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EXPERIMENTS ON THE EFFECTIVE VISCOSITY OF CONCENTRATED SUSPENSIONS OF SOLID SPHERES

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Abstract—The effective viscosity of concentrated suspensions of solid spherical particles was determined experimentally, under zero shear conditions, by means of a convection experiment. For solid concentrations in the range 30–50 per cent the results verify extrapolations to low shear rates of previously reported conventional viscosity measurements, and suggest that, in this range of concentrations and at low shear, a suspension of neutrally buoyant spheres behaves effectively as an equivalent Newtonian fluid.

1. INTRODUCTION

It is well known, that significant differences exist among the reported values for the effective viscosity of concentrated suspensions containing spherical particles (cf. the sets of data by Rutgers 1962, Thomas 1965, Chong, Christiansen & Baer 1971 and Krieger 1972) which appear to depend on the measuring technique and the design of the experimental apparatus, as well as on the shear conditions prevailing during the course of the experiments. Questions have been raised, therefore, as to whether these data truly represent an intrinsic property of the suspension or whether they reflect to some extent the particular flow configuration existing in each experiment. From the practical point of view this is also an important matter because the empirical and semi-empirical models that are used to predict effective viscosities of concentrated suspensions invariably contain a parameter whose value must be adjusted with the aid of experimental data; hence, such a parameter cannot be uniquely determined but will depend on the particular set of data that are being used. Similarly, the asymptotic theoretical model of Frankel & Acrivos (1967) is also not complete in that it requires knowledge of the maximum volume fraction of the solids in the suspension which also varies from one type of experiment to the next. Clearly then it would be desirable to carry out the experiment under conditions of essentially zero shear, where the influence of many extraneous factors arising from the presence of a bulk motion would be minimized.

It is already well established both theoretically and experimentally that, when heated from below and cooled from above, a stagnant horizontal layer of a Newtonian fluid will become unstable and begin to transfer heat by free convection when the Rayleigh number

$$Ra = \frac{\beta g \Delta T d^3 \rho^2 c_p}{k\mu}$$

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exceeds a certain critical value which, for the case of isothermal top and bottom surfaces, depends only on the aspect ratio of the layer, d/l* (see for example Davis 1967, Catton & Edwards 1967). Thus, if q, the heat flux across the fluid layer is plotted versus ΔT, the slope of the resulting curve has a discontinuity at the point of instability owing to the appearance of the additional (convective) mode of heat transfer. Consequently then, for a fluid for which all but one of the properties comprising the Rayleigh number are known, the location of this critical point can serve to determine the remaining parameter.

Using this approach, Liang & Acrivos (1970) recently obtained the zero-shear viscosity of certain non-Newtonian fluids and showed that their results were comparable to those arrived at by conventional techniques (e.g. by means of a Weissenberg rheogoniometer). It thus seems reasonable to suppose that this method could also be applied to layers of concentrated suspensions of spherical particles, and thereby yield values for their bulk effective viscosities, provided, of course, that the other bulk properties of the suspension appearing in Ra can be predicted or measured independently.

The main advantage of this experiment derives from the fact that, at the critical point, the fluid layer is essentially stagnant and hence the measured effective viscosity, being independent of any ambient motion or of the shape of the apparatus, should represent an intrinsic property of the suspension. The remaining portion of the heat flux vs ΔT curve could then be used to test the extent to which this viscosity is affected by a modest increase in the rate of strain, thereby providing a qualitative indication of whether or not the suspension behaves as a Newtonian fluid in this range of low shear. The disadvantage of such a technique stems, of course, from the expected long duration of convection experiments and from the fact that the result will depend on the many other physical parameters entering into the expression for Ra, which, as was indicated above, must be obtained independently.

