

## Effective viscosity of suspension

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THE AVERAGE rate of mechanical energy dissipation in homogeneous suspensions is expressed in the form of a scalar parameter — effective viscosity. The present approach follows as close as possible EINSTEIN'S original paper and, on the other hand, has some thing in common with the cell models. However the radii of these cells are not constant but obey the distribution law determined on the basis of a closest neighbour distance distribution of statistically independent points. This enables to take into account the complex statistics of suspensions. The proposed theory covers wide range of volume concentrations of suspensions.

Średnia dysypacja energii mechanicznej w przepływach jednorodnej zawiesiny została wyrażona za pomocą wielkości skalarnej — lepkości efektywnej (lub zastępczej) zawiesiny. Przedstawione w pracy podejście jest w istocie uogólnieniem metody zaproponowanej przez EINSTEINA dla zawiesin rozrzedzonych. Z drugiej strony proponowane przez nas podejście jest bliskie tzw. modelowi komórkowemu z tym jednak, że promienie komórek nie są stałe, zaś rozkład promieni opiera się na znanym rozkładzie odległości do najbliższego punktu dla zbioru punktów losowo rozmieszczonych w przestrzeni. Teoria dotyczy szerokiego zakresu objętościowych koncentracji zawiesin.

Средняя диссипация механической энергии в течениях однородной суспензии выражена в виде скалярной величины — эффективной (или эквивалентной) вязкости суспензии. Представленный в работе подход является в сущности обобщением метода предложенного Эйнштейном для разреженных суспензий. С другой стороны предложенный нами подход близок т. наз. ячеечной модели с тем однако, что радиусы ячеек не являются постоянными, распределение же радиусов опирается на известном распределении расстояний к самой близкой точке для совокупности точек случайно распределенных в пространстве. Теория касается широкого интервала объемных концентраций суспензий.

### Notation

- $a$  particle radius,
- $b$  characteristic length,
- $D_i$  dissipation rate in the set  $G_i$  in the simple fluid,
- $D_i^p$  dissipation rate in the set  $G_i$  in the particulate fluid,
- $D_t$  total energy dissipation rate in the reference spheres
- $D(g, R, s)$  dissipation rate in the  $R$ -sphere,  $\tilde{D} = D/\beta^2$ ,
- $d(g, s)$  reduced non-dimensional dissipation rate,  $\tilde{d} = d/\beta^2$ ,
- $f(R)$  distribution function of a distance ( $2R$ ) to the nearest point,
- $f(a, R)$  distribution function of a distance ( $2R$ ) to the centre of the nearest particle,
- $f(s, g)$  distribution function of the reference spheres radii,
- $g = R/b$  non-dimensional distance from the centre of a particle,
- $K, L, M$  functions defined in (6.3),
- $n$  mean number of particles in a unit volume,
- $N$  actual number of particles in the volume  $V$ ,
- $n_i$  directional cosine,
- $p, p_0$  pressure,
- $R$  radial distance from the centre of a particle,
- $s = a/b$  non-dimensional particle radius,

$u, u_i$	velocity and velocity components,
$W$	global volume of the reference spheres per unit volume,
$\alpha_{ij}$	imposed rate-of-strain tensor,
$\bar{\alpha}_{ij}(R)$	mean value of the rate-of-strain tensor in the $R$ -sphere,
$\alpha_{ij}(\mathbf{r})$	rate-of-strain tensor inside the $b$ -sphere,
$\beta$	function defined by formula (8.2),
$\eta = \bar{\mu}/\mu$	viscosity ratio,
$\mu$	viscosity of ambient fluid,
$\bar{\mu}$	viscosity of suspended particles,
$\mu^*$	effective viscosity,
$\rho$	density of ambient fluid,
$\Phi$	volume concentration of suspended particles,
$\Phi_m$	maximum volume concentration.

## 1

IN THIS PAPER we are concerned with the dissipation of mechanical energy in suspensions in the case of simple shear flow. The average rate of energy dissipation can be expressed in the form of an "effective viscosity", a scalar parameter, which assimilates the dissipation rate in a particulate fluid and that of a simple one.

The experimental values of effective viscosity, especially for greater concentrations obtained using different kinds of viscometers vary significantly, sometimes even by an order of magnitude. This may be due to the fact that near the walls the concentration is not the same as in the bulk and, what seems to be more important, the presence of the walls induces in their neighbourhood some ordering of particles. In the bulk flow itself some authors observed the existence of clusters and regions with locally ordered distribution of particles and in most cases significant fluctuations of concentration (see for example [1]). Hence the dense suspension can hardly be treated without taking into account its statistical properties.

Statistical features of suspensions have been discussed by several authors (PRAGER [2], BUEVICH [3]), but still we are lacking an adequate description of the global mechanical behaviour of dense suspensions.

From the theoretical point of view the notion of effective viscosity encounters serious difficulties: besides the change of the concentration near the walls the isotropic structure of the suspension may be violated by the flow and hence the necessary condition for the proper definition of the viscosity — the isotropy of the medium — may be not fulfilled.

In the first part of the paper we discuss the questions of homogeneity and isotropy of particulate media and this leads us to the critical recapitulation of the assumptions commonly used in derivation of the effective viscosity of suspension.

The second part of the paper contains the theory of energy dissipation which can be applied to low Reynolds number flows of an incompressible liquid for a wide range of concentrations.

The present approach follows as close as possible Einstein's original paper and we do believe that (as it was expressed by Eddington in relation to Einstein's paper on radiation) his investigation gives the clearest insight into the considered phenomenon. In the Appendix the Einstein original paper is recapitulated and confronted with the proposed approach.

