

Interaction between Particles Suspended in Solutions of Macromolecules

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I. Introduction

Most of the theories which have been presented on forces acting between suspended colloidal particles are concerned with van der Waals' force and a force caused by electric double layers on particles.^{1,2} These theories can explain many phenomena observed in experiments relating to the stability of suspended particles. Forces of other origins, however, can also act between the particles.^{3,4} In this paper we derive an attractive force acting between particles suspended in a solution of macromolecules where there is neither direct interaction between two particles nor between particles and solute macromolecules.⁵ The magnitude of this force is of the order of osmotic pressure of the solution of macromolecules and the range is of the order of diameter of macromolecules. This force becomes strong in solutions of chain macromolecules or dissymmetrical macromolecules and is greatly intensified by giving electric charges to the macromolecules. In such a case, a macroscopic aggregation of suspended particles can take place. We give some numerical examples of the critical concentration at the macroscopic aggregation. Even a minute change in the condition of solute macromolecules can completely change the state of particles. Direct experimental proof of the reality of the force derived here has not yet been obtained. Some phenomena, however, suggest that this force actually acts between suspended particles.

II. Interaction in Solution of Spherical Macromolecules

Let us consider two large parallel plates immersed in a solution of rigid spherical macromolecules. When the distance between the inner surfaces of these two plates is smaller than the diameter of solute macromolecules, none of these molecules can enter the space between the plates and this space becomes a phase of the pure solvent. Therefore, a force equivalent to the osmotic pressure of the solution of macromolecules acts on the outer surfaces of these plates. Such a force also appears between two spherical particles suspended in this solution.

When the concentration of macromolecules is sufficiently low, the force P between two particles suspended in the solution is given by⁵

$$P = kTN \partial \ln Q / \partial a \quad (1.1)$$

$$Q = \int_V \exp \{-w(x,a)/kT\} dx \quad (1.2)$$

where N denotes the total number of solute macromolecules, V the total volume of the solution, and a the distance between the centers of two particles. The quantity $w(x,a)$ is the potential energy of a macromolecule situated at x generated by its interaction with the suspended particles or, more rigorously, w is the average free energy of a macromolecule at x when two particles are fixed at distance a . The negative value of P corresponds to the attraction between particles.

If macromolecules are rigid spheres with no interaction between them and suspended particles, then Q is equal to the volume of space in which each macromolecule can move freely. In other words, we can obtain Q by subtracting from the total volume of the solution the volume into which the centers of macromolecules cannot enter owing to hindrance by the particles. When particles are also spherical, we have

$$\begin{aligned} Q &= V - \pi(4/3)[((D+d)/2)^3 + (D+d)^2a - (1/12)a^3] \\ & \qquad \qquad \qquad D \leq a \leq D+d \\ &= V - (8\pi/3)((D+d)/2)^3 \qquad \qquad D+d \leq a \end{aligned} \quad (2)$$

where D and d are diameters of suspended particles and solute macromolecules, respectively. Consequently, we obtain

$$P = -p_0S \quad (3.1)$$

where

$$\begin{aligned} S &= (\pi/4)D'^2 = (\pi/4)[(D+d)^2 - a^2] \qquad D \leq a \leq D+d \\ &= 0 \qquad \qquad \qquad D+d \leq a \end{aligned} \quad (3.2)$$

and

$$p_0 = kTN/V \quad (3.3)$$

When two particles approach each other, two spherical volumes of diameter $D+d$ which exclude macromolecules begin to overlap at mutual distance a equal to $D+d$. With further decrease of a the volume of the overlapping region increases and therefore the attraction acts between particles. The quantity S in (3.1) is an area of the circular cross section of the overlapping region at the middle of a , and p_0 is the osmotic pressure of the solution of macromolecules. Since the interaction between macromolecules was neglected in deriving the above result, p_0 was expressed by van't Hoff's formula.

The physical interpretation of the simple result (3) can be obtained easily. Except on the part of the surface facing the phase of the pure solvent of cross section area S between two particles, the osmotic pressure of the solution acts uniformly from every side on the surface of suspended

particles. The total force acting on the outside half of one of the two particles is larger by Sp_0 than the force acting on its inside half. Thus we can expect that the result (3.1) may be applicable even when the concentration of macromolecules is so high that the osmotic pressure cannot be expressed by van't Hoff's formula. Especially when the diameter D of particles is very much larger than the diameter d of macromolecules and therefore the phase of the pure solvent between particles can be defined clearly, (3.1) can give the force almost exactly.