2. THE EXPERIMENTAL SET-UP

The set-up for the convection cell was similar to that reported by Liang (1969), though of different dimensions. It consisted of two $6 \times 6 \times 2\frac{1}{2}$ in. ($0.152 \times 0.152 \times 0.063$ m) aluminum blocks surrounded by thin plastic walls and separated by replaceable thin plastic spacers. The surfaces of the blocks were carefully machined and black anodized to prevent corrosion. The lower block was heated from below by a 10 Ω resistive wire which was spread and embedded between mica sheets to ensure a uniform temperature profile at the upper surface of the block. The thickness of the block served the same purposes. The upper block was cooled by passing water through copper channels connected to its top such that, at sufficiently high water flow rates, the readings at various locations on its lower surface were indistinguishable. The walls of the cell were built out of a $\frac{3}{8}$ in. (0.095 m) transparent P.V.C. sheet which resists most of the common organic solvents. These walls were connected to the lower block by means of reinforced polyester bolts to minimize heat losses through them. Teflon sheets were used as gaskets. The spacers were cut from P.V.C. in different heights to enable easy change and determination of height. Insulation was provided by

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* Here d is the thickness of the layer and l its horizontal dimension, while β, ρ, c_p, k and μ are, respectively, the thermal expansion coefficient, the density, the heat capacity, the thermal conductivity and the viscosity of the fluid. Also ΔT is the temperature difference across the layer and g is the gravitational acceleration.

having wool in the bottom and by means of an 8 in. (0.203 m) thick layer of foamed Polystyrene placed all around the experiment.

The heating element was fed from a d.c. power supply and the maximum power input did not exceed 10 W. The current was measured with a milliammeter with a reading error of less than 1 per cent, and the voltage across both sides of the heating element read by a digital voltmeter. Cooling water was supplied from a constant temperature bath in which the temperature was controlled by continuous refrigeration and regulated heating. Both the heat input and the cooling water temperature served to regulate the conditions under which the cell operated.

The temperature was measured along the diagonal of the square surface of each block by means of a digital thermometer attached to thermistor probes inserted close to the surface. Temperatures at the lower (hot) surface were determined with an error of ± 0.02 deg and at the upper (cold) surface with the accuracy of the meter (± 0.01 deg). These are relative to temperature differences across the sample of the order of 5.5 deg. The room temperature was read at the edge of the insulation because, as will be seen below, it served to estimate the heat losses in the system.

The suspended materials consisted of two kinds of Polystyrene spherical particles. The early experiments were performed with spheres of mixed size (0–500 μm) made of linear Polystyrene* while, later on, batches of Cross-linked Polystyrene† spheres were used having narrow size distributions (45 ± 10 per cent and 115 ± 10 per cent μm). These sizes are in the range which ensures the absence of any effects due to Brownian motion while still allowing the suspension to be considered as an equivalent homogeneous material.

The suspending fluids were mixtures of Ucon Oils‡ and Butyl alcohol. Two types of oils were used, 50-HB-2000 and 75-H-450, to provide various mixtures with a density close to the nominal density of the spherical particles (1.05), but with different ambient viscosities such that the resulting effective viscosity of the suspension, for all solid concentrations, was kept at about 10 P.

The remaining properties appearing in the Rayleigh number were determined as follows: The bulk thermal expansion coefficient of the suspension was set equal to the volume fraction of the fluid times its value for β, which varied from 7.8×10^{-4} to 8.75×10^{-4} in c.g.s. units at the average temperature of the experiment (i.e. the temperature in the middle of the layer). This assumed that the particles were essentially incompressible. The bulk thermal conductivity for the stationary suspension was obtained from the experimental data using that part of the heat flux vs ΔT curve where conduction was the sole mode of heat transfer. We note parenthetically that a recent study of the effective conductivity of sheared dilute suspensions of spherical particles (Leal 1973) shows that at low particle Péclet numbers (as was the case in this experiment) the contribution of the shear is minute. The measured conductivities were then assumed to apply over the entire range of Rayleigh numbers encountered in this study.

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* Product of Sinclair-Kopper Co., Pittsburgh, PA.

† Product of Diamond Shamrock Co., Redwood City, CA.

‡ Union Carbide Polyglycol oils.