On the other hand, the present theory has much in common with the cell (or cage) theory proposed originally by SIMHA [4]. Different versions of this approach are presented in [5]. There are, however, several important deviations from previous contributions. First, the radius of the cell (sphere) is not constant but obeys the distribution law determined on the basis of a closest neighbour distance distribution of statistically independent points. This enables us to take indirectly into account the fluctuations of the velocity field in suspensions and their contribution to the rate of energy dissipation. It is also important to note that the enveloping spheres are not overlapping. Second, the boundary conditions are chosen in such a way that the mean simple shear flow in the neighbourhood of each particle is the same as the imposed one.

The proposed theory gives in the limit of dilute suspension the Einstein-Taylor expression for the effective viscosity.

## 2

For finding the average values needed for the description of a suspension we must start with the definition of the homogeneous distribution of particles. A medium is considered homogeneous when the number of particles in a volume large in comparison with the size of the individual particle is proportional to this volume. However for, the theory of dense suspensions we need a more precise definition.

The random distribution of statistically independent points in space obeys the Poisson distribution. Let  $G_\alpha$  be sets with a given measure,  $\text{mes}(G_\alpha) = V$  for all  $\alpha$ , and let  $n$  be the mean number of particles in a unit volume. Then the relative number of sets containing  $N$  points is

$$(2.1) \quad w(N) = \frac{(nV)^N e^{-nV}}{N!}.$$

Hence the mean number of points within a distance between  $2R$  and  $2(R+dR)$  from a given point is equal to  $32\pi R^2 dR$  and the probability that the nearest point to a given one is at such a distance is

$$(2.2) \quad f(R)dR = 32\pi R^2 n \cdot \exp\left[-\frac{32}{3}\pi n R^3\right] dR.$$

It is clear that these expressions are not valid for spheres of a finite radius (constant radius  $a$ ); for large  $N$  the appropriate  $w_a(N)$  function is identically zero, since only a finite number of spheres can be embedded in a finite volume  $V$ ; also  $f_a(R)$  is identically zero for  $R < a$  and, moreover, this function is not even monotonous because for a close packing and in its neighbourhood there exists a given order of spheres.

Still for small volume concentrations  $\Phi$  i.e. for  $na^3 \ll 1$  the previous formulae can be safely used and they give the value of the relative fluctuation of the volume concentration  $\Phi_N = \frac{4\pi}{3}Na^3$  in the form

$$(2.3) \quad \frac{\sqrt{\Delta\Phi_N^2}}{\Phi} = \sqrt{\frac{\frac{4}{3}\pi a^3}{V\Phi}} = \frac{1}{\sqrt{nV}}, \quad \text{where} \quad \Phi = \frac{4}{3}\pi na^3.$$

In spite of many efforts (connected mainly with the kinetic theory of fluids) the precise definition of the random distribution of finite spheres is not known [6].

However using physical arguments, similar to those used originally for (2.1) by SMOLUCHOWSKI, BUEVICH [3] has derived the distribution function which reads

$$(2.4) \quad w(N) = C_{N_v}^N \Phi^N (1 - \Phi)^{N_v - N},$$

where  $C_{N_v}^N$  is the normalizing coefficient and  $N_v$  the maximal number of particles which can be embedded in a volume  $V$ , i.e.,  $N_v \cdot v_e = V$ , where  $v_e$  is a volume of an elementary cell defined as a ratio of the particle volume to the volume concentration of the close packing  $\Phi_m$ . This distribution gives instead of (2.3) the expression

$$(2.5) \quad \frac{\sqrt{\Delta \Phi_N^2}}{\Phi} = \sqrt{\frac{\Phi_m - \Phi}{\Phi}} \frac{1}{\sqrt{nV}}.$$

On the basis of this consideration the homogeneous distribution can be defined as such for which the relation (2.5) holds.

Instead of the Eq. (2.2) we will (see Sec. 6) introduce the distribution function which in the dilute limit gives (2.2) and for the close packing tends to the delta-function,  $\delta(R-a)$ .

The homogeneity of the suspension is the necessary but not sufficient condition for its isotropy. It is clear that for non-spherical particles the flow itself induces the orientation of particles and the isotropy is lost. A deeper result has been obtained by BATCHELOR and GREEN [9]; they have shown that even in the case of spherical particles in a simple shear flow the structure of suspension is flow-dependent due to the presence of closed trajectories of one particle center relative to the other one. The Brownian motion and the hydrodynamic interference with other particles presented in the flow undoubtedly try to regain the isotropic structure.

### 3

The viscosity is intrinsically connected with the isotropic structure of the medium.

As the flow itself violates the isotropy the viscosity in the usual sense cannot be determined and in this paper the term "effective viscosity" is used only as a convenient measure of the average rate of energy dissipation. This measure can be used only when some conditions are fulfilled, namely:

(a) the suspension is homogeneous (but not necessarily isotropic);

(aa) the distribution function of the orientation of particles (if non-spherical) does not depend on the initial positions of particles.

These two conditions are similar to the condition of thermodynamic equilibrium which is necessary for the definition of the usual viscosity of simple liquids: its determination on the basis of the kinetic equation involves the assumption that the appropriate distribution function is close to the Maxwellian; the phenomenological definition is also possible only in the state of equilibrium, i.e., if the gradients of velocity, pressure and temperature are small [8, § 49].

