

Use of the Coulter Counter and a Digital Computer for the Evaluation of Stability Ratios in Flocculating Monodisperse Systems

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The combined use of the Coulter counter and a digital computer has been shown to be a powerful tool in evaluating stability ratios of flocculating monodisperse polystyrene lattices. The method involves comparing the experimental and theoretical depletion rates of the singlet species and corrections have been applied for the presence of higher species at the start of the flocculation experiment. The influence of particle size, particle concentration, electrolyte concentration, and surfactant upon suspension stability has been examined.

INCREASING ATTENTION has been focused in recent years upon the interfacial properties of powdered drugs. This interest may be attributed, at least in part, to the growing importance of suspensions as a dosage form. Although a considerable number of useful research and review articles on flocculation in pharmaceutical systems, has been published (1-12), there is still need for further investigation. A number of areas in this field are still in need of further illumination; there is, for example, some controversy as to the exact nature of the role of zeta potential in the flocculation of supracolloidal systems (2-9, 13).

The process of flocculation in a disperse system involves the increase in apparent particle size due to the formation of doublets, triplets, and higher species. Thus, *a priori*, one can consider any experimental technique of use in particle-size measurement, for the study of flocculation kinetics. A number of classical physicochemical methods such as light scattering (14), sedimentation (15, 16), and microscopy (11) can be used for work on flocculation kinetics. However, many of these techniques are of limited utility since they measure the total size of floccules and do not distinguish between the singlet, doublet, triplet, *etc.*, species. The Coulter counter (17, 18) measures particle volume and if the original system is monodisperse, the presence of doublets,

triplets, and even quadruplets can be detected. The Coulter counter technique is capable of measuring several thousand particles per second and once the operator has gained experience in the use of the equipment, data can be accumulated in a remarkably short space of time. However, even with the use of electromechanical or electronic calculators, the conversion of raw counts to singlet and multiplet concentrations is both tedious and time consuming. When such data have to be substituted into the somewhat complex flocculation equations (see later), the calculation process is further extended and the possibility of error greatly increased. In the present paper the combination of a Coulter counter and a digital computer is shown to provide a powerful tool for the study of the interfacial properties of monodisperse supracolloidal suspensions.

Most of the physicochemical studies of flocculation kinetics have been limited to systems containing particles well within the colloidal range, the upper limit of which is about 0.5μ (19), whereas powdered drugs are almost invariably of supracolloidal dimensions (20, 21). Equations governing colloidal flocculation might well be expected to require modification on application to supracolloidal systems, since sedimentation may occur even in the singlet species. In the present paper a method is presented for studying the flocculation of particles in the size range $0.5-5.0 \mu$.

THEORY

The kinetics of flocculation in colloidal systems is dependent upon two factors, the Brownian motion

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of the particles and the extent of their mutual interaction when in close proximity. The simplest case may be defined as when there is no mutual repulsion between the particles, such that any particle entering the sphere of interaction of another coalesces irreversibly with it. This situation was considered by Von Smoluchowski (22, 23) in his theory of rapid coagulation.

Smoluchowski assumed that at the start of flocculation, N_0 spherical particles of equal size are present per unit volume of liquid and that flocculation is started by a sudden removal of the mutual repulsion of the particles. The number of particulate encounters is determined by considering the process as a diffusion problem.

He showed that the number of aggregates of k single particles at any time, t , N_k , is given by the expression

$$N_k = \frac{N_0(t/T_{1/2})^{k-1}}{(1+t/T_{1/2})^{k+1}} \quad (\text{Eq. 1})$$

$T_{1/2}$ is the coagulation time, defined as the time required to reduce the total number of particles by one-half. The coagulation time may be calculated from the equation

$$T_{1/2} = \frac{1}{4\pi D_i R N_0} \quad (\text{Eq. 2})$$

where D_i is the diffusion constant of the particles and R is the distance between the centers of two particles when flocculation occurs. This is, in the simplest case, $2r$, where r is the radius of the primary particles. The derivation of these equations has been given by Overbeek (24).

Equation 1 becomes for the singlet

$$N_1 = \frac{N_0}{(1+t/T_{1/2})^2} \quad (\text{Eq. 3})$$

and for the doublet

$$N_2 = \frac{N_0 t/T_{1/2}}{(1+t/T_{1/2})^3} \quad (\text{Eq. 4})$$

The total number of particles in the system becomes equal to

$$\sum_{k=1}^{\infty} N_k = \frac{N_0}{1+t/T_{1/2}} \quad (\text{Eq. 5})$$

Figure 1 shows a graph of the number of each species up to triplet as a function of time (taken from Reference 24, p. 282).

