

Viscosity of Double Diffuse Layer

by

R. P. Kulkarni*

Introduction

The structure of the double diffuse layer surrounding clay particles is supposed to be either 'Solid' or 'Two dimensional'. According to either model, the bond between water molecules of this layer is stronger than that of free water. In case of 'rigid' model the resistance to shear as well as to normal stress, offered by the water in the form of double diffuse layer is supposed to be significant whereas in the 'two dimensional' model, the bond between water molecules is believed to offer resistance only to the normal stress and the resistance against shear stress is equal to or smaller than that offered by water molecules in free water. Rosenqvist (1955) favours the 'rigid' model, Lambe (1958), Micheals (1959), Martin (1962) believe the 'two-dimensional' model to be more realistic. Martin (1962) had opined that at present there is no evidence available which is in disfavour to any of the above two models that describe the structure of the double diffuse layer surrounding clay particles.

Rosenqvist (1955) had tried to find indirectly, from the diffusion coefficient of water in a clay water system, the average viscosity of the double layer water of Aserum clay at different water contents. The magnitude of viscosity obtained by him varied from 240 to 1530 millipoises when water content of the soil was decreased from 30 to 10 percent.

Bready and Brown (1954), Davies and Rideal (1961) had shown that the viscosity of monolayers of liquid is very high (10^4 to 10^5 poises) due to surface orientation.

In order to determine the viscosity of the double diffuse layer around clay particles by as direct a method as possible use is made of the information giving relationship between the viscosity of suspension and that of suspending medium.

Literature Study

A suspension is composed of two phases, solid and liquid. The solute particles are solid and suspending medium is liquid. Solute particles do not react chemically with the suspending medium. Nevertheless, a double diffuse layer is formed around the solute particle if conditions are favourable. In case of clay-water suspensions, clay particles are solute and

* Assistant Research Officer, Maharashtra Engineering Research Institute, Nasik-422004.

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water is the suspending medium. A double layer is formed around clay particles in suspension.

It is observed that the viscosity of the suspension is more than that of the suspending medium. The viscosity of suspension depends upon the characteristics of the solute, e.g. their shape, size, formation of double layer around them or otherwise, their concentration etc. It was further observed that for the same solute concentrations, the viscosity of the suspension with solute particles of spherical shape is smaller than that of the suspension with solute particles of anisodimensional shape. Different hypotheses are proposed to explain the higher magnitude of the viscosity of the suspension with elongated solute particles in comparison to that of the suspension with spherical particles of same concentration. In addition, there are a number of equations developed relating the viscosity of suspension and that of suspending medium.

Equations relating Viscosity of Suspension and of Suspending Medium :

Arrhenius (1887), Einstein (1906), Waltmann & Green (1943) had found out different relationships.

A relationship between viscosity of suspension and volume fraction of solids, s , applicable to any volume of s , was developed by Robinson (1949) as :

$$\frac{\mu_s - \mu_L}{\mu_L} = \frac{xs}{1 - ys}$$

Where 'x' is the fraction component and 'y' is the relative sediment volume. The value of 'y' depends upon the 'packed volume' of the suspended solid. μ_s and μ_L are the viscosities of the suspension and suspending medium respectively.

Crawley and Kitzes (1957) had used Robinson's equation in their theory of viscosity of highly concentrated thoria slurries and selected Einstein's value of 2.5 for 'x'.

Kearsey (1962) extended Crawley and Kitzes' (1957) work further for non-Newtonian suspensions containing nonspherical particles. In the experimental work done by Kearsey, concentration of thoria in the suspension was varied from 500 to 1500 gms of thoria per litre of water. He found that the value of 'x' is 6.1 ± 0.1 if Robinson's (1949) equation (5) is applied to these thoria slurries. The increase in the value of 'x' from 2.5 to 6.0 is because of the elongated shape of thoria particles. Alexander and Johnson (1949) had shown that the value of 'x' is not more than 6.0 even if the axial ratio of solute particles is as large as 113.

Kearsey (1962) had further shown that Robinson's equation is applicable to flocculated thoria slurries i.e. to non-Newtonian suspensions.

From the above given literature review, it may be inferred that Robinson's equation is applicable to :

- (i) a suspension of any solid concentration ;
- (ii) a suspension containing solute of anisodimensional shape ;

- (iii) a suspension with non-Newtonian behaviour ; and
- (iv) the value of 'x' is not more than 6.0 for elongated particles.

