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The Viscosity of Concentrated Suspensions and Solutions*

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An expression for the viscosity of solutions and suspensions of finite concentration is derived by considering the effect of the addition of one solute-molecule to an existing solution, which is considered as a continuous medium.

IN recent years much attention has been given to the viscosity of concentrated solutions and suspensions. Guth and Simha[1] have derived the quadratic term in the concentration-dependence of the viscosity of suspensions of spherical particles by considering the hydronamic interaction of two spheres as derived by von Smoluchowski.[2] Their derivation, which leads to very involved calculations, seems to contain a numerical error, as was kindly pointed out to me by Dr. Simha.

Riseman and Ullman[3] have derived formulas for the quadratic term in the concentration-dependence of the viscosity for dumbbell, rigid rod, and flexible chain macromolecules.

Burgers and, independently, Saito[4] have developed a theory for spherical solute-molecules in which a single solute-molecule is placed in a field of flow, obtained by averaging over all possible positions of a second solute-molecule. This averaging process, however, leads to diverging integrals which make the result questionable.

In this paper we wish to point out that there is a very simple method of estimating the concentration-dependence of any quantity at higher concentrations if the result for infinite dilution is known. An analogous treatment was given by Onsager in his theory of the dielectric constant.[5]

Let us consider a suspension containing n solute-particles in a total volume V. Its viscosity is a function of the concentration n/V and will be indicated by $\eta(n/V)$.

Let us now consider the effect of adding one more solute-particle. If such a particle were added to a pure solvent of volume V we could immediately give an expression for the viscosity-increase. If the solute-particle has a volume v_0 and a spherical shape, the viscosity would be multiplied by Einstein's well-known factor $(1+2.5v_0/(V+v_0))$.

Let us now assume this factor to be approximately valid for the result of the addition of one solute-molecule to an already existing solution containing n solute-molecules. This means that this solution is represented by a pure solvent of the same viscosity. The assumption would be exact if the n solute particles already present were much smaller than the one added. In our case, however, the substitution of a continuum for the solution is an approximation. In this way we obtain the difference-equation,

$$\eta\left(\frac{n+1}{V+v_0}\right)=\eta\left(\frac{n}{V}\right)\cdot\left(1+\frac{5}{2}\frac{v_0}{V+v_0}\right),$$

which leads to

$$d\eta/dc_v=5\eta/2(1-c_v);$$

where c_v is the concentration by volume nv_0/V and therefore

$$\eta=\eta_0/(1-c_v)^{5/2}.$$

By expansion we find

$$\eta_{sp}/c_w=[\eta]+0.7[\eta]^2c_w+0.42[\eta]^3c_w^2+\cdots,$$

where η_{sp} is the specific viscosity, $[\eta]$ the intrinsic viscosity, and c_w the concentration by weight. For spheres $[\eta]=2.5v_0/M$, where M is the mass of a sphere.

This equation agrees quite well up to the term indicated with very careful measurements on asphalt-emulsions by H. Eilers.[6] Eilers represents his measurements by a formula

$$\eta=\eta_0\left\{1+\frac{1.25c_v}{1-(c_v/0.78)}\right\}^2.$$

This is equivalent to

$$\eta_{sp}/c_w=[\eta]+0.76[\eta]^2c_w+0.64[\eta]^3c_w^2+\cdots.$$

The method discussed here for spherical molecules may of course be applied to any kind of molecules.

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[5] L. Onsager, J. Am. Chem. Soc. **58**, 1986 (1936).

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