The potential energy U of this force P is written

$$\begin{aligned} U(a) &= -p_0(\pi/12)[2(D+d)^3 - 3(D+d)^2a + a^3] \quad D \leq a \leq D+d \\ &= 0 \quad D+d \leq a \quad (4) \end{aligned}$$

If $D \gg d$, (4) can be approximated by

$$U(a)/kT = -(3/2)\phi\beta x^2 \quad 0 \leq x \leq 1 \quad (5)$$

where $x = (D+d-a)/d$, $\beta = D/d$, and $\phi = (\pi/6)d^3N/V$; here ϕ means the volume concentration of macromolecules. At $x = 1$ or $a = D$, U takes the minimum value $-(3/2)kT\beta\phi (= U_m)$.

For example, if $\beta = 50$ and $\phi = 10^{-2}$, then $U_m/kT = -0.75$. The attractive potential between particles is of long range, but usually its absolute value is not very much larger than kT in dilute solutions of rigid spherical macromolecules.

III. Interaction in Solutions of Chain Macromolecules and of Dissymmetrical Macromolecules

Calculations have heretofore been made for particles in solutions of rigid spherical macromolecules. A similar method of calculation can be applied to particles in solutions of coiled chain macromolecules. In this case also, the essential feature of the relation between the potential U of particles and their mutual distance a is not altered, if we regard ϕ as the apparent volume concentration of coiled macromolecules and d as their apparent diameter.⁵ It is remarkable that in solutions of chain macromolecules the apparent volume concentration can be large in very small net concentration. The aggregates are formed with comparative ease in solutions of chain macromolecules.

Assuming that the square of diameter d of chain macromolecules is proportional to their polymerization degree or molecular weight, we can prove from (5) that as the molecular weight of chain macromolecules is increased at a constant weight concentration, the range of force between suspended particles is increased and the value of U_m is kept nearly constant. Therefore, with increasing molecular weight we can expect an increasing number of aggregates.

Now, let us consider a solution of rigid ellipsoidal macromolecules of length A and diameter B . By the same method as in the previous section, we can obtain the potential energy U between spherical particles suspended

in this solution if we take into account the limited freedom of orientation of ellipsoidal macromolecules near particles. When D is very much larger than A and B , the result for $A/B(=q) \geq 1$ is given by

$$-U(a)/kT = (3/2)\beta\phi[(1/3)q^2 + (2/3) - q(q^2 - 1)^{-1/2}(1 - x) \ln (q + (q^2 - 1)^{1/2}) - (1 - x)x] \quad (6)$$

for $x = (D + A - a)/A$ between 1 and $1 - q^{-1}$; here ϕ is the volume concentration of ellipsoidal macromolecules and $\beta = D/A$. The formula for $0 < x < 1 - q^{-1}$ is not written here because it is too long. The result for $A/B < 1$ is obtained by replacing $\ln (q + (q^2 - 1)^{1/2})$ in (6) by $((\pi/2) - \sin^{-1}q)$ and x in (6) by $x' = (D + B - a)/B$. The value of U at $a = D$, namely, when two particles are in contact with each other, is written

$$\begin{aligned} -U_m/kT &= (3/2)\beta\phi((1/3)q^2 + (2/3)) \\ &= (3/2)\phi D(4\pi/3)^{1/3}v^{-1/3}[(1/3)q^{4/3} + (2/3)q^{-2/3}] \quad (7) \end{aligned}$$

over the whole range of q ; here v is the volume of the macromolecule. At a constant concentration and a constant volume v , $-U_m$ takes a minimum value when $q = 1$. With increasing dissymmetry of solute macromolecules, both the range of force and the absolute value of potential energy are increased. The ratio of $-U_m$ in a solution of ellipsoidal macromolecules to $-U_m (= -U_m^0)$ in a solution of spherical macromolecules of the same volume at the same concentration is given by $[(1/3)q^{4/3} + (2/3)q^{-2/3}]$. For example, the value of this ratio U_m/U_m^0 becomes 3 at $q = 5$ and 18 at $q = 20$. When $\phi = 10^{-3}$, $\beta = 6$, and $q = 20$, $-U_m/kT = 1.2$. Thus, in a solution of dissymmetrical (for example, rodlike) macromolecules, a larger number of aggregates of suspended particles can be formed than in solutions of spherical macromolecules of the same volume.