Clay soil-water suspensions behave like non-Newtonian fluids [Bingham (1922), Terzaghi (1931), Houser and Le Beau (1941) Norton et al (1944), Gueze and Tan (1954), Murayama and Shibata (1961), Howard (1963)], unless the concentration of solute particles is very small. Clay particles are anisodimensional with axial ratio less than 100 even for montmorillonite clay particles, Lambe (1948). It was found that because of these two peculiarities of clay water suspension, Robinson's equation is more applicable, in comparison to others, to relate the viscosity of clay water suspension and of suspending medium.

Increase in the Viscosity of Suspension because of Solute Particles of Elongated Shape :

The different reasons to explain the increase in viscosity of the suspension with anisodimensional solute particles above that of the suspension containing spherical solute particles of same concentration are (McBain and McBain 1937)

- (1) Hydration of particles;
- (2) Electro-viscous effect;
- (3) Influence of the axis ratio; and
- (4) Structural viscosity.

It is observed that the viscosity of suspension with anisodimensional particles as solute could be quite high although the concentration of solute was so small that structural viscosity as described above could not be developed. Houser and Reed (1936) had shown that as little as 0.05 percent bentonite in water is sufficient to develop a viscous suspension which stops the rise of air bubbles to the surface. Norton et al (1944) has shown that the viscosity of suspension increases considerably with 0.005 solid concentration of kaolinite. The concentration of solute in these cases was so small that particle-particle contact or ramming of particles is not possible, which is necessary to develop structural viscosity as given by McBain and McBain (1937). A modification to the above hypothesis to explain the increase in the viscosity of the suspension with anisodimensional solute more than that of the suspension with spherical particles is given below.

Let it be assumed that in a clay-water suspension, the concentration of clay particles is so small that even after complete development of double diffuse layer surrounding them they are not in contact with each other but that there is free water present between them (Figure 1A). In this case the viscosity of the suspension with elongated particles would be more than that of the suspension with spherical particles of same concentration because of the first three reasons given above. When the concentration of solute is increased to such an extent that double layers of adjoining particles interpenetrate each other (Figure 1B) then the viscosity of the suspending medium would increase and would be average of the viscosity of double layers at contact with each other. This is because the viscosity of the double layer water is more than that of free water and

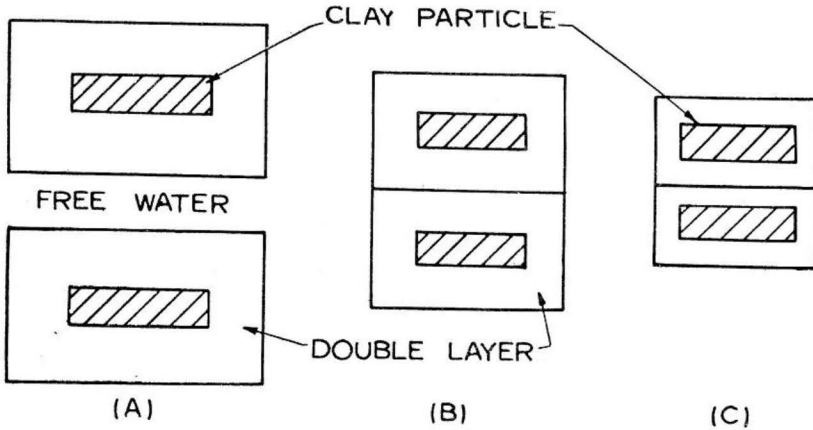


FIGURE 1. Interpretation of Double Layers.

it increases with decreasing distance from the particle surface, Terzaghi (1931), Rosenqvist (1955), Davies and Rideal (1961). With further increase in the concentration of solute, particles some nearer to each other with consequent increase in the interpenetration of double layers of adjoining clay particles; which in their turn increase the viscosity of the suspending medium. The suspending medium is not free water in this case but it is in the form of double layers which are in contact with each other at various amount of interpenetration. The viscosity of the suspending medium is, therefore, the average of that at contact of double layers, which, increases with the increase in the concentration of solute. It is, therefore, hypothesised that the viscosity of the suspension with elongated particles as solute is more than that of suspension with equivalent concentration of spherical particles because of the increase in the viscosity of the suspending medium itself due to the formation of interpenetrating large double layers around anisodimensional particles, which are electrically charged.

Experimental Evidence

As an evidence in support of this hypothesis, viscosity observations of *AgI* sol as given by Harmsen, Schooten and Overbeck (1953) are considered. *AgI* particles are elongated in shape and have electrically charged surfaces. The data given by these authors is compiled in Table 1-A and 1-B.