Now let a simple shear flow be assumed in the whole space. Further let  $G_i$ ,  $i = 1, 2, \dots$  are such sets that  $G_i \subset G_j$ , if  $i < j$  and such that  $V_i = \text{mes}(G_i)$  is finite for all  $i$  but  $\lim_{i \rightarrow \infty} V_i = \infty$ . The prescribed velocity field imposes the boundary conditions on  $\Gamma G_i$ . The rate of dissipation of mechanical energy in  $G_i$  is equal to

$$(3.1) \quad D_i = \int_{G_i} \psi dV$$

where, assuming the Cauchy-Poisson relation, the dissipation function  $\psi$  can be written in the form

$$\psi = \delta \theta^2 + 2\mu \alpha_{ij} \alpha_{ij}, \quad \delta, \mu - \text{viscosity coefficients}$$

with  $\theta = \text{div } \mathbf{u}$ ,  $\mathbf{u}$  being the velocity field and  $\alpha_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i})$  is the rate-of-strain tensor

In principle it is possible to solve the equation of motion both in the case when in  $G_i$  we have only a simple fluid (and to obtain  $D_i$ ) and in the case when the particles are distributed in this volume. In the last case it is assumed that the particles are not deformable and hence that there is no additional contribution to dissipation due to the deformation. Both the material inside and outside the particles is assumed Newtonian but possibly with the different values of viscosity coefficients (resp.  $\bar{\mu}$  and  $\mu$ ). The particular flow field  $\mathbf{u}$  and the dissipation depend on the positions of particles in  $G_i$  but due to the assumptions introduced above there exists a mean value  $\langle D_i^t \rangle$  for  $t \rightarrow \infty$ . The quantity

$$(3.2) \quad \mu^* = \mu \lim_{t \rightarrow \infty} \frac{\langle D_i^t \rangle}{D_i}$$

is called the effective viscosity.

This definition takes into account the very fact that the kinematics of the flow of a medium remains the same when the simple liquid is replaced by a particulate one (at least far from the walls).

It is important to notice that the rheological properties of suspensions will exhibit themselves through the response to the change of the imposed velocity field. These properties are due to the reorientation of particles or their trajectories. The effective viscosity as defined above does not describe this phenomenon and the relaxation time involved. The result of BATCHELOR and GREEN [9] shows that the effect of reordering can be significant even in the case of spherical particles (the change of the coefficient of the  $\Phi^2$ -term).

#### 4

In such a general form the problem hardly can be examined and several simplifications have been introduced. They are of different kinds:

The first simplification is that we confine ourselves to incompressible flows. Only for these flows we know how to compute the rate of dissipation, because instead of (3.1) we have simply

$$(4.1) \quad D_i = 2\mu \int_{G_i} \alpha_{kl} \alpha_{kl} dV.$$

As only the rate-of-strain tensor influences the energy dissipation rate then for the simple shear flow for which  $\alpha_{ki}(\mathbf{r}) = \text{const}$  we have at once  $D_i = 2\mu V_i \alpha_{ki} \alpha_{ki}$ .

Second, in most papers the authors discuss only spherical non-deformable particles to remove the question of orientation and deformation of particles.

Third, it is assumed that there is no particle-particle interactions (besides hydrodynamical), that no external forces are acting on particles and that the Reynolds number of the flow is small. These conditions justify the use of the Stokes equation.

The next, fourth assumption presented in Einstein's paper and in most other papers relates to the level of concentration. The arguments run as follows: if the suspension is very dilute then the interactions of particles are negligible and one can consider the contribution of each particle separately. This means that we prescribe (or compute) an additional dissipation  $D'$  due to the presence of a single particle and hence the energy dissipation rate in  $G_i$  is  $D_i^p = 2\mu V_i \alpha_{ke} \alpha_{ke} + N_i D'$ , where  $N_i$  is the number of particles in the set  $G_i$ . Hence the effective viscosity can be written directly as

$$(4.2) \quad \mu^* = \mu \left( 1 + \frac{D'}{2\mu \alpha_{ke} \alpha_{ke}} \lim_{i \rightarrow \infty} \frac{N_i}{V_i} \right) = \mu \left( 1 + \frac{nD'}{2\mu \alpha_{ke} \alpha_{ke}} \right)$$

or,

$$(4.3) \quad \mu^* = \mu(1 + nC),$$

where  $C$  is an unknown constant.

These arguments are however open to some criticism. From the use of linear Stokes equation it follows immediately that the velocity (at a given point) is an additive function but in no case this is true for the dissipation. In essence Einstein's approach is not connected with the idea of noninteracting particles but with the conviction that the presence of particles changes the dissipation significantly only in their closest neighbourhoods and that the additional dissipation far from the particle plays a negligible role in the total energy balance.

The fifth and final assumption concerns the way of computing  $D'$ . EINSTEIN found the solution of Stokes equation in the case of a rigid sphere assuming straining motion condition at infinity. However the range of validity of Stokes equation is [7, p. 250]:

$$(4.4) \quad \frac{R}{a} < \left( \frac{|\alpha_{ij}| a^2 \rho}{\mu} \right)^{-1/2}$$

and hence it is not surprising that the use of the velocity field far from the particle may lead to some kind of "paradox". In the considered case it is the following one: the integral describing  $D'$  is divergent and the proper mathematical method of avoiding this very fact does not exist.

The ways to go around this difficulty are of different kinds. The first one used by EINSTEIN himself and criticized just after publication of his work is based on the use of the principle value of the divergent integral. The second one proposed by LANDAU [8] and essentially improved by BATCHELOR [7] is based on the use of the divergence theorem. BATCHELOR considers the finite region (ours  $G_i$ ) and the straining motion on  $\Gamma G_i$ . The dissipation rate can be expressed as the sum of integrals over the surface of the (unique) particle in  $G_i$ . However in the final calculation instead of use of the assumed straining

motion condition on  $\Gamma G_i$  the authors are using the condition which follows from the imposed straining motion at infinity.

These are some indications that if the Navier-Stokes equation is used instead of the Stokes equation this will lead to the finite value of the integral describing  $D'$ ; BERKER [10, §77] has shown though only for a particular flow field at infinity that in the steady flow of a viscous incompressible fluid past a fixed body the disturbed velocity vanishes faster for the Navier-Stokes solution than this for the Stokes solution. The velocity field in the Einstein problem vanishes as  $R^{-2}$  and as it has been stated many times if the solution of the full equation vanishes as  $R^{-2-\epsilon}$ ,  $\epsilon > 0$ , then  $D'$  will be finite.