IV. Interaction in Solutions of Charged Macromolecules

The osmotic pressure p of solutions of charged macromolecules is very much larger than the osmotic pressure p_0 of solutions of noncharged macromolecules of the same concentration. The ratio $p/p_0 (=f)$ is equal to the number of free counter ions per macromolecule when the solution contains no low molecular salts. When low molecular salts are added, f is decreased but is still somewhat larger than unity. If two parallel and infinitely large plates be immersed in such a solution of charged macromolecules, the attractive force P between the plates would be f -times as large as the force P_0 in a solution of noncharged macromolecules of the same concentration. When two spherical particles are immersed, the ratio of the forces $P/P_0 (=f')$ does not become f because some counter ions can enter the narrow region between two particles. Nevertheless, this ratio is larger than unity. It is difficult to obtain the exact expression of P , but the approximate value of P can be estimated as follows.

Let us consider spherical particles of diameter D suspended in a solution of charged spherical macromolecules of diameter d which is very much

smaller than D . When two particles approach each other, there appears a region between particles into which the macromolecules are forbidden to enter. This region has a circular cross section area S with diameter $D' = [(D + d)^2 - a^2]^{1/2}$. Some counter ions entering this region are attracted by macromolecules near the boundary of the region and are drawn outward. Consequently, they are moving near the circumference of the region. Few counter ions are in the central part of the region. The thickness of the circumference layer wherein counter ions are moving is of the order of the Debye-Hueckel ionic atmosphere radius $D''/2$ which is determined by the molar concentration of free counter ions in the solution: With decreasing distance a between particles, D' becomes larger than $D'' + d$ and the region containing few counter ions begins to appear. Then the force due to osmotic pressure of counter ions begins to act between the particles. When D' is sufficiently larger than $D'' + d$, the force P can be estimated by the equation

$$P = -p(c)S[(D' - (D'' + d))/D']^2 \quad (8)$$

where $p(c)$ is the osmotic pressure of the solution of charged macromolecules of concentration c . By integrating this force, if $D \gg d$, we have

$$U(a)/kT = (U_m^0/kT)f[x^2 + [(D'' + d)^2/Dd]x - (4/3)((D'' + d)/d)(2d/D)^{1/2}x^{3/2} - (1/3)((D'' + d)^2/2dD)^2] \quad (9)$$

where $U_m^0/kT = -[p_0(c)/kT](\pi/4) = -[p(c)/kT](\pi/4)(1/f)$, and U_m^0 is the potential energy of particles at $x = (D + d - a)/d = 1$ in a solution of noncharged macromolecules at the same concentration c .

We put $D = 5000A$ and $d = 100A$. When two particles are in contact with each other, $D' = 1000A$, and when the distance between the surfaces of two particles is $50A$, $D' = 700A$. We assume that each macromolecule has 200 ionized groups and that there is no added low molecular salts. Most of the counter ions are bound by macromolecules and, in ordinary solutions of charged macromolecules without added salts, the osmotic coefficient is of the order of 0.1.^{6,7} Therefore, in the above numerical example we can put $f = 20$. When the molar concentration of ionized groups in the solution is between 10^{-1} and 10^{-3} , the molar concentration of free counter ions is between 10^{-2} and 10^{-4} . Then the thickness $D''/2$ of the layer of counter ions entering the region between two approaching particles is between $40A$ and $400A$. (We can expect that $D''/2$ is a little less than the ionic atmosphere radius determined by the molar concentration of free counter ions estimated from the osmotic coefficient or the activity coefficient of counter ions.) Hence, we can see that D' is larger than $D'' + d$ unless x is too small. From (9) it is found that in the above example, U/kT at $x = 1$, U_m/kT , is of the order of $10(U_m^0/kT)$ to $3(U_m^0/kT)$. Thus the force between suspended particles is intensified by giving charges to macromolecules. The magnitude of the force is changed by changes in the charge of macromolecules and in concentrations of macromolecules

and salts. By addition of low molecular salts, D'' is slowly decreased and f is more rapidly decreased; consequently, $-U/kT$ is decreased.