TABLE 1-A
Viscosity data of suspending medium of *AgI* sol

No.	CH^+ $Mg\ ion/l$	Thickness of double layer	Viscosity (μ_L) centipoises
1	0.035	500	0.8937
2	0.12	278	0.8937
3	2.3	64	0.8938
4	15.0	25	0.8943
5	39.0	16	0.8954

TABLE 1—B
Viscosity data of AgI sol suspension

1		2		3		4		5	
<i>S</i>	μ s	<i>S</i>	μ s	<i>S</i>	μ s	<i>S</i>	μ s	<i>S</i>	μ s
0.107	0.896	0.111	0.896	0.101	0.897	0.075	0.8981	0.037	0.898
0.209	0.901	0.215	0.901	0.206	0.8999	0.162	0.9015	0.103	0.901
0.411	0.907	0.418	0.908	0.382	0.9063	0.312	0.9059	0.225	0.905
0.797	0.925	0.830	0.9268	0.773	0.9188	0.678	0.9170	0.476	0.913
1.58	0.966	1.61	0.968	1.59	0.9460	1.34	0.9380	1.09	0.933
2.99	1.10	4.85	1.311	3.04	1.0028	2.69	0.9855	2.11	0.9654
6.37	1.889	7.08	1.761	4.54	1.0698	4.27	1.045	2.95	0.996
				6.53	1.179	6.35	1.136	4.87	1.068

VISCOSITY OF DOUBLE LAYER

The magnitudes of viscosity of suspending medium with different electrolyte concentrations ranging from 0.035 to 39.0 mmoles/litre are given in Table 1-A. Data regarding thickness of double layer for each electrolyte concentration is presented in the same table. The values of the viscosity of suspensions with different volume concentrations of the *AgI* soil, ranging from 0.1 to 7.5 volume percent and for various electrolyte concentrations is given in Table 1-B.

Robinson's (1949) equation was applied to relate the viscosity of the suspension and of the suspending medium. The values of constants x and y for suspensions of different electrolyte concentrations are given in Table 2. For the determination of these constants, value of the viscosity of suspending medium with different electrolyte concentrations as given in Table 1-A are used. It may be noted that magnitude of x decreases from 3.71 to 3.34 with the increase in the electrolyte concentration from 0.035 to 2.3 mmoles/litre. When the electrolyte concentration was increased to 15 and 39 mmoles/litre, Einstein's equation could be applied with the value of x modified to 4.06 and 4.43 respectively.

TABLE 2
Values of constants x and y for *AgI sol*.

	Electrolyte concentration mgm/litre	Parameters of Robinson's (1949) equation	
		x	y
1.	0.035	3.71	14
2.	0.12	3.64	9
3.	2.3	3.34	6
4.	15.0	4.06	0
5.	39.0	4.43	0

For the first three electrolyte concentration, the double layers surrounding *AgI sol* were in contact with each other. With the increase in the electrolyte concentration the thickness of the double layer decreased with consequent decrease in the amount of interpenetration of double layers of adjoining particles. This led to the decrease in the viscosity of the double layers at the boundary of contacts with consequent decrease in the viscosity of the suspending medium. This led to the decrease in the value of constants x and y with the increase in the electrolyte concentration. On the other hand at later two concentrations of electrolyte, namely at 15.0 and 39.0 mmoles/litre, the thickness of the double layers decreased to such an extent that with given concentration of *AgI sol*, there was no contact between double layers of adjoining particles but free water was present between them and hence Einstein's equation was applicable. With the increase in the electrolyte concentration from 15.0 to 39.0 mmoles/litre the thickness of the double layer decreased with consequent increase in the axis ratio* (length/dia) of particles. This led to the increase in the value of x .