## 5

In what follows we use the first three assumptions in the unchanged form. The fourth assumption is relaxed and we indirectly take into account the hydrodynamic interactions of particles. This assimilates the present approach with those based on general statistical considerations [2] and permits its applicability to a wide range of concentrations. Unlike pure statistical theories ours essentially uses hydrodynamical equations which enables us to avoid the use of numerical values of parameters taken from experiments.

Also the fifth assumption is not used, instead we exploit the idea that the dissipation can be presented as a sum of the dissipation near the particles (inside the reference spheres) and the dissipation rate in the region outside the reference spheres.

## 6

Let in a volume  $G$  be given the velocity field  $\mathbf{u}(\mathbf{x})$ . In the neighbourhood of each point the velocity field can be split according to the Helmholtz theorem into the translational, rotational and straining part. Only the last term contributes to the dissipation.

Let each single particle be surrounded by its own reference sphere with radius  $R$  such that no other particle enter this sphere. The dissipation rate inside the reference sphere can be calculated by solving the following problem: to find the velocity field in an incompressible liquid sphere of radius  $b$  and viscosity  $\mu$ , surrounding a concentric incompressible liquid sphere (with viscosity  $\mu$ ) of radius  $a$ ,  $a < b$ . The equations describing this problem are

$$(6.1) \quad \begin{aligned} \mu \Delta u_i &= p_{,i} & a \leq r \leq b \\ \bar{\mu} \Delta u_i &= p_{,i} & 0 \leq r \leq a, \\ u_{i,i} &= 0 & r \leq b. \end{aligned}$$

We assume the outer boundary condition (on the  $b$ -sphere) in the form of the pure straining motion i.e.

$$(6.2) \quad u_i = \alpha_{ij} n_j \quad \text{for} \quad r = b.$$

The liquid sphere (of radius  $a$ ) remains spherical due to surface tension and on this sphere we assume the no-slip condition. The functions  $u$  and  $p$  must be continuous in the whole

field  $r \leq b$ . The condition of no net force or torque at the inner sphere must be fulfilled and besides the radial component of  $u$  must vanish and tangential component of the normal stress must be continuous. This problem can be solved using LAMB [11] method and the solutions for some particular cases were found independently by several authors. The full solution for all admissible values of  $\bar{\mu}$  is given in [12] and the results are as follows:

$$\begin{aligned}
 u_i(\mathbf{r}) &= \frac{1}{\Delta(s, \eta)} [K(r, s, \eta) \alpha_{ij} n_j + L(r, s, \eta) \alpha_{ji} n_j n_i], \\
 (6.3) \quad p(\mathbf{r}) &= p_0 + \frac{\bar{\mu}}{\Delta(s, \eta)} M(r, s, \eta) \alpha_{ij} n_i n_j, \quad \text{for } r \leq a, \\
 p(\mathbf{r}) &= p_0 + \frac{\mu}{\Delta(s, \eta)} M(r, s, \eta) \alpha_{ij} n_i n_j, \quad \text{for } a \leq r \leq b,
 \end{aligned}$$

where for  $r \leq a$

$$(6.4) \quad K = \left( -5 \frac{r^3}{a^2} + 3r \right) \Phi, \quad L = 2 \frac{r^3}{a^2} \Phi, \quad M = -21 \frac{r^2}{a^2} \Phi$$

and for  $a \leq r \leq b$

$$\begin{aligned}
 (6.5) \quad K &= \Phi_1 r - 5 \frac{r^3}{a^2} \Phi_2 + 4 \frac{a^5}{r^4} \Phi_3, \\
 L &= 2 \frac{r^3}{a^2} \Phi_2 + 2 \frac{a^3}{r^2} \Phi_0 - 10 \frac{a^5}{r^4} \Phi_3, \\
 M &= - \left[ 21 \frac{r^2}{a^2} \Phi_2 - 4 \frac{a^3}{r^3} \Phi_4 \right].
 \end{aligned}$$

Here  $\eta = \bar{\mu}/\mu$ ,  $s = a/b$  and the expressions for the functions introduced above are [12]:

$$\begin{aligned}
 (6.6) \quad \Phi(s) &= 5s^7 - 7s^5 + 2, \\
 \Phi_1(s, \eta) &= - [5(2 - 5\eta)s^7 + 21\eta s^5 + 4(1 + \eta)], \\
 \Phi_2(s, \eta) &= s^5 [5\eta s^2 - (2 + 5\eta)], \\
 \Phi_3(s, \eta) &= (1 - \eta)s^5 + \eta, \\
 \Phi_4(s, \eta) &= 5(1 - \eta)s^7 + 2 + 5\eta, \\
 \Delta(s, \eta) &= 4(1 - \eta)s^{10} - 5(2 - 5\eta)s^7 - 42\eta s^5 + 5(2 + 5\eta)s^3 - 4(1 + \eta).
 \end{aligned}$$

The energy dissipation rate  $D(g, R, s)$  in the  $R$ -sphere is

$$\begin{aligned}
 (6.7) \quad D(g, R, s) &= \frac{8}{3} \pi R^3 \bar{\mu} \alpha_{ij} \alpha_{ij} d(g, s), \quad 0 \leq R \leq a, \\
 D(g, R, s) &= \frac{8}{3} \pi R^3 \mu \alpha_{ij} \alpha_{ij} d(g, s), \quad a \leq R \leq b,
 \end{aligned}$$

where  $g = R/b$ ,  $g \in [s, \infty)$  and

$$d(g, s) = \frac{1}{5R\Delta^2} \left[ K \left( \frac{7}{2} K_{,R} + 2L_{,R} + \frac{3}{R} L + \frac{3}{2R} K - M \right) + L(2K_{,R} + 2L_{,R} - M) \right];$$



in the above formulae we have not visualized the dependence of the  $D$  and  $d$  functions on the viscosities. The function  $d(g, s)$  denotes the reduced, non-dimensional dissipation rate per unit volume.

