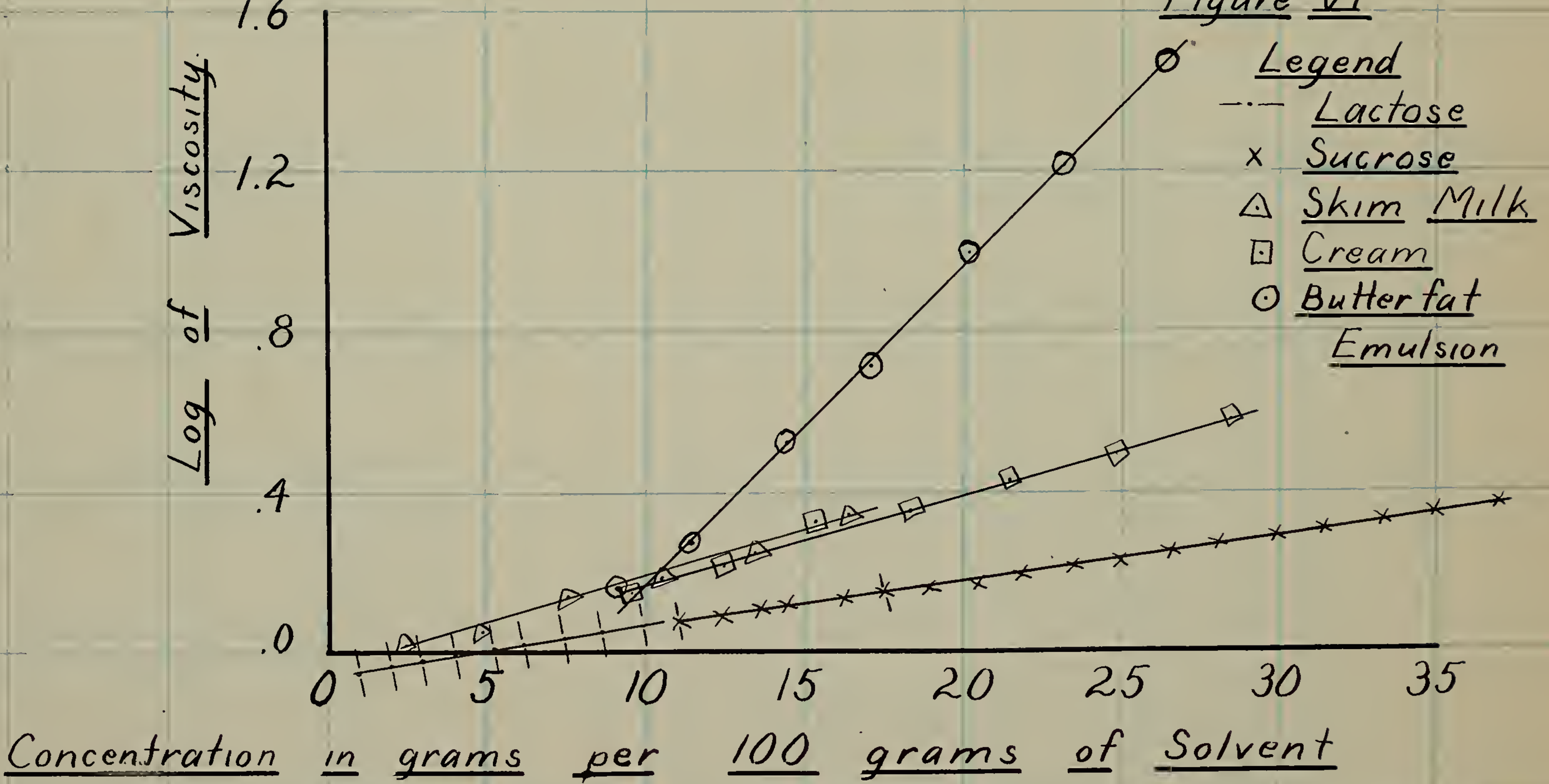
Figure V

Log η as a function of the Concentration of Butterfat in Cream.

A comparison of the slopes of the curves for the various materials gives an index of the effect of increasing concentration on the viscosity. The slope for sucrose was found to be 0.0115 and for lactose, 0.0114. These two sugars give molecular dispersions in water. For skim milk powder the slope was found to be 0.0225. This is to be expected since proteins have a very marked effect on the viscosity. In the case of skim milk powder, the dispersion is considered to be partly colloidal, at least. For the butterfat emulsion the slope is 0.079 which is much greater than any of the others. This rapid increase in the viscosity with increases in concentration may be explained by the very small size of the fat globules. The homogenization process used in making the emulsion resulted in very small fat globules. Doan (16) reported, also, that homogenization led to a peculiar distribution of fat globules, or a grouping, which would have an effect on the viscosity. The slope of the curve for cream is 0.020 which is much less than the slope for the butterfat emulsion which shows very clearly the increase in viscosity due to homogenization.