*The axis ratio of particles that effect the value of x includes thickness of the double layer around particles (Crawley and Kitzes, 1957, 1958, Kearsy, 1962)

Kearsey (1962) had observed that magnitude of constant x to be used in Robinson's equation could be 2.5 for *AgI sol* if the shape of the solute particles becomes ellipsoidal because of the presence of double layer. The values of x as given in Table 2 are more than 2.5, possibly because the magnitude of the viscosity of the suspending medium considered for calculations was not correct. The actual value of the suspending medium (denoted as μ_{L1}) could be found by assuming value of x to be 2.5. The magnitude of the viscosity of the suspending medium would change depending upon the electrolyte concentration and solute concentration of the suspension. The magnitude of viscosity of suspending medium (μ_{L1}) for suspensions with electrolyte concentration of 0.035, 0.12 and 2.3 mmoles/litre and also for different solute concentrations are illustrated in Figure 2. It may be noted that for a given electrolyte concentration the viscosity of suspending

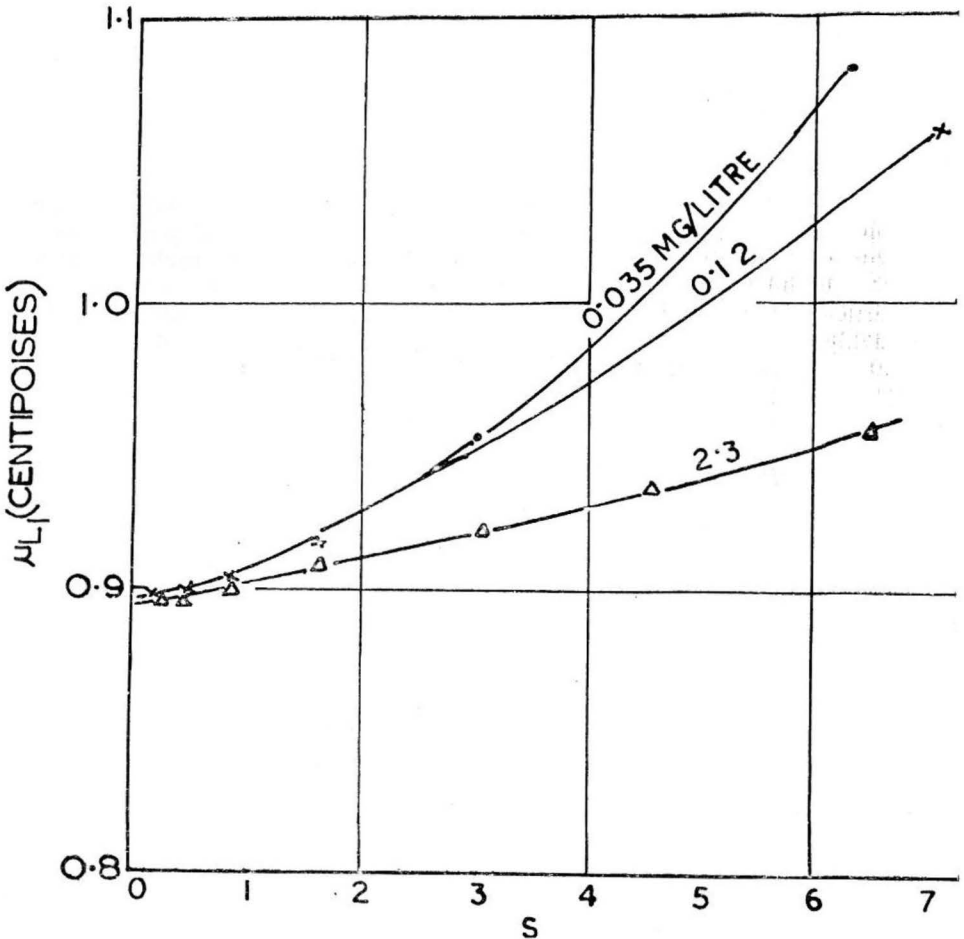


FIGURE 2. Relationship between solute concentration (*AgI sol*) and viscosity of suspending mediums

medium increases with the increase in the solute concentration. Further, for a given solute concentration the viscosity of the suspending medium decreases with the increase in the electrolyte concentration.

TABLE 3
Calculated values of viscosity of suspending medium of AgI sol

1		2		3	
S	μL_1	S	μL_1	S	μL_1
0.107	0.8952	0.111	0.8952	0.101	0.8950
0.209	0.8966	0.215	0.8966	0.206	0.8956
0.411	0.8980	0.418	0.8980	0.382	0.8958
0.797	0.9042	0.830	0.9042	0.773	0.9007
1.58	0.916	1.61	0.9158	1.59	0.9072
2.99	0.9523	4.85	0.9938	3.04	0.9189
6.37	1.081	7.08	1.057	4.54	0.9337
				6.53	0.9526