For further use it is important to note that for  $s = 1 - \varepsilon$ ,  $\varepsilon \ll 1$ , we get

$$(6.8) \quad d(1,1) \simeq \frac{-14 + (112 - 70\eta)\varepsilon}{-140\varepsilon^3 + (490 - 175\eta)\varepsilon^4} \sim \varepsilon^{-3},$$

and, on the other hand, for  $s \rightarrow 0$

$$(6.9) \quad d(1, s) \underset{s \rightarrow 0}{\rightarrow} 1 + \frac{5\eta + 2}{2(1 + \eta)} s^3 + \frac{5(5\eta + 2)^2}{8(1 + \eta)^2} s^6 + \dots$$

## 7

As mentioned above we divide the flow field into two regions: one near the particles taken as a sum of the reference spheres and the particle-free region.

A detailed description of the position and hydrodynamical interactions of the particles is not possible and hence we are forced to use an indirect method. We assume that the reference radii vary and the boundary conditions are those following from the imposed velocity field. In this approach we may use as a guide the distribution function of the distance from the given point to its closest neighbour.

The characteristic length of the suspension,  $b$ , is taken as proportional to  $n^{-1/3}$ . For taking into account the limit of dense suspension we assume that for the close packing i.e. for  $\Phi = \Phi_m$ ,  $b = a$ . Hence we arrive at the following definition of  $b$ :

$$(7.1) \quad b^3 = \frac{3\Phi_m}{4\pi n} \quad \text{or} \quad \Phi = \Phi_m \left( \frac{a}{b} \right)^3.$$

Using this value (2.2) can be presented in the form

$$(7.2) \quad f(R) = AR^2 \exp \left[ -8\Phi_m \left( \frac{R}{b} \right)^3 \right],$$

where  $A$  is the normalizing coefficient.

Now our main assumption is that for the spheres of constant radius  $a$  the distribution function of  $R$  is given by

$$(7.3) \quad f(a, R) = \tilde{A}(R-a)^2 \exp \left[ -8\Phi_m \left( \frac{R-a}{b-a} \right)^3 \right],$$

where  $\tilde{A}$  is again the normalizing coefficient. For the very dilute suspensions  $a \ll b$  and in the limit  $a \rightarrow 0$  we get the classical formula (2.2) and for the dense suspensions,  $a \approx b$ , we get the  $\delta$ -function as it has to be expected from the geometry of close packing.

It must be however stressed that in spite of the fact that both in the limit of dilute and dense suspensions the formula (7.3) gives proper results it cannot be considered as an exact formula describing the distribution of the distance to the closest sphere. We claim only that one may expect on the basis of physical properties that (7.3) gives a good approximation to such a distribution.

The function (7.3) will be taken as the function describing the variation of the reference radius  $R$  for a given value of characteristic length  $b$ .

The physical meaning of (7.3) is that we surround each particle by a sphere of radius equal to half of the distance to its closest neighbour. Hence the reference spheres of different particles are not overlapping.

By introducing the non-dimensional variables  $s$  and  $g$  we arrived at

$$(7.4) \quad f(s, g) = \frac{24\Phi_m}{(1-s)^3} (g-s)^2 \exp \left[ -8\Phi_m \left( \frac{g-s}{1-s} \right)^3 \right],$$

where

$$\int_s^\infty f(s, g) dg = 1.$$

## 8

The kinematics of the flow of suspensions shows that the mean velocity for the particulate medium is the same as for the simple fluid. However by introducing  $R$ -spheres we have violated this property and we aim now to restore it keeping simultaneously the pattern of the simple shear flow in the reference sphere unaltered.

The mean value of  $\alpha_{ij}$  in the  $R$ -sphere (for  $R \leq b$ ) is

$$(8.1) \quad \bar{\alpha}_{ij}(R) = \frac{1}{K_R} \int_{K_R} \alpha_{ij}(\mathbf{r}) dV, \quad K_R = \frac{4}{3}\pi R^3,$$

and by the use of (6.3) we get

$$(8.2) \quad \bar{\alpha}_{ij}(R) = \beta(g, s) \alpha_{ij},$$

where

$$\beta(g, s) = \frac{1}{\Delta} \left[ \left( \frac{s}{g} \right)^3 \left( -\frac{6}{5}\Phi - \Phi_1 + \frac{21}{5}\Phi_2 \right) - \frac{21}{5} \left( \frac{g}{s} \right)^2 \Phi_2 + \Phi_1 \right].$$

(At this point we again follow Einstein's idea, see Appendix).

It is important to notice that the function  $\beta$  is a scalar one with scalar arguments and that  $\beta(1, s) = 1$ . The general shape of  $\beta$ -function for  $\eta = \infty$  is shown in Fig. 1 ( $\Phi_m = 0.74$ ).

From the linearity of Stokes equation it follows that by changing the boundary condition on  $b$ -sphere, i.e. by setting

$$(8.3) \quad \tilde{u}_i(\mathbf{r}) = u_i(\mathbf{r})/\beta(g, s) \quad \text{for} \quad |\mathbf{r}| = b,$$

we obtain that the mean value of  $\tilde{\alpha}_{ij}(\mathbf{r})$  in the  $R$ -sphere is equal to the rate-of-strain tensor of the superimposed velocity field, i.e. to  $\alpha_{ij}$ .

Because of the scalar character of the  $\beta$  function the pattern of the flow remains unaltered. Using this velocity field we obtain for the dissipation rate in the  $R$ -sphere the expression

$$(8.4) \quad \tilde{D}(g, R, s) = \frac{2\mu}{\beta^2} \int_{K_R} \alpha_{ij}(\mathbf{r}) \alpha_{ij}(\mathbf{r}) dV = \frac{8}{3} \pi \mu R^3 \alpha_{ij} \alpha_{ij} \tilde{d}(s, g), \quad 0 \leq R \leq b,$$

where  $\tilde{d}(s, g) = d(s, g)/\beta^2$ .