Measurements of the heat capacity for the individual phases introduce one of the more severe experimental errors in the evaluation of *Ra* because the scatter in such calorimetric determinations amounts to about 10 per cent. In the various runs the liquid phase heat capacity ranged from 1884 to 2386 J/kg deg at the average temperature of the experiments, while that of the solids was measured as 1214 J/kg deg.* The bulk heat capacity was then estimated to be the volumetric average of the two quantities. In all cases, care was taken to render the density of the fluid as close to 1.05 as possible in order to minimize the rate of settling (or of rise) of the particles. It is, however, of little advantage to attempt to reduce the difference between the densities of the two phase below about 10^{-2} in view of the relatively large temperature differences across the layer and the differences in the thermal expansion coefficients of the two phases.

3. EXPERIMENTAL PROCEDURE

The suspension was prepared outside the cell at the desired concentration, mixed well, and left to settle for 24 hr. This ensured the disappearance of air bubbles which otherwise might have affected the thermal and rheological properties of the medium. Before being poured into the cell, the suspension was again mixed gently to disperse the particles homogeneously. The cell, which had been leveled beforehand, was then filled with the suspension and, immediately thereafter, heating of the bottom block and cooling of the top was begun. The conditions under which the cell operated were determined by the heat supply and by the temperature of the cooling water.

Each heat flux vs ΔT curve was established with a constant average temperature in the cell (to within ± 0.3 deg). In view of the fact that a thermal steady state was achieved only after 12–24 hr and that one to three steady state measurements were required before the average temperature approximately equaled the desired value, a final reading for a point on the heat transfer curve required 1–3 days, and a full curve approximately 2 weeks. This long duration together with unavoidable buoyancy effects acting on the individual particles presented of course a major obstacle. Thus, it was found impossible to construct the heat transfer curve starting at the pure conduction region because, by the time the expected critical point was reached, the mixture had already separated into a tightly-packed suspension and a clear fluid layer, thereby inhibiting any possible motion. The experiments had to start, therefore, well into the convection regime where sedimentation was retarded by the presence of finite circulation velocities. The additional points on the curve were then obtained by reducing the heat input until the state of pure conduction was achieved. In contrast to experiments with pure fluids, the curve did not retrace itself upon reversal of the process, but instead followed the conduction line even for temperature differences much greater than the critical. The inability to construct the heat flux vs ΔT curve by approaching the critical point from the conduction region is of course unfortunate because, whereas such a procedure would have ensured a random distribution of particles in the fluid at the

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onset of the instability, the fact that the experiment began in the convection regime probably introduced into the layer an ordered structure in the form of rolls, thereby affecting to some extent the uniformity of the solids concentration at the critical point.

In spite of the heavy insulation, it was found that heat losses to the surroundings could amount to a few per cent of the heat input. These were corrected in the following manner: it was assumed that most of the heat losses occurred through the lower block, in which case they should have been proportional to the difference between the temperature of the lower block, T_h, and that of the room T_r. The coefficient of proportionality was found by performing conduction experiments with a known fluid, without supplying any electrical power but by cooling the upper block. It was then assumed that, at steady state, the heat leaking into the system, proportional to $T_r - T_h$, exactly equaled the heat conducted across the fluid layer. In this way, after varying the cooling water temperature and the room temperature, an average value was estimated for the heat losses equal to $7.5 \times 10^{-3} (T_h - T_r) \pm 10$ per cent J/kg deg.

4. RESULTS

Experimental results are reported here for suspensions with solid concentrations equal to 30, 40 and 45 per cent. Runs 1 and 2 were carried out at an average temperature of 18.9 deg; in all other cases it equaled 20 deg. The results are summarized in table 1 and a typical heat transfer curve is shown in figure 1, where the data collected for run No. 5 are presented. The discontinuity of the slope at the critical point is clearly evident although, of course, there is some uncertainty as to its exact location. This, together with the uncertainty in the value for the heat capacity, are the major sources of any inaccuracies in the results. The isolated point in figure 1 represents the last measurement for this run, obtained by *increasing* ΔT

Table 1. Summary of experimental results.