This theory has been tested experimentally by Turkevich (25) using a gold sol of spheres having a diameter of 400 Å., and good agreement was found with K values up to 6 in the early stages of flocculation.

The Eq. 5 gives no information on the stability of colloids, and although these systems are thermodynamically unstable they can remain, with little apparent change, over long periods of time.

Smoluchowski extended his theory to cover slow flocculation by assuming that only a fraction, α , of the collisions leads to permanent coalescence. The course of coagulation is still governed by Eq. 1, but Eq. 2 now becomes

$$T_{1/2} = \frac{1}{4\pi D_i R N_0 \alpha} \quad (\text{Eq. 6})$$

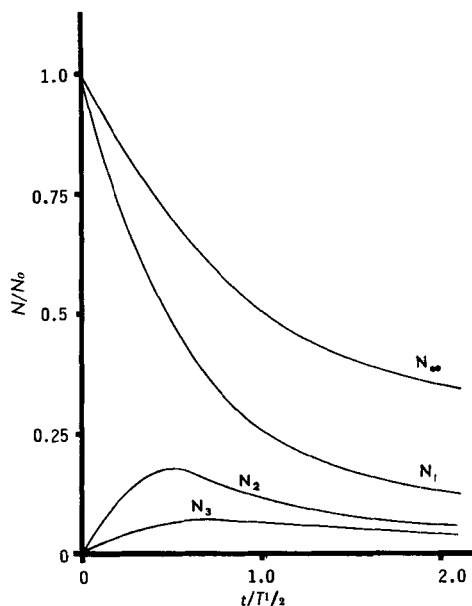


Fig. 1—Theoretical curves for the proportion of various species as a function of time in rapid flocculation (taken from Reference 24).

Smoluchowski assumed that α , which can be called the "collision efficiency," remains constant over the whole period of coagulation but Van Arkel and Kruyt (26, 27) and Tuorila (28) showed that this is not the case and that the rate of coagulation decreases with time. This has been explained by Reerink and Overbeek (29) by the fact that the repulsion between aggregates is larger than that between individual particles.

No theory was available to link the quantity α with such parameters as the zeta potential and electrolyte concentration, and Fuchs (30) modified Smoluchowski's theory to enable collision efficiency to be predicted from a consideration of the energy barrier between the particles.

The total energy of interaction between two particles, V_t , is the sum of the attractive contribution V_a and the repulsive contribution V_r . This potential V_t retards the process for rapid coagulation by a factor W such that

$$W = 2r \int_{2r}^{\infty} \left[\exp\left(\frac{V_t}{kT}\right) \right] \frac{dS}{S^2} \quad (\text{Eq. 7})$$

where r is the particle radius, S is the distance between the particle centers = $2r$ at coagulation, k is the Boltzmann constant, and T is the absolute temperature.

W denotes the ratio of rapid to slow coagulation, and when V_t is zero for all separations of the particles, $W = 1$ and rapid coagulation occurs according to the Smoluchowski equation. For slow coagulation, Eq. 5 becomes

$$N_k \frac{N_0}{1+t/T_{1/2}(\text{slow})} = \frac{N_0}{1+t/T_{1/2}(\text{fast})} W \quad (\text{Eq. 8})$$

and the coagulation time becomes

$$T_{1/2} = \left(\frac{1}{4 \pi D_1 R N_0} \right) W \quad (\text{Eq. 9})$$

From a comparison of Eqs. 6 and 9

$$W = \frac{1}{\alpha} \quad (\text{Eq. 10})$$

with α always < 1 and W in theory, always > 1 . W can be < 1 (24), and this is explained by the presence of London attraction forces which accelerate coagulation. Equation 7 describes the retardation in flocculation in the initial stages when not many secondary particles have formed, and Overbeek (Reference 24, p. 285) has stated that it is not possible to extend the theory to cover collisions between multiple particles. The dependence of W on such factors as particle size, electrolyte concentration, and valency, has been reviewed by Verwey and Overbeek (31), Ottewill and Watanabe (32-36), Somasundaran, Healy, and Fuerstenau (37), and the theory was extended to a dispersion containing two types of particle by Hogg, Healy, and Fuerstenau (38).