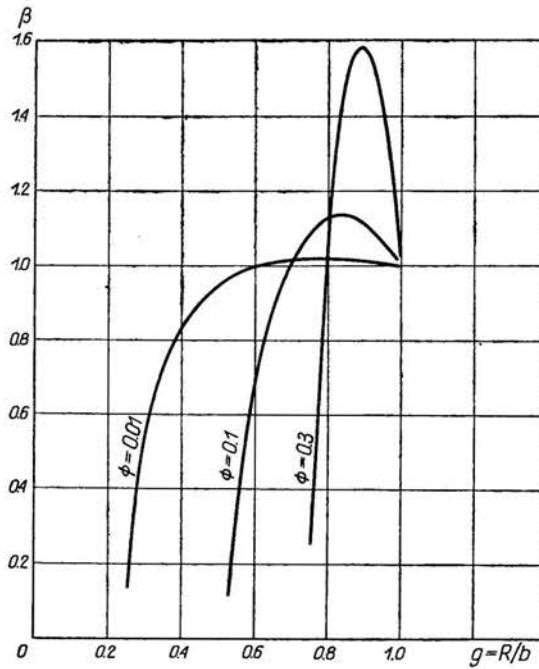


FIG. 1. The plot of the function  $\beta$  versus  $g = R/b$  for the rigid particles and for different volume concentrations.

For  $g > 1$ , i.e.,  $R > b$  we must take

$$\tilde{d}(s, g) = \tilde{d}(s, 1) + g^3 - 1$$

according to the physical sense of the proposed approach.

## 9

Having the distribution function  $f(s, g)$  and the energy dissipation rate  $\tilde{d}(s, g)$  in a reference sphere of radius  $R$  we are able to represent the total dissipation inside the reference spheres in the form

$$(9.1) \quad D_t = 2\mu\alpha_{ij}\alpha_{ij}\Phi_m \int_s^\infty g^3 \tilde{d}(s, g) f(s, g) dg = 2\mu\alpha_{ij}\alpha_{ij}D_t^*$$

In the region outside the reference spheres the energy dissipation rate is equal to

$$(9.2) \quad 2\mu\alpha_{ij}\alpha_{ij} \cdot (1 - \Phi_m) \int_s^\infty g^3 f(s, g) dg = 2\mu\alpha_{ij}\alpha_{ij}(1 - W)$$

and hence the effective viscosity  $\mu^*$  is

$$(9.3) \quad \mu^* = \mu(1 + D_t^* - W).$$

The effective viscosity  $\mu^*$  depends on  $s$  or, what is the same on the volume concentration  $\Phi$ . The maximum concentration  $\Phi_m$  plays a role of parameter.

Now we will show that for small concentrations we obtain the Einstein-Taylor formula which is independent of  $\Phi_m$ . Using the asymptotic expansion of  $d(s, g)$  [formula (6.8)] from (9.3) it follows that

$$(9.4) \quad \frac{\mu^*}{\mu} = 1 + \frac{5\eta + 2}{2(1 + \eta)} \Phi + (\text{higher terms}) + \Phi_m \int_s^1 \left[ \tilde{d}(s, g) - 1 - \frac{\tilde{d}(s, 1) - 1}{g^3} \right] g^3 f(s, g) dg.$$

The expression for  $\tilde{d}(g, s)$  reads

$$(9.5) \quad \tilde{d}(g, s) \xrightarrow{s \rightarrow 0} 1 + \frac{5\eta + 2}{2(1 + \eta)} \frac{s^3}{g^3}$$

and the simple calculations show that the integral in (9.4) is of an order of magnitude higher than  $s^3$ .

It is important to mention that we cannot use the Stokes equation in the region larger than the one prescribed by the inequality (4.4). From this inequality it follows also that the admissible values of the imposed rate-of-strain tensor are greater for denser suspensions since the characteristic length  $b$  is smaller or, in other words, the Reynolds number based on the effective viscosity decreases with the increase of concentration. From this point of view the classical Einstein formula is valid not only for the limit of very dilute suspension but also for the limit of very small imposed rate-of-strain tensors.

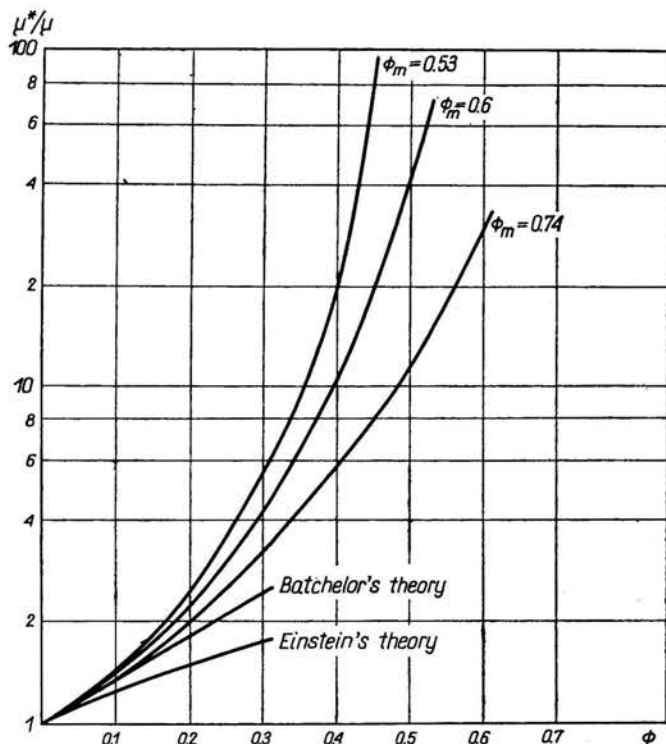


FIG. 2. The dependence of the effective viscosity on volume concentration (rigid particles) for different values of the maximal volume concentration  $\Phi_m$ .