Run	Fluid		Particles		Gap (mm)	Suspension viscosity (P)	$\frac{\mu_{\text{eff}}}{\mu}$
	Composition	Viscosity (P) (at exp. temp.)	Concentration (%)	Size (μm)			
1	Ucon oil 50-HB-2000	14.16	30	mixed 0–500	25.02	45.3	3.2
2	Ucon oil 75-H-450 + butanol	1.60	40	mixed 0–500	25.02	20.8	13.0
3	Ucon oil 50-HB-2000	12.21	30	100–130	25.02	41.6	3.4
4	Ucon oil 75-H-450 + butanol	1.148	40	40–50	25.02	15.5	13.5
5	Ucon oil 75-H-450 + butanol	1.08	45	40–50	25.02	35.7	33.1
6	Ucon oil 50-HB-5100	29.91	0		25.02	29.91	1.0
7	Polybutene 16	41.7	0		39.98	41.7	1.0
8	Ucon oil 75-H-450 + butanol + methoxyethoxy ethanol	2.27	50	40–50	39.98	240–280	110–130

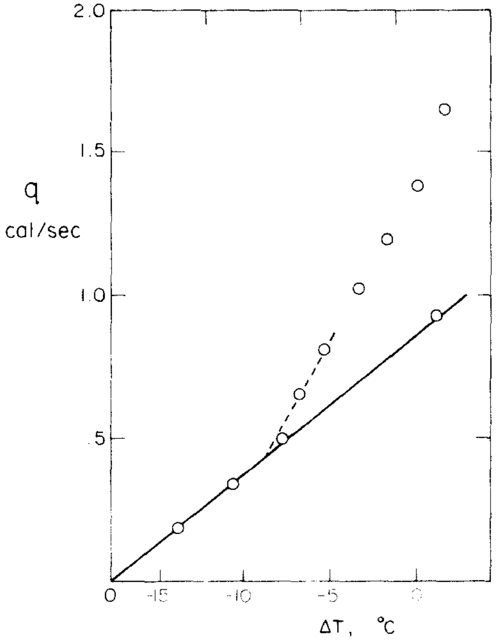

Figure 1. A typical set of data for heat flux vs ΔT. The results correspond to run 5 where the solid concentration was 45 per cent and the effective viscosity 33.1 P.

from its lowest value, and clearly demonstrates a hysteresis effect since it corresponds to a state of pure conduction at a temperature difference twice the critical.

Figure 2 contains all the experimental points from runs 1–6. The continuous curve was obtained from run 6 in which the cell was filled with a pure Newtonian fluid, Ucon oil No. 50-HB-5100. It is evident that, in spite of the experimental uncertainties discussed earlier, all the results closely follow the Newtonian curve in this region especially when one takes into account that the appropriate Rayleigh numbers beyond the critical were calcu-

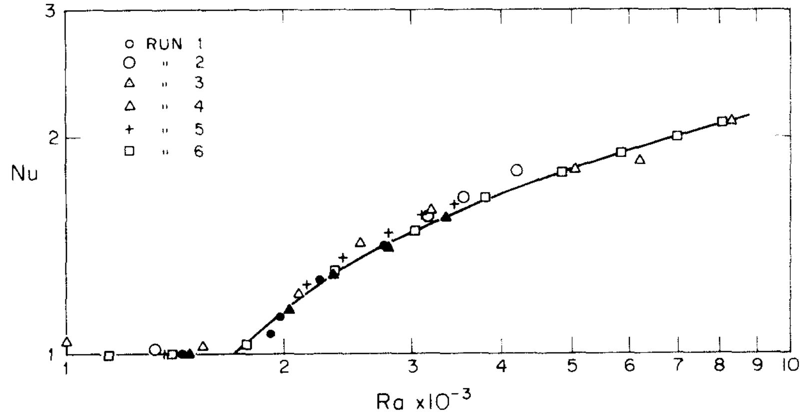

Figure 2. The Nusselt number as a function of the Rayleigh number for all runs.

lated using the corresponding value for the effective viscosity as measured at the critical point. Of course, it would be of interest to extend the measurements to higher shear rates since, as reported recently by Hoffman (1972), discontinuities in the viscosity have been encountered when using a cone and plate viscometer, apparently owing to a change in the structure of the suspension from an ordered array to a disordered one.