Determination of Stability Ratios in Mono-disperse Systems—From Eqs. 6 and 9 the collision efficiency α , or stability ratio W , could be determined by simply measuring the change in total concentration of particles with time and recording the $T_{1/2}$. This would, however, not take into account any changes in collision efficiency during the coagulation process and, if coagulation were slow, would require a lengthy experiment. Equations 1, 3, and 4 can be used to determine α or W at any period of time during the reaction provided a method is available to distinguish between the various species in the system. A recent series of papers (39-41) by Higuchi *et al.*, has demonstrated the suitability of the Coulter counter to determine the relative proportions of singlets, doublets, triplets, and sometimes quadruplets in flocculating monodisperse polystyrene lattices. Study of Fig. 1 shows that the species that undergoes the greatest change in concentration during flocculation is the singlet, and therefore measurement of the concentration of this species is likely to give greatest precision, especially where flocculation is slow, *i.e.*, where α is small and W large, or in systems of low concentration. We have used the Coulter counter to follow the rate of depletion of the singlet species in flocculating monodisperse systems and have used a digital computer to compare this rate with that predicted by the Smoluchowski theory, to evaluate the stability ratio.

Method of Calculation—From Eq. 3 the relative proportion of singlets to the total original concentration at any time t , is equal to

$$\frac{N_1}{N_0} = \frac{1}{(1 + t/T_{1/2})^2} \quad (\text{Eq. 11})$$

by rearranging

$$t = \left(\sqrt{\frac{N_0}{N_1}} - 1 \right) T_{1/2} \quad (\text{Eq. 12})$$

and a graph of t against $\sqrt{(N_0/N_1 - 1)} T_{1/2}$ should be a straight line with a slope of 1 if rapid coagulation occurs. When coagulation is slow, the graph has a slope of α or of $1/W$.

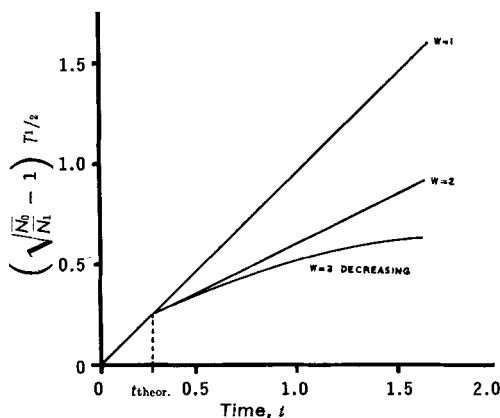


Fig. 2—Theoretical plot of $\left(\sqrt{\frac{N_0}{N_1}} - 1 \right) T_{1/2}$ as a function of time at different stability ratios.

Since, however, it was not possible to start with 100% singlets, the systems at $t_{\text{exp.}} = 0$ always appeared to have already been flocculating for a small time $t_{\text{theor.}}$. This value was obtained by substituting the value of N_1/N_0 obtained at $t_{\text{exp.}} = 0$, into Eq. 11 and it was added to all subsequent values of $t_{\text{exp.}}$ in the flocculation process. This method was considered to be valid since at the beginning of the experiment the relative proportion of singlets, doublets, and triplets was always in accord with that which would be predicted from Fig. 1.

Figure 2 shows a graph of t against $\left(\sqrt{N_0/N_1} - 1 \right) T_{1/2}$ for three systems where $W = 1$, $W = 2$ and where $W = 2$ initially, but falls during the course of the experiment. The graph depicts results obtained from theoretical calculations and illustrates the application of the correction factor $t_{\text{theor.}}$ to obtain the true effective zero time.

The coagulation time $T_{1/2}$ was calculated from Eq. 2 (see Reference 24, p. 282), which for water at 298°K. becomes equal to

$$T_{1/2} = \frac{2 \times 10^{11}}{N_0 \times 3.6 \times 10^9} \text{ hr.} \quad (\text{Eq. 13})$$

In this paper the above approximate equation has been used. However, in studying the flocculation of true colloidal systems when absolute rather than apparent W values can be determined the numerator, under the above conditions, should be stated more precisely as 1.63×10^{11} .