## 10

As was mentioned earlier the effective viscosity  $\mu^*$  depends on the chosen value of the close packing parameter  $\Phi_m$ . For the rigid particles this dependence is shown in Fig. 2 which presents the plot of effective viscosity as a function of volume concentration. Besides the curve which corresponds to the theoretical value of close packing ( $\Phi_m = 0.74$ ), two other curves are presented; first for  $\Phi_m = 0.6$ , the value of close packing parameter determined experimentally and the second,  $\Phi_m = 0.53$  which corresponds to simple cubic packing. Our results are also compared with those which follow from Einstein's original formula and with BATCHELOR and GREEN second order formula  $\mu^*/\mu = 1 + 2.5\Phi + 7.6\Phi^2$ . This comparison shows that the differences became significant even for small concentrations; our curve for  $\Phi_m = 0.74$  coincides with Einstein's results only up to the concentration 0.04 and with Batchelor's up to 0.1.

It is of some interest to compare contributions due to different terms entering into the expression (9.3), namely  $W$  and  $D_t^*$  (Fig. 3). It is clear that the contribution of  $D_t^*$ , i.e., of

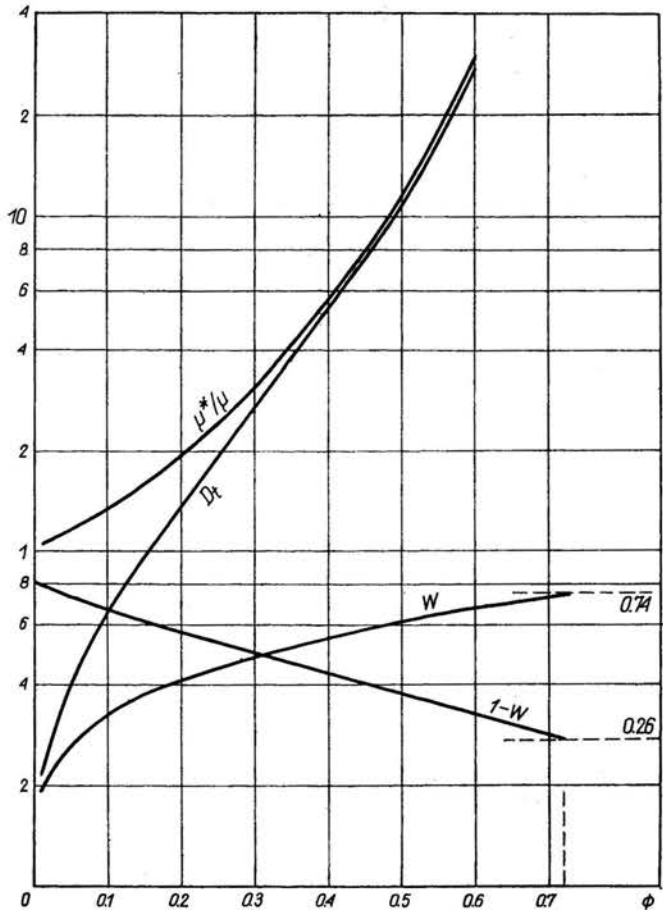


FIG. 3. Contributions of different terms,  $D_t^*$  and  $W$ , to the effective viscosity ( $\eta = \infty$ ,  $\Phi_m = 0.74$ ).

the dissipation in the vicinity of particles grows rapidly with the volume concentration and that the dissipation outside the reference spheres plays an important role only for dilute suspension. With the increase of concentration the term  $1 - W$  becomes small in comparison with  $D_i^*$ .

There are many experimental investigations concerning the measurements of the effective viscosity and the most extensive presentation of available data was given by THOMAS [13]. He collected the data obtained with both rotational and capillary viscometers

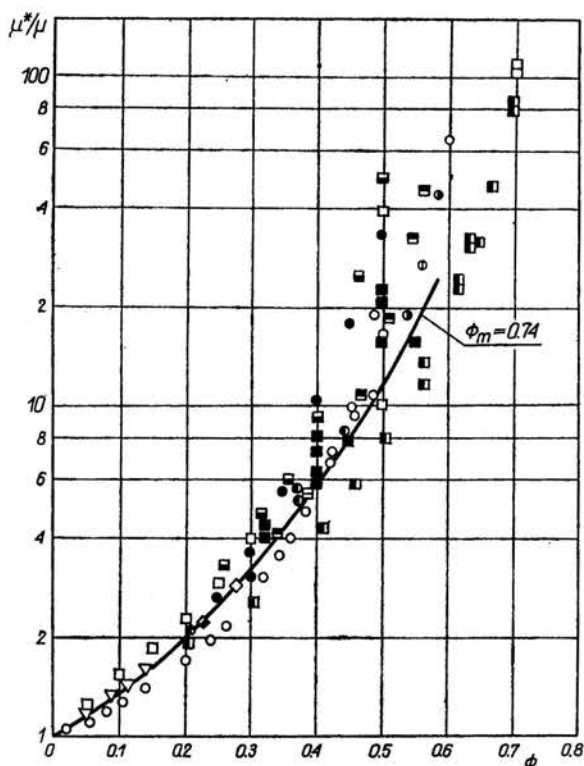


FIG. 4. The comparison of the theoretical curve for the rigid spheres and  $\Phi_m = 0.74$  with the data collected by THOMAS [13].

for a wide range of particle diameters and materials. The scatter of experimental data is large and hence the good fitting of our curve cannot be treated as its full verification (Fig. 4).

Similar results to these shown in Fig. 2 were obtained for different viscosity ratio  $\eta$ . Here we present the influence of the viscosity ratio on the effective viscosity (Fig. 5). The effective viscosity for a given concentration increases with  $\eta$ . The lowest value is reached by suspensions of gas bubbles, the highest for suspensions of solid particles. The general shape of all curves is the same but it must be noticed that it may be due to the simplifying assumption that in all cases the particles are not deformable.