V. Aggregation of Suspended Particles

When a large number of particles are suspended in the solution of macromolecules, they are attracted to each other through the negative potential U , and some of them form aggregates. In ordinary solutions of non-charged rigid spherical macromolecules, however, the number of these aggregates is not very large. From (5), the ratio of the number of pairs of two particles M_2 to the number of dispersed particles M_1 is calculated by

$$M_2/M_1 = 12\Phi(K/\beta) \quad (10.1)$$

$$K = (1/t_0) \int_0^{t_0} \exp \{t^2\} dt \quad (10.2)$$

where $t_0^2 = -U_m/kT$ and $\Phi (= (\pi/6)(D^3M_1/V))$ is the volume concentration of dispersed particles. Two particles at a distance less than $a = D + d$ are regarded as a pair. In the numerical example of Section II where $U_m/kT = -0.75$, M_2/M_1 becomes $(1/4)\Phi$. This ratio M_2/M_1 increases rapidly with increasing $-U_m/kT$. Therefore, the numerical estimation in Sections III and IV suggests that in solutions of chain or dissymmetrical macromolecules and especially in solutions of charged macromolecules, a large number of aggregates can be formed and even a macroscopic aggregation can occur.

We calculate the critical (volume) concentration Φ^* of suspended particles where the macroscopic aggregation takes place. It is assumed that in the macroscopic phase of aggregation each particle has 12 neighboring particles and that the interaction energy between two neighboring particles can be given by (5), (6), or (9). When $D \gg d$, the interstices between particles in the macroscopic aggregate are large enough for macromolecules to enter and, as will be shown later, the average distance $a_m(x_m)$ between neighboring particles in the aggregate satisfies the condition $1 \gg 1 - x_m > 0$. Accordingly, we can use the above assumption for the interaction energy between particles in the aggregate.

At constant values of the average distance a between neighboring particles, we can obtain the partition function z per particle composing the aggregate by using a method similar to the free volume method in the theory of liquid;^{8,9} namely, where z is given by

$$z(a) = \int_0^{(a-D)/2} \exp \{-U_a(a')/kT\} 4a'^2 da' \quad (11)$$

Energy $U_a(a')$ is the average potential energy of a particle surrounded by 12 neighboring particles when the position of the surrounded particle deviates from the center of the free volume by distance a' . When the concentration of macromolecules and other conditions of the medium are given, the average value of a , a_m , in the macroscopic aggregate is determined by the maximum condition of $z(a)$ as a function of a . The value of z at this maximum, z_m , gives the activity of a particle in the macroscopic

aggregate. The volume concentration Φ^* of dispersed particles in equilibrium with this macroscopic aggregate is given by

$$\Phi^*/(\pi D^3/6) = M_1/V = z_m^{-1} \quad (12)$$

Some examples of the results of numerical calculation are shown in Table I.

TABLE I
Critical Volume Concentration of Suspended Particles at Macroscopic Aggregation at Various Values of Interaction Energy

D/d	$-U_m/kT$	$\phi(U_m/U_m^0)$	$1 - x_m$	Φ^*
50	2.16	2.9×10^{-2}	0.054	6.9×10^{-2}
50	2.91	3.9×10^{-2}	0.046	1.9×10^{-5}
50	3.21	4.3×10^{-2}	0.041	8.4×10^{-7}

In a solution containing suspended particles of volume concentration 10^{-5} , when the value of U_m/kT is changed from -2.2 to -3.2 , the state of particles is changed from complete dispersion to complete aggregation. When the volume concentration of macromolecules is 10^{-2} , this change of U_m/kT corresponds to the increase of U_m/U_m^0 from about 3 to about 4. In real solutions of charged macromolecules, such an increase of U_m/U_m^0 is easily brought about by increasing the activity coefficient of counter ions or by increasing the charge of macromolecules. Inversely, a decrease of charge, a decrease of activity coefficient of counter ions, or an increase of concentration of added salts can bring about the dispersion of aggregated particles.