EXPERIMENTAL

A Coulter counter model "B"¹ was employed with 30- μ and 50- μ orifices. The model B is a dual threshold machine, and by setting the upper and lower threshold controls at suitable values, about that which corresponds to the volume of the species to be measured, counts can be obtained directly. Electrolyte, consisting of 1% sodium chloride, filtered through 0.45 μ Millipore² filters, was used with the 50- μ orifice and 2% sodium chloride filtered through 0.45 μ and 0.100 μ Millipore filters for the

¹ Manufactured by Coulter Electronics Ltd., Dunstable, Bedfordshire, England.

² Marketed by Millipore (U.K.) Ltd., Wembley, Middlesex, England.

30- μ orifice. Background counts, which were always low, were taken on the electrolyte and measurements of the particles were made at concentrations which enabled coincidence to be ignored.

The flocculation was carried out in 150-ml. stoppered flasks which were immersed in a water bath maintained at $25 \pm 0.1^\circ$.

Materials—Samples of Dow³ polystyrene lattices of diameter 0.714 μ and 1.305 μ were used, as obtained from the manufacturer.

The standard deviations quoted by the manufacturer for these lattices were 0.0053 μ for the 0.714- μ sample and 0.0158 μ for the 1.305- μ sample. Standard deviations determined from cumulative number/percentage size distribution curves obtained using the Coulter counter were somewhat higher than these values, 0.02 μ and 0.025 μ , respectively. However, the samples are sufficiently monodisperse to enable the vast majority of singlets to be differentiated from higher species, and for the Smoluchowski equation to be employed. Wachtel and La Mer (42) have also noted larger standard deviations of monodisperse polystyrene lattices, measured on the Coulter counter, than those stated by the manufacturers.

Suspensions were diluted in filtered distilled water which had been freshly distilled from an all-glass still and the working level was between 1.0×10^7 and 1.0×10^8 particles/ml. Aluminum chloride hexahydrate, $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$, was selected as the flocculating electrolyte since it was hoped to extend the work to pharmaceutical suspensions and aluminum was considered the most suitable trivalent metal from the viewpoint of toxicity.

General Method—All glassware used in the flocculation experiments was cleaned with chromic acid and rinsed six times with distilled water and twice with filtered distilled water. All apparatus used for measurement on the Coulter counter was rinsed with filtered electrolyte. Suspensions of the latex particles were prepared at 90% of the required concentration and 90.0 ml. was placed in the reaction flasks and allowed to reach the experiment temperature. Volumes of 2.0 ml. of the suspension were withdrawn and diluted in filtered electrolyte to a suitable working level. A count of singlets, doublets, triplets, and all species above was taken and the results used to determine the original total number of latex particles in the system.

Although ultrasonics and a high speed dispersion mixer were tried, it was not found possible to produce a system of pure singlets to start the reaction. This is in line with the experience of Higuchi *et al.* (39) who took as their starting point 90% singlets, 8% doublets, 1% triplets, and 1% larger aggregates. The method of dilution was standardized as far as possible although variation in the technique was not found to affect the count significantly.

The experiment was initiated by adding 10.0 ml. of the flocculating electrolyte at a concentration of 10 times that required in the reaction mixture and at suitable intervals samples were withdrawn and

assayed for the singlet species and the total count. The concentration of singlet species, in the system initially, was calculated from the counts taken at the beginning and the dilution factor.

Experiments were designed to demonstrate the effects of particle size, concentration of particles, concentration of electrolyte, and the presence of surfactant on the stability ratio W .

Computer Program—An Elliot 803 digital computer was programmed in Algol 60 to compute values of W by means of the method outlined above. A print-out of the program is shown in the *Appendix*. Values of the Smoluchowski collision efficiency α were computed also but all results are expressed in terms of W . Mean values of W are given except in a few instances where a pronounced reduction in flocculation rate occurred at the end of the experiment. In these cases initial values of W are used.

RESULTS AND DISCUSSION

Effect of Concentration of Particles on W —The effect of different concentrations of particles on the stability ratio W , at high electrolyte concentrations, was measured using 0.714- μ polystyrene latex at concentrations of 10^7 to 10^8 particles per ml. The concentration of aluminum chloride was $5 \times 10^{-3} M$, which is considerably above the critical electrolyte concentration.