The dependence of  $\mu^*/\mu$  on the value of the imposed velocity field reported by several authors as an experimental result (see for example [13]) can be perhaps partially attributed to the deformability and compressibility of suspended particles.

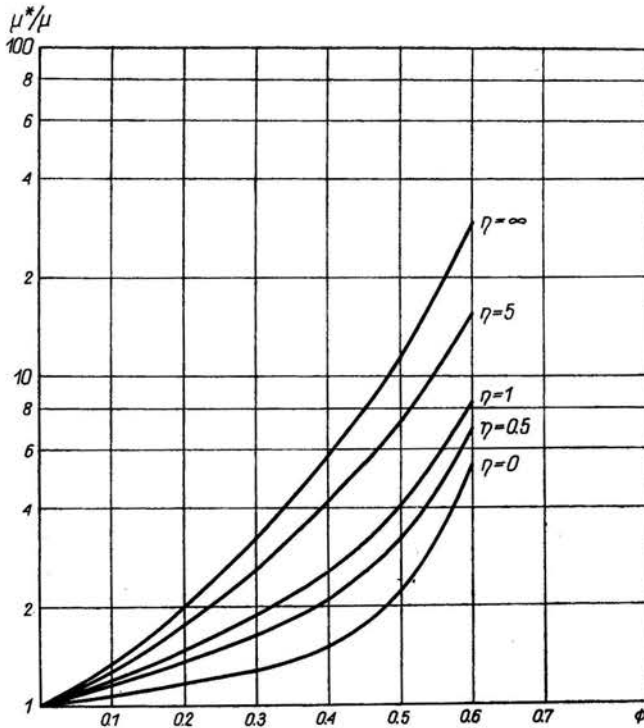


FIG. 5. The dependence of the effective viscosity on the viscosity ratio  $\eta = \bar{\mu}/\mu$ ,  $\Phi_m = 0.74$ .

## Appendix

As was mentioned earlier our approach is very close to the one given in Einstein's original work [14]. To our best knowledge his approach has never been repeated and hence it seems worthwhile to recapitulate briefly his paper.

Einstein starts with the solution of the flow field assuming the pure straining motion conditions at infinity ( $b = \infty$ ). This leads (in our notation) to the expressions:

$$(A1) \quad K = \left( -\frac{a^5}{r^5} + 1 \right) r, \quad L = \frac{5}{2} \left( \frac{a^5}{r^4} - \frac{a^3}{r^2} \right), \quad M = -5 \frac{a^3}{r^3},$$

instead of our formula (6.5). The dissipation rate based on (A1) is

$$(A2) \quad D(a, R., 0) = \frac{8}{3} \pi R^3 \mu \alpha_{ij} \alpha_{ij} \left[ 1 + \frac{1}{2} \left( \frac{a}{R} \right)^3 - \frac{15}{2} \left( \frac{a}{R} \right)^6 + 12 \left( \frac{a}{R} \right)^8 - 6 \left( \frac{a}{R} \right)^{10} \right].$$

In Einstein's original version only terms up to  $(a/R)^3$  are given due to the assumption of a very low concentrated suspension. Hence it follows that the additional dissipation rate due to the presence of a single particle is  $D' = \frac{4}{3} \pi a^3 \mu \alpha_{ij} \alpha_{ij}$  and that the global dissipation is of the form

$$(A3) \quad D = 2\mu \alpha_{ij} \alpha_{ij} \left( 1 + \frac{\Phi}{2} \right)$$

Einstein calculates the influence of the presence of particles on the straining motion field and finds that the relation between  $\alpha_{ij}$  and the mean straining motion in the suspension is

$$(A4) \quad \bar{\alpha}_{ij} = \alpha_{ij}(1 - \Phi).$$

If  $\mu^*$  denotes the effective viscosity then the dissipation rate in the field  $\bar{\alpha}_{ij}$  is  $2\mu^*\bar{\alpha}_{ij}\bar{\alpha}_{ij}$ . Comparing this expression with (A3) and using (A4) Einstein finally finds

$$(A5) \quad \frac{\mu^*}{\mu} = 1 + \frac{5}{2}\Phi.$$

In our approach the role of relation (A4) is played by (8.2) which for the dilute suspension of solid spheres leads to

$$(A6) \quad \beta(a, R, \infty) = 1 - \left(\frac{a}{R}\right)^3.$$

For the low concentrated suspension it is convenient to introduce the variable  $\delta = a/R$  and by using it the formula (7.4) for solid particles gives

$$(A7) \quad f(\delta, s) = \frac{24\Phi_m}{(1-s)^3} \left(\frac{s}{\delta}\right)^2 (1-\delta)^2 \exp\left[-8\Phi_m \frac{s^3(1-\delta)^3}{\delta^3(1-s)^3}\right],$$

and the formula (9.3) leads immediately to

$$(A8) \quad \frac{\mu^*}{\mu} = 1 + \frac{5}{2}\Phi - \frac{84\Phi^2}{(1-s)^3} \int_0^1 \frac{(1-\delta)^2}{\delta} \exp\left[-8\Phi_m \frac{s^3(1-\delta)^3}{\delta^3(1-s)^3}\right] d\delta$$

It is interesting to mention that the first non-linear term in concentration is negative as follows also from (A2).

The above derivation seems to have several advantages over those commonly used, namely it uses explicitly the randomness of the particles distribution and the dissipation is calculated in separate, not overlapping volumes.

However due to the involved assumption about the form of distribution function this derivation does not pretend to satisfy those who wish to have a precise mathematical proof. The only way to obtain such a proof is to find the upper and lower limits of dissipation in the set occupied by many particles as it has been done by KELLER, RUBENFELD and MOLYNEUX [12].

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