From Table I and the result obtained in Section III, it is found that in solutions of macromolecules having a great dissymmetry the macroscopic aggregation of suspended particles can actually take place and the state of particles is strongly affected by the change of shape of solute macromolecules. Chain macromolecules can change their shape with the change of composition of solvents. When they have charge, extended linear macromolecules in a solution without salts are contracted to a coiled spherical shape by the addition of a small amount of salts.

With decreasing radius of suspended particles, the critical volume concentration of these particles at macroscopic aggregation is rapidly decreased.

The number of M_i of aggregates composed of i particles is calculated by

$$M_3/M_1 = 36\Phi^2(K/\beta)^3 \quad (13.1)$$

$$M_i/M_3 = 6H^{i-3}/i(i-1)(i-2) \quad 4 \leq i < 10 \quad (13.2)$$

$$H = (6 \cdot 2^{1/2}/\pi)\Phi(K/\beta)^3 \quad (13.3)$$

where K is given in (10.2) and the interaction energy between two neighboring particles is assumed to be written in the form of (5). When the macroscopic aggregation occurs, H becomes larger than unity. When the volume concentration of suspended particles is 10^{-4} , the value of (K/β)

at $H = 1$ is about 15. In this case, $M_2/M_1 = 0.02$ and $M_3/M_2 = 0.07$. This means that at macroscopic aggregation, the number of aggregates composed of i particles takes a minimum at $i = 3$ as a function of i . Such a minimum is found in most cases of the condensation phenomena.

VI. Discussion

We have assumed that there is no direct interaction between particles and solute macromolecules, and that the particles themselves have no electric charge. In reality, we have no such ideal example, but we have examples in which suspended particles are aggregated by addition of macromolecules. Chain macromolecules are more effective than rigid macromolecules. Aggregation of red blood cells can be brought about non-specifically by adding to the suspension chain macromolecules or highly dissymmetrical macromolecules.¹⁰⁻¹² According to experimental results,¹² many electrolytic and nonelectrolytic macromolecules have the ability to make aggregates of blood cells. The high molecular weight and the dissymmetrical shape of molecules were found to be most important factors.^{11,12} As the degree of polymerization of chain macromolecules increases, the aggregation effect becomes stronger. Monomer or polymer of a low polymerization degree cannot bring about the aggregation. Most of the rigid spherical macromolecules, for instance, globular protein molecules, have no ability to make aggregates of the cells. Even when they have the ability to aggregate, a very high concentration of macromolecules is necessary for the occurrence of aggregation. As another example, it is well known that aggregates of rubber latex particles can be formed by adding various chain macromolecules, including fibrous proteins and linear polyelectrolyte molecules.^{13,14} Soil particles can also be aggregated by chain macromolecules.¹⁵ Rigid spherical macromolecule cannot form aggregations of these particles. In most of these nonspecific aggregation phenomena, there are found only few macromolecules adsorbed on particles and therefore, in current opinion, the aggregation has been considered to be caused by the dehydration of particles by hydrophilic macromolecules. Such explanation, however, cannot satisfy us because the dehydration is an ambiguous concept. We want to emphasize that the qualitative features of these phenomena can be reasonably explained by our theory. The osmotic force derived in this paper probably takes an important role in these phenomena.

Of course, the interaction between suspended particles and solute macromolecules cannot always be disregarded.^{12,16,17} In some cases, macromolecules are remarkably adsorbed on particles and form bridges between them. As a result, a gel-like aggregation can occur. We can easily develop a general theory based on eq. (1), of the force between particles mediated by solute molecules and investigate the effect of energetic interaction between particles and solute molecules. Discussions on this kind of force were made by Tezak and others.⁴ We describe here some results of our calculation. When the interaction energy u between particles and