The results shown in Fig. 3 indicate a reduction in W with increasing concentration up to a level of about 5×10^7 particles per ml., above which no further decrease occurs. However, in the experiments at the two highest concentrations, it was found that the initial values of N_1/N_0 were very low (0.41 and 0.36, respectively) compared with values of about 0.84 for all of the other systems. This indicates that some spontaneous flocculation, or lack of deflocculation on dilution from the concentrate, occurs at these higher concentrations. While it is known (39) that these polymers contain small amounts of an anionic surfactant which affects stability, the energy barrier is evidently not sufficient to prevent flocculation at this particular concentration. Although values of W on these two systems were, therefore, determined on colloids which were already considerably flocculated, they are in good agreement with the systems at between 4 and 7×10^7 particles per ml. and which were only very slightly flocculated at the start of the experiment. Earlier workers (24, p. 322) with true colloids have reported both increases and decreases in stability with increasing concentration of colloidal

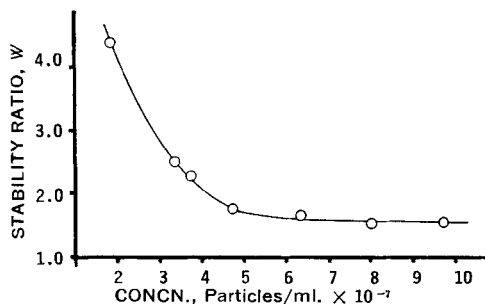


Fig. 3—Influence of particulate concentration on stability ratio.

³ Kindly supplied by Mr. B. J. Lippie, Dow Chemical Company, Midland, Mich.

⁴ This sample when calibrated by means of the 0.714- μ latex and a 1.099- μ latex was found to have a diameter of only 1.20 μ but this did not affect the experiments since Coulter threshold settings were calculated from the results of calibration by taking readings at $\frac{3}{2}$ and $\frac{1}{2}$ of the mean pulse height as recorded on the oscilloscope, as recommended in the instruction manual (43).

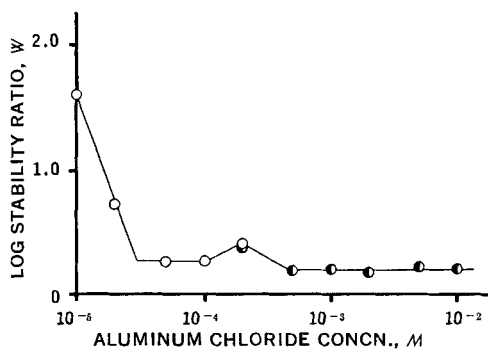


Fig. 4—Stability ratio W as a function of aluminum chloride concentration for 0.714- μ polystyrene latex. \circ and \bullet are separate experiments.

particles, but Klompé (24, p. 323) who corrected for exchange of flocculating ions against double-layer ions, reported that for a trivalent electrolyte, higher concentrations of sol required lower concentrations of electrolyte to produce flocculation. Further experiments are required with the lower concentrations of particles to determine whether different concentrations of electrolyte can produce reductions in the value of W . With aluminum salts there is also the possibility of peptization (44) although results on this system suggest that the concentration used in this experiment is higher than the peptizing concentration.

Effect of Electrolyte Concentration on W —After consideration of the results of the above experiment, a concentration of 5×10^7 particles per ml. was selected, and the stability ratio was determined at concentrations of aluminum chloride from 10^{-5} to 10^{-2} M . The results are shown in Fig. 4 (combined results of two experiments).

An initial rapid reduction in W occurs with very low concentrations of electrolyte. If this portion of the graph is extrapolated to $\log W = 0$, the critical flocculation concentration is 3.5×10^{-6} M . This is in good agreement with the range 9×10^{-6} to 9×10^{-5} given by Overbeek (24, p. 309) for the flocculation of negatively charged sols by aluminum salts. A small region of partial peptization occurs at 2×10^{-4} M and above this value W reverts to just below its original minimum.

Effect of Particle Size on W —It is of interest that the flocculation rate never reaches the Smoluchowski maximum for rapid coagulation where $W = 1$ and $\log W = 0$, in contrast to the experience of earlier workers with conventional colloids (24, p. 32) and of Ottewill and Shaw (45) with monodisperse polystyrene latex particles of diameter 0.06–0.4 μ . The particles used in the above experiments are less than twice the diameter of the largest used by Ottewill and Shaw. The minimum value of W is 1.5 which means that the maximum flocculation rate is 67% of the Smoluchowski rate. This is in good agreement with the results of Higuchi *et al.* (39) who found that the singlet/singlet flocculation rate constant of 1.83- μ polystyrene latex particles approached the Smoluchowski rate to within about a factor of two. Higuchi *et al.* (39) and Swift and Friedlander (46) have suggested that this deviation from theory is due to hydrodynamic interactions