Another set of test results given by Hauser and Le Beau (1941) is now considered. They had determined viscosity of bentonite clay-water suspensions with different concentrations of clay particles each of which was of single size. The first type clay-water suspensions was having particles of 96μ size. The second and third type of suspensions were having particle sizes of 60 and 20μ respectively. Clay-water suspensions of each type with different concentrations of solute were tested. The test data is given in Table 4. Table 5 presents this information in

TABLE 4
Viscosity of Bentonite-water suspension (Hauser and Le Beau, 1941)

Apperent Particle diameter (μ)	Concentration percent dry wt. (%)	Viscosity at 25°C (μ_s) centipoises
96	0.372	1.12
96	0.516	1.25
96	0.742	1.55
60	0.340	1.19
60	0.525	1.44
60	0.713	1.65
20	0.424	1.23
20	0.612	1.49
20	0.800	1.91

Note :—Viscosity was measured by Hoesppler Viscosimeter

TABLE 5
Data relating solid concentration and viscosity of suspension

Apparent particle diameter	S	$\frac{1}{S}$	$\frac{\mu_s}{\mu_L} - 1$	$\frac{1}{\frac{\mu_s}{\mu_L} - 1}$
96	0.00155	645.9	0.12	8.34
96	0.00214	466	0.25	4.00
96	0.00309	324.5	0.55	1.82
60	0.001415	706.8	0.19	5.26
60	0.00218	458.7	0.44	2.27
60	0.00298	336.7	0.65	1.54
20	0.00177	566.6	0.23	4.35
20	0.00255	392.2	0.49	2.04
20	0.00334	299.4	0.91	1.099

Note :—Specific gravity of bentonite assumed = 2.4, & $\mu_L = 1.0$ centipoise

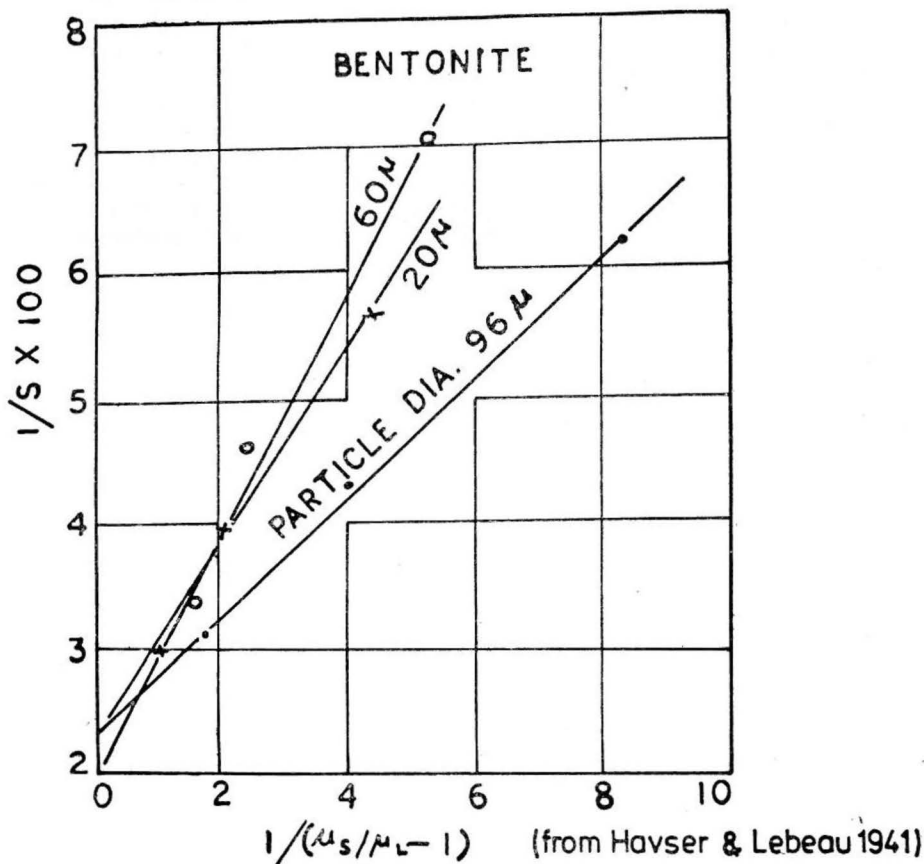


FIGURE 3. Relationship between $1/s$ and $\left(\frac{1}{\frac{\mu_s}{\mu_L} - 1}\right)$ for bentonite