solute molecules is positive, namely when the interaction is repulsive, the interaction energy U between particles is always negative and an attraction appears. When u is negative, U as a function of distance a depends on the magnitude and range of u in a complex way. When two particles approach each other very closely, adsorbed molecules must be detached from the surface of particles and therefore, a repulsion usually appears between particles. If, however, molecules sandwiched between two approaching particles can interact with both particles and, for example, have negative energy $2u$, then an attraction can appear between particles and U is negative when a is not too small for molecules to enter the narrow region between particles. This attraction which is caused by bridging molecules becomes stronger with increasing concentration of solute molecules and takes a maximum value at the concentration where the surface of independent particles is not yet saturated by adsorbed molecules and the region between two approaching particles is saturated by them. At higher concentrations, where the surface of independent particles is also saturated, the attraction is weakened. Under suitable conditions of the medium, U as a function of a can have two minimum points. The one is caused by the bridging effect of solute molecules and the other taken at $a = D$ is caused by the osmotic effect. Between these two minimum there is a maximum caused by the detachment of adsorbed molecules. The details of this calculation will be reported elsewhere.

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Synopsis

An attractive force appears between particles suspended in solutions of macromolecules when there is neither direct interaction between two particles nor energetic interaction between particles and solute macromolecules. The magnitude of this force is of

the order of the osmotic pressure of the solution of macromolecules and the range is of the order of the diameter of macromolecules. This force is calculated as a function of concentration, shape, and charge of macromolecules, and it is shown that it becomes stronger in solutions of chain macromolecules or of macromolecules of dissymmetrical shape than in solutions of rigid spherical macromolecules at the same net concentration. If macromolecules have charge, the force can be greatly intensified. In every case numerical estimation is made, and it is found that actually this kind of force can have a remarkable influence on the state of suspended particles. Numerical examples of the critical concentration of particles at their macroscopic aggregation are given. Finally a short description is added on the effect of energetic interaction between particles and macromolecules.

Résumé

Il existe une force d'attraction entre les particules en suspens dans des solutions de polymères, lorsqu'il n'y a aucune interaction directe entre les particules elles-mêmes ni entre les particules et les macromolécules dissoutes. La grandeur de cette force est de l'ordre de la pression osmotique de la solution polymérique, et leur domaine est de l'ordre du diamètre des macromolécules. Cette force est calculée en fonction de la concentration, la forme et la charge des macromolécules, et on montre qu'elle devient plus forte dans des solutions de macromolécules linéaires ou de macromolécules de forme dissymétrique, que dans des solutions de macromolécules sphériques rigides pour une même concentration. Si les macromolécules sont chargées, la force peut être considérablement accrue. Dans chaque cas une estimation numérique a été faite, et on trouve qu'actuellement cette sorte de force cause une influence notoire sur l'état de particules en suspens. Des exemples numériques de la concentration critique de particules à l'état d'aggrégation macroscopique sont indiquées. Finalement on donne également une brève description de l'effet d'interaction énergétique entre les particules et les macromolécules.

Zusammenfassung

Zwischen Teilchen, die in einer Lösung von Makromolekeln suspendiert sind, tritt eine Anziehungskraft auf, auch wenn weder eine direkte Wechselwirkung zwischen zwei Teilchen noch eine energetische Wechselwirkung zwischen den Teilchen und den gelösten Makromolekeln besteht. Die Stärke dieser Kraft ist von der Grössenordnung des osmotischen Druckes der Lösung der Makromolekel und ihre Reichweite von der Grössenordnung des Durchmessers der Makromolekel. Diese Kraft wird als Funktion der Konzentration, Gestalt und Ladung der Makromolekel berechnet und es wird gezeigt, dass sie in Lösungen von kettenförmigen Makromolekeln oder von Makromolekeln von unsymmetrischer Gestalt grösser wird als in Lösungen starrer, kugelförmiger Makromolekel bei derselben Nettokonzentration. Wenn Makromolekel Ladungen tragen, kann sich die Kraft sehr verstärken. Für jeden betrachteten Fall wird eine zahlenmässige Abschätzung durchgeführt und es stellt sich heraus, dass tatsächlich eine Kraft von dieser Art einen bemerkenswerten Einfluss auf den Zustand der suspendierten Partikel haben kann. Für die kritische Konzentration von Teilchen bei ihrer makroskopischen Aggregation werden zahlenmässige Beispiele gegeben. Schliesslich wird noch zusätzlich eine kurze Beschreibung der Auswirkung der energetischen Wechselwirkung zwischen Teilchen und Makromolekeln gegeben.

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