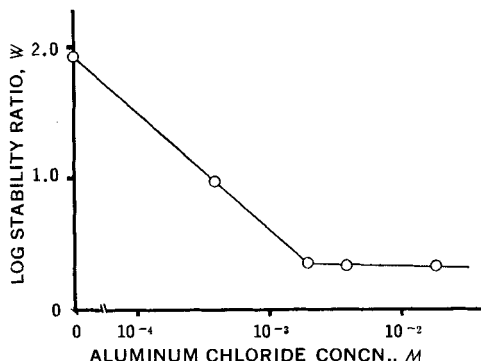


Fig. 5—Stability ratio W as a function of aluminum chloride concentration for 1.20- μ polystyrene latex.

when the particles are within one diameter of each other.

The maximum concentration of electrolyte used was 10^{-2} M and providing that peptization does not occur, the energy barrier at such high concentrations will be very low, or nonexistent. Values of W at these concentrations will not be an expression of the energy barrier but of the retardation of coagulation due to these hydrodynamic interactions.

At any concentration of electrolyte

$$W_{\text{exp.}} = W_{\text{true}} + C \quad (\text{Eq. 14})$$

where

$W_{\text{exp.}}$ = measured value of stability ratio.

W_{true} = contribution to stability due to energy barrier.

C = additional contribution due to hydrodynamic interaction.

Below a certain critical size, C becomes insignificant and

$$W_{\text{exp.}} = W_{\text{true}} \quad (\text{Eq. 15})$$

This critical size would seem to be somewhere in the region of 0.5 μ which is significantly the upper limit which some authors would place on the colloidal state.

If this explanation is correct one would expect that the minimum values of $W_{\text{exp.}}$ would be greater still for particles larger than 0.7 μ .

To test the above theory a similar experiment was performed using the 1.305- μ diameter latex from Dow (Coulter diameter 1.20 μ).

The results (Fig. 5) show that the general rule of increase in stability with increase in particle size stated by Verwey and Overbeek (31, p. 177) is obeyed, the critical concentration having been increased by a factor of about 10^2 . The minimum value of $W_{\text{exp.}}$ has increased from 1.5 to 2.0, as predicted above. Further experiments are planned on systems in the diameter range 0.5–2.0 μ to investigate this relationship in greater detail.

Effect of Surfactant on W —In order to determine whether adsorbed surfactant was likely to have any effect on the $\log W$ /concentration curve an experiment was performed using the same concentration of particles in the presence of 2.5×10^{-5} M ammonium lauryl ether sulfate.⁵ This has been shown

⁵ Marketed as "Empicol EAB" by Marchon Products Ltd., Whitehaven, Cumberland, England.

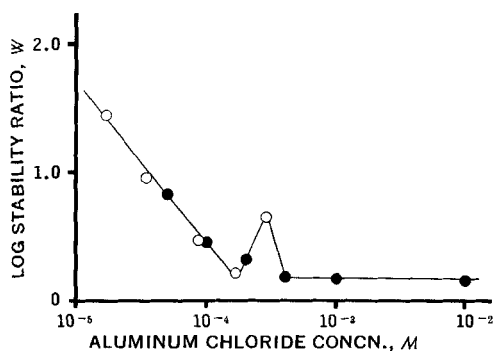


Fig. 6—Stability ratio W , as a function of aluminum chloride concentration for 0.714- μ polystyrene latex containing 2×10^{-5} M ammonium lauryl ether sulfate. —○— and —●— are separate experiments.

(13) to be compatible with aluminum salts, whereas more commonly used anionic surfactants such as sodium lauryl sulfate and sodium dioctyl sulfosuccinate cause precipitation (6, 13). The results of two experiments are shown in Fig. 6. The graph is similar in form to that obtained in the absence of surfactant, but the latter confers some protective action on the particles at low concentrations of elec-

trolyte. The region of partial peptization occurs at about the same concentration of electrolyte and the ultimate value of W_{exp} is identical, 1.5. This indicates that the presence of the surfactant has little effect on coagulation at high electrolyte concentrations and is not the cause of the departure from Smoluchowski theory.