TABLE 6

Values of constants x and y for bentonite water suspension

Apparent particle diameter (μ)	x	y
96	49.2	255
60	97.0	187.7
20	82.2	216.4

such a way that a straight line, for each type of clay-water suspension, describing Robinson's equation could be obtained. This data is plotted in Figure 3. The magnitude of the viscosity of the suspending medium was taken for this calculation as one centipoise. The values of constants x and y for each type of bentonite-water suspension are given in Table 6. It is seen that the magnitude of x is larger than 6.0. It is known that the value of x for a flocculated suspension with anisodimensional particles, with axis ratio upto 113, does not increase more than 6.0. This shows that the viscosity of the suspending medium has increased as the double layers of bentonite clay particles are in contact with each other. If the value of x is assumed to be 6.0 and the magnitude of the viscosity of the suspending medium is determined, then it will give the average value of the viscosity of the double layer. The magnitudes of the viscosity of the suspending medium corresponding to different suspensions with different solute concentrations and with decreasing particle sizes are shown in Figure 4. It is seen that the viscosity of the suspending medium increases with the increase in the solid concentration as the interpenetration of adjoining double layers increases with it.

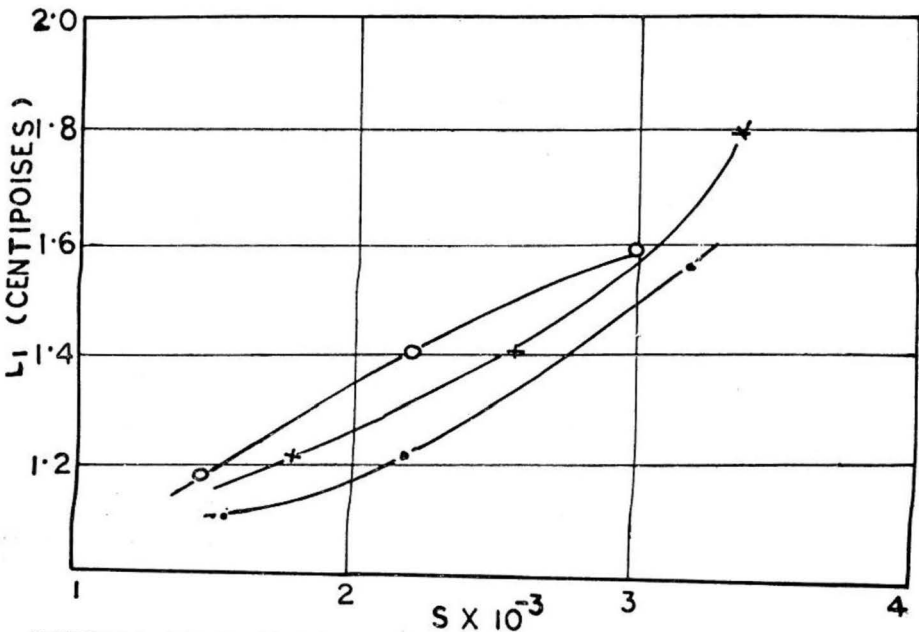


FIGURE 4. Relationship between solute concentration (bentonite) and viscosity of suspending medium.

A test series of viscosity determination was carried on a typical sample of Bombay marine clay-water suspensions with different amounts of solute and also with different concentrations of electrolyte. Another soil used for this study was a typical black cotton soil. The classification properties of these two soils are given in Table 7.

TABLE 7
Classification properties of Bombay Marine Clay and Black Cotton Soil

	Bomay marine clay	Black cotton soil
Liquid limit (%)	115.0	75.0
Plastic Limit (%)	45.0	35.0
Plasticity index	70.0	40.0
% Clay ($<2\mu$)	48.0	41.5
Activity (PI/clay %)	1.46	0.96
Specific gravity	2.70	2.67

The viscosity of soil water suspension was measured by transportable type 'Ferranti' double cylinder viscometer. The electrolyte added was a mixture of sodium hexameta-phosphate (32 gms) and sodium carbonate (7 gms) so as to make one litre of water. Different concentrations of the electrolyte added in soil-water suspensions were 2, 6 and 14 percent. Each of these different concentrations of the electrolyte was added to soil-water suspensions with proportion of soil varying from 5 to 50 percent. The results of viscosity tests for Bombay marine clay and black cotton soil are