The log n-concentration curves for the different materials were all plotted on one graph in order to show graphically the difference in the rate of change of the logarithm of the viscosity with respect to changes in the concentration. (Fig. VI).

Figure VI



Comparison of Log η - Concentration Curves of the Different Materials.

Surface Tension

The surface tensions of sucrose, lactose, skim milk powder, butterfat emulsions, and cream, were determined over the same range of concentrations as the viscosities. The dispersions used in the viscosity determinations were also used in the surface tension measurements.

The surface tensions of solutions of sucrose from 10 to 27 per cent sucrose were determined. The results are given in Table VI. These results show a slight increase in surface tension with increasing concentration. The increase in surface tension was found not to be directly proportional to the concentration. No attempt was made to plot the results since inspection shows no regular increase. The lack of a direct proportionality, however, may be due to the fact that the increase in surface tension with small changes in concentration was so small that the error involved in making the measurements obscured the proportionality.

The surface tensions of solutions containing from 1 to 10 per cent lactose were determined. The results are given in Table VII. Here again a slight increase in the surface tension with increasing concentrations was found. The results show no direct proportion between concentration and surface tendion.

The surface tensions of dispersions of skim milk powder in water were determined over a range of concentratione from 2.35 to 14.08 per cent skim milk powder. The results are given in Table VIII. A marked decrease in the surface tension was found since the surface tension of water is 72.8 dynes per centimeter at 25°C. while the lowest concentration of skim milk powder has a surface tension of 51.0 dynes per centimeter. The surface tension of the skim milk dispersions was found to decrease as the concentration increased up to concentrations of 9.38 per cent. Above that concentration no change occurred in the surface tension with increasing concentration up to 14.08 per cent.

According to DuNouy (19), it is necessary to allow the surface of a colloid to stand undisturbed for a period of at least two hours in order to permit complete surface adsorption to take place. Since this work was carried out with materials which undergo decomposition very quickly and which , in the case of cream of butterfat emulsions, separate into a layer of higher concentration at the surface due to the rising of the fat globules, it was impossible to allow the surfaces to age. While an error may occur from a lack of time to complete surface adsorption, this error is unavoidable with this type of substances.

The surface tensions of butterfat emulsions containing from 8 to 20 per cent butterfat were determined. The results are given in Table IX. The results show no relationship, either general or direct, between the concentration and the surface tension. The surface tension varied very little and was found to be low.

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The surface tensions of creams from 8 to 20 per cent butterfat were determined. The surface tension showed a decrease with increasing concentrations of butterfat as is shown in Table X. It is impossible to state, however, that a mathematical relationship exists between the concentration and surface tension from these results.

The formula connecting surface tension, concentration, and surface adsorption developed by Gibbs and Thomson independently, gives a very good qualitative explanation of the different effect of materials lowering the surface tension and materials raising the surface tension. The formula states that,

$$u = -\frac{c}{RT} \cdot \frac{dr}{dc}$$

Where u = the excess or defect in the concentration of the dispersed phase in the surface layer.

c = concentration

R = gas constant

T = absolute temperature

$\frac{dr}{dc}$ = the rate of change of the surface tension in respect to changes in concentration.

R, T, and c will always be positive. Therefore, if $\frac{dr}{dc}$ is positive, u will have a negative value. If $\frac{dr}{dc}$ is negative, u will be positive. This formula, therefore, states that in the case of materials increasing the surface tension, there will be a lower concentration in the surface layer than in the rest of the system. In the case of materials lowering the surface tension, the concentration in the surface layer will be greater than in the rest of the system.

The equation shows that a small quantity of solute can increase the surface tension slightly, but may decrease the surface tension very considerably. Now a study of the data in Tables VI to X shows that in the case of sucrose and lactose, which increased the surface tension, the increase was very slight even at the highest concentrations used. In the case of the skim milk powder, which lowered the surface tension very markedly, the effect of the lowest concentration used was very nearly the same as the effect of the highest concentration used. Percival's (33) work shows, in the case of the proteins, casein, albumin, and gelatin, that the addition of small amounts of these proteins caused a large decrease in the surface tension, but the effect decreased as the concentration increased which is to be expected from Gibbs' equation. As the concentration in the surface layer approaches the limiting value, or saturation, the effect of further additions approaches zero.