SUMMARY

The use of the Coulter counter and a digital computer has been shown to be eminently suitable for evaluating stability ratios in flocculating monodisperse systems, in the particle size range 0.5–2.0 μ . This is above the limits which most workers would set for true colloidal systems and many powdered drugs are likely to have a considerable proportion of particles in this region. Such monodisperse suspensions have been shown to behave in a similar manner to colloidal systems, yet to exhibit important differences particularly in maximum flocculation rates. Further experiments are planned to evaluate more fully the factors which are likely to be of importance in governing the flocculation behavior of pharmaceutical suspensions. Such information is likely to be of great value in rationalizing the behavior of these systems and providing the pharmacist with a sound theoretical basis for formulation

APPENDIX

Sample of Data Input

| Program Symbol | Data | Key |
|----------------|--|--|
| X | 5 | Number of times at which results were taken |
| B | 400 | Background count on electrolyte |
| N | 20,662 | The initial count less background computed as singlets |
| M | 2 | Number of counts at each time |
| DIL | 125.0 | The dilution factor |
| VOL | 0.05 | The volume counted on the Coulter counter |
| F | 17503 14018 11679 10380 9689 | X rows of M experimental results |
| V (I) | 0.0 0.50 1.00 1.50 2.00 | X times in hours when results were taken |

Computer Program

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Written in Elliot 803 Algol 60
COULTER COUNTER DUAL THRESHOLD ALPHA AND STABILITY RATIO'
BEGIN INTEGER X,B,N,I,P,J,F,M,XM1'
      REAL T,R,W,Z,D,V,SUM,NUM,TAU,PHI,DIL,VOL'
SWITCH S: = L1'
L1: READ X,B,N,M,DIL,VOL'
XM1: = X-1'
T: = (2@8)/(N*DIL*(1/VOL)*3.6)'
BEGIN ARRAY R,W,V(1:X)'
      FOR I: = 1 STEP 1 UNTIL X DO
BEGIN P: = 0' FOR J: = 1 STEP 1 UNTIL M DO
BEGIN READ F' P: = P+F'
END'
      R(I): = ((P/M)-B)/N'
      W(I): = (SQRT(1/R(I))-1)*T'
      Z: = W(I)'
END'
PRINT £ N/NO EXPT N/NO THEO SQNO/N-1T T THEO ?
BEGIN SUM: = 0' TAU: = 0'
      FOR I: = 1 STEP 1 UNTIL X DO
BEGIN READ V(I)'
PRINT FREEPOINT [4], R(I), £ ?, SAMELINE,1/(1+(Z+V(I))/T)**2, £ ?,

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```

W(I), £ ?, (Z+V(I))'
END'
PRINT £
?'
FOR I: = 1 STEP 1 UNTIL XMI DO
BEGIN NUM: = (W(I+1)-W(I))/(V(I+1)-V(I))'PHI: = 1/NUM'SUM: = SUM+NUM'
TAU: = `TAU+PHI'
PRINT FREEPOINT (4), NUM, SAMELINE, PHI'
END'
PRINT £
ALPHA = ?, SAMELINE, FREEPOINT (4), SUM/XMI'
PRINT £
STABILITY RATIO W=?, SAMELINE, FREEPOINT (4), TAU/XMI' PRINT £
?'
END'
END'
GO TO L1'
END'

```

Sample of Print-out

| N/No exp. | N/No theor. | $(\sqrt{N_0/N} - 1)T$ | $T_{\text{theor.}}$ |
|-----------|-------------|-----------------------|---------------------|
| 0.8073 | 0.8073 | 0.1214 | 0.1214 |
| 0.5780 | 0.4016 | 0.3390 | 0.6214 |
| 0.4377 | 0.2395 | 0.5500 | 1.1214 |
| 0.3425 | 0.1589 | 0.7619 | 1.6214 |
| 0.2856 | 0.1131 | 0.9365 | 2.1214 |

Values of α and W Between Successive Times

| α | W |
|------------------------|----------------------------------|
| 0.4353 | 2.297 |
| 0.4217 | 2.371 |
| 0.4240 | 2.359 |
| 0.3491 | 2.864 |
| Mean $\alpha = 0.4075$ | Mean Stability Ratio $W = 2.473$ |

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Keyphrases

Stability ratios—floculating monodisperse systems
 Particle concentration—stability ratios
 Electrolyte concentration—stability ratios
 Particle size—stability ratios
 Surfactant—stability ratios
 Coulter counter—particle counting
 Digital computer—calculation, stability ratios