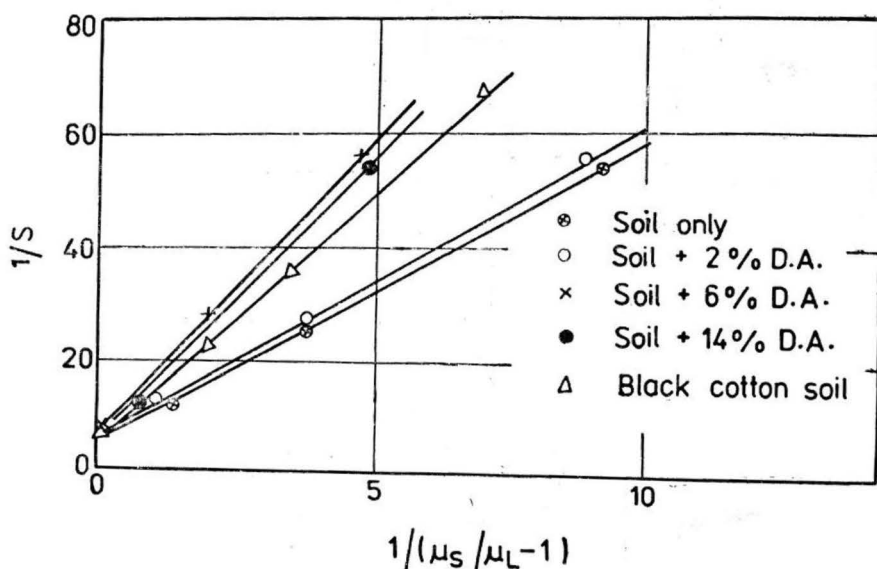


FIGURE 5. Relationship between $1/S$ and $\left(\frac{1}{\frac{\mu_s}{\mu_L} - 1}\right)$
for Bombay Marine Clay & Black Cotton Soil

TABLE 8
Data of Viscosity Test of Bombay Marine Clay

$\frac{1}{S}$	1 Soil only		2 Soil + 2% D.A.		3 Soil + 6% D.A.		4 Soil + 14% D.A.	
	(millipoises) $\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$	$\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$	(millipoises) $\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$	$\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$	(millipoises) $\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$	$\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$	(millipoises) $\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$	$\frac{1}{\left(\frac{\mu S}{\mu L} - 1\right)}$
0.0184	12.5	8.8	10.26	8.45	10.28	4.5	10.28	4.5
0.038	12.8	3.58	12.8	3.58	15.4	1.85	15.4	1.85
0.0855	18.0	1.25	20.4	0.96	26.6	0.6	25.6	0.64
0.157	96.0	0.116	72.0	0.161	20.6	0.051	175.0	0.0607

Note : D.A.—Defflocculating agent.

given in Tables 8 and 9. The relations between

$$1/S \text{ and } \frac{1}{\frac{\mu_S - \mu_L}{\mu_L}}$$

are illustrated in Figure 5. Straight line relationships are obtained showing that Robinson's equation is applicable to all soil-water suspensions. The values of constants x and y are given in Table 10. The value of constant y is not zero in any case showing that the double layers surrounding soil particles are in contact with each other.

TABLE 9
Data of viscosity test of Black Cotton Soil

Percent Soil* %	Viscosity millipoises (μ_S)	$\frac{1}{S}$	$\frac{1}{\frac{\mu_S}{\mu_L} - 1}$	S
4.0	11.5	68.0	6.66	0.0147
8.0	13.22	34.5	3.10	0.0292
12.0	15.4	23.2	1.85	0.0432
25.0	41.0	12.35	0.323	0.081

* 4.0% soil means a suspension containing 4 gm soil and 100 gms water.

TABLE 10
Values of constants x and y for Bombay Marine Clay and Black Cotton Soil

No.	Name of soil	x	y
1	Bombay marine clay only	5.54	5.7
	„ + 2% D.A.	6.01	5.1
	„ + 6% D.A.	10.85	7.2
	„ + 14% D.A.	11.0	6.87
2	Black Cotton Soil	8.7	7.5

In case of suspensions with Bombay marine clay as solute, when electrolyte concentrations were zero and two percent, the magnitude of x is less than 6.0 and more than 2.5. This shows that the viscosity of suspending medium, in other words the viscosity at the contact of double layers, is more than that of free water, but as the value of x could not be defined for these two cases, it is difficult to determine the magnitude of viscosity of the suspending medium. For soil-water suspensions with electrolyte concentrations of 6.0 and 14.0 percent, the value of x is more than 6.0. The magnitude of viscosity of suspending medium (μ_{L1}), for these two cases can, therefore, be approximately determined by assuming value of x to be 6.0.