The results of the surface tension determinations on the butterfat emulsions and cream cannot be readily explained by the use of Gibbs' formula. In the case of emulsions, the droplets are believed to carry an electric charge which causes a repulsion of the globules, and from this repulsion a lower effective surface tension results. The surface tension of the emulsions was lower than that of the skim milk dispersions. The surface tensions of the creams were much lower than either the skim milk dispersions or the butterfat emulsions, and showed a decrease in surface tension with increasing concentrations of butterfat.

TABLE VI

The Surface Tension of Sucrose Solutions from 10 to 27 per cent Sucrose.

Per cent	Dynes per cm.	Per cent	Dynes per cm.
10	73.3	19	74.9
11	73.7	20	74.7
12	74.7	21	75.3
13	74.4	22	75.0
14	74.4	23	75.2
15	74.4	24	75.2
16	74.8	25	75.4
17	74.7	26	75.4
18	74.8	27	75.2

TABLE VII

The Surface Tension of Lactose Solutions from 1 to 10 per cent Lactose.

Per cent	Dynes per cm.	Per cent	Dynes per cm.
1	72.8	6	72.6
2	73.1	7	72.8
3	73.2	8	73.1
4	73.3	9	73.3
5	73.1	10	73.9

TABLE VIII

The Surface Tension of Solutions of Skim Milk Powder from 2.35 to 14.08 per cent Skim Milk Powder.

Per cent	Dynes per cm.
2.35	51.0
4.69	50.8
7.04	50.9
9.38	50.6
11.73	50.6
14.08	50.6

TABLE IX

The Surface Tension of Butterfat Emulsions from 4 to 20 per cent Butterfat.

Per cent	Dynes per cm.
4	48.9
6	49.1
8	49.1
10	49.5
12	49.7
14	49.0
16	48.3
18	49.1
20	49.8

TABLE X

Surface Tension of Cream from 8 to 20 per cent Butterfat.

Per cent	Dynes per cm.
8	39.6
10	39.3
12	38.9
14	38.6
16	38.2
18	37.6
20	36.9

CONCLUSIONS

The relationship between the concentration and the viscosity of sucrose solutions ranging from 10 to 27 per cent sucrose may be expressed by a mathematical equation. The equation was found to be,

$$\log \eta = 0.05428 + 0.01153x \quad \text{or}$$
$$\eta = 0.8825e^{.0266x}$$

The relationship between the concentration and the viscosity of lactose solutions is similar to that of sucrose. The relationship may be expressed by a formula which was found to be,

$$\log \eta = -0.0488 + .0114x$$

or in form $\eta = ae^{kx}$ the equation was found to be:

$$\eta = 0.894e^{.0262x}$$

The concentration and viscosity of dispersions of skim milk powder, containing from 2.5 to 15 per cent skim milk powder, were found to be related by the equation,

$$\log \eta = 0.0225x - 0.0480, \text{ or}$$
$$\eta = 0.895e^{0.052x}$$

The concentration and viscosity of emulsions of butterfat, containing from 10 to 20 per cent butterfat, were found to be related by the equation,

$$\log \eta = 0.079x - 0.619 \quad \text{or}$$
$$\eta = 0.240e^{0.182x}$$

This equation does not hold for emulsions containing less than 10 per cent butterfat.

It is possible to express the relationship between the concentration and viscosity of creams containing from 8 to 20 per cent butterfat by an equation. The equation was found to be,

$$\log \eta = 0.020x + .023 \quad \text{or}$$

$$\eta = 1.05e^{0.046x}$$

The surface tension of solutions of sucrose was found, in general, to increase with increasing concentrations of sucrose. No direct mathematical relationship was found.

The surface tension of lactose solutions from 1 to 10 per cent lactose was found to increase with increasing concentrations of lactose. No direct relationship was found.

The surface tension of dispersions of skim milk powder from 2.38 to 14.08 per cent skim milk powder was found to be much lower than the surface tension of the dispersion medium and to be almost constant for the concentrations used.

The surface tension of butterfat emulsions from 8 to 20 per cent butterfat showed little change with changes in the concentration of the butterfat. The surface tension was slightly lower than that of a skim milk powder dispersion having the same percentage of skim milk powder as the emulsion.

The surface tension of creams from 8 to 20 per cent butterfat decreased with increasing concentrations of butterfat.

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