Figure 6 gives the relationship between solid concentration and the viscosity of the suspending medium when electrolyte concentrations were 6 and 14 percent. It may be noted again that the viscosity of the suspending medium increases with the increase in solute concentration as the interpenetration of double layers increases with the increase in solid concentration. Further, for a given solid concentration the viscosity of the suspending medium is slightly smaller for higher concentration (14 percent) of electrolyte. It seems that the change in the thickness of double layers due to the increase in the electrolyte concentration from 6.0 to 14.0 percent is negligible. This led to the negligible change in the viscosity of the suspending medium with this increase in electrolyte concentration. The relationship between the solute concentration and the viscosity of suspending medium for suspensions with black cotton soil as solute is given in Figure 6.

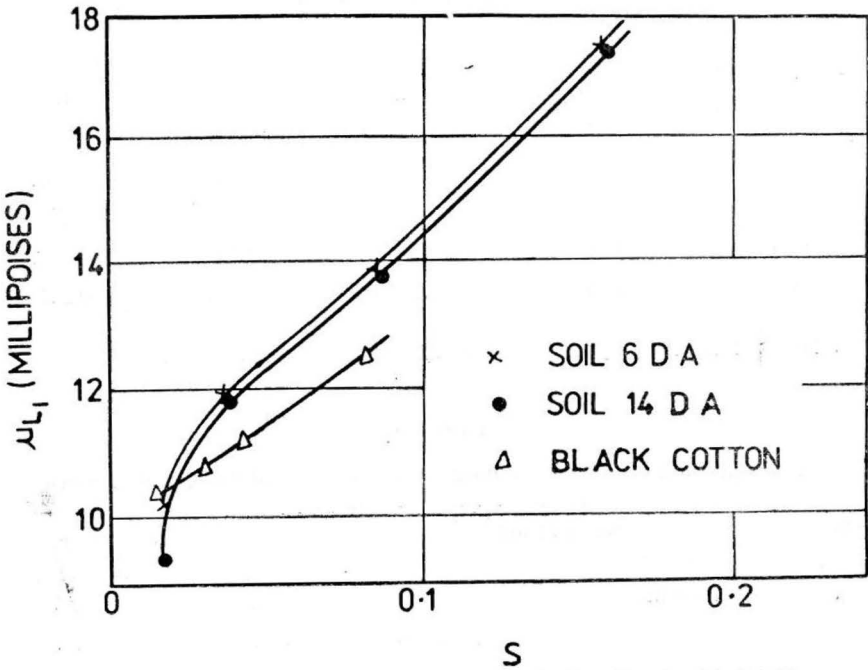


FIGURE 6. Relationship between solute concentration (Bombay Marine Clay and Black Cotton Soil) and viscosity of suspending medium

Summary and Conclusion

It is shown above that the viscosity of suspension with anisodimensional solute is mainly due to the interpenetration of the double diffuse layers of adjoining particles. The viscosity of the suspending medium at such conditions changes to the viscosity of the double layers at their contact. It was shown that the viscosity of suspension increases with the increase in the solute concentration because of increasing interpenetration of double layers surrounding solid particles with consequent increase in the viscosity of the suspending medium. This shows that the viscosity of double layer increases with the decrease in the distance from the particle surface. This fact was further confirmed by the observation that the viscosity of the suspension decreases with the increase in electrolyte concentration. For the same solid

concentration, with the increase in electrolyte concentration the thickness of double layers decrease which leads to their lesser interpenetration in consequence of which the viscosity of the suspending medium decreases.

The values of viscosity of suspending medium obtained are approximate to the extent to which the assessment of the value of constant x is correct. It is, however, shown by interpretation of many results of viscosity tests that the double layers surrounding clay particles are viscous and their viscosity increases with the decrease of the distance from the particle surface.

Viscosity is the measure of shear resistance the fluid can offer. It, therefore, seems that the rigid model of the double layer is more representative than two-dimensional one. The double diffuse layer surrounding a clay particle is able to resist not only normal forces but also shear stresses.

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Appendix A

Definition of notations used in the paper :

A & B = material constants

C = the weight of solute per cubic centimeter of the suspending medium

k = empirical constant

v = plastic viscosity

s = volume concentration of solute

x = a constant

y = a constant

ρ = volume percent of the soil

μ_L = viscosity of the suspending medium

μ_{L1} = actual value of viscosity of suspending medium depending on viscosity of the double layer

μ_S = viscosity of suspension.