The measured effective viscosities reported in table 1 are compared with Rutgers' (1962) reduced data in figure 3. Evidently, the good agreement between the two sets suggests that extrapolations of viscometric experiments to low shear can predict correctly the viscosity at this range of solid concentration. At concentrations 50 per cent and above, Rutgers indicated that no consistent data could be obtained for low rates of strain. Similarly, the low shear limit measurements reported by Kreiger (1972) extend only to a maximum of 45 per cent solid concentration. It was not surprising, therefore, that, in our case, no successful runs could be performed with solid concentrations 50 per cent and above. Perhaps this difficulty could be attributed to the relative importance of surface interactions at this range of concentrations. Besides, at these high concentrations, it is not unreasonable to regard the suspension as a porous medium where convection would be expected to occur for much larger values of ΔT than those of the present experiments. A partially successful experiment though is reported in table 1 and figure 3 (run 8). Here the convection did not start spon-

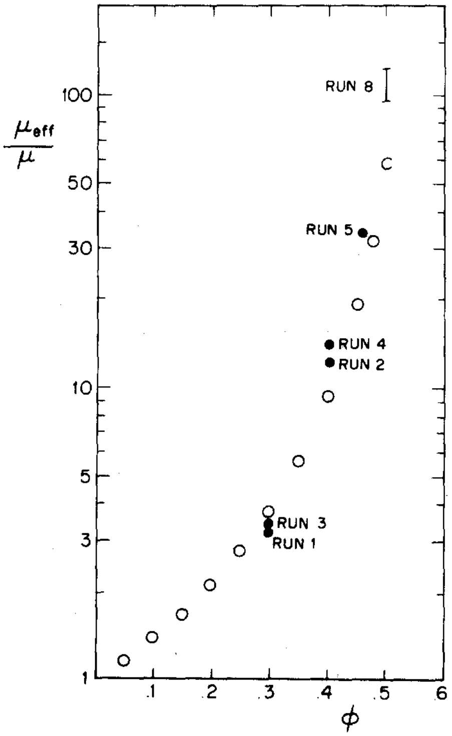

Figure 3. The effective viscosity of the various concentrations. ○—data of Rutgers (1962); ●—present results.

taneously but had to be induced by mechanical mixing. Also, the scatter of the data was much larger than in all the other cases.

It should be noted also that the experiments with a mixed size distribution yielded lower values for the effective viscosity than those with a uniform particle size at the same overall solid concentration. Although this difference is well within the experimental inaccuracy, the trend is consistent with the results of earlier studies (Ward & Whitmore 1950, Williams 1953, Eveson 1959 and, recently Chong, Christiansen & Baer 1971) where a reduction in the effective viscosity was observed due to the mixing of spheres of different sizes. This effect is expected to be more noticeable with solid concentrations of 50 per cent and above (Williams 1953).

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Sommaire—La viscosité effective de suspensions concentrées de particules solides sphériques a été déterminée expérimentalement dans des conditions de cisaillement nul par une expérience de convection. Pour des concentrations de solides dans la gamme de 30–50 pour cent, les résultats vérifient les extrapolations à des taux de cisaillement faibles de mesures de viscosité classiques précédemment rapportées, et suggèrent que dans cette gamme de concentrations et à faible cisaillement, une suspension de sphères neutralement flottantes se comporte effectivement comme un fluide équivalent Newtonien.

Auszug—Die wirksame Viskosität von konzentrierten Suspensionen kugelförmiger Teilchen wurde mittels eines Konvektionsexperiments unter schubfreien Bedingungen experimentell ermittelt. Die Ergebnisse bestätigen Extrapolationen auf niedrige Schubgrade von früher berichteten üblichen Viskositätsmessungen für feste Konzentrationen im Bereich von 30–50 N pro cwt (knapper Zentner) und regen an, daß eine Suspension von neutral schwebenden Kugeln in diesem Konzentrationsbereich und bei niedrigem Schub sich praktisch wie ein Äquivalent einer Newton'schen Flüssigkeit verhält.

Резюме Зффективная вязкость концентрированных твердых сферических частиц определялась экспериментально при нулевых условиях трения конвективным опытом. При твердых концентрациях в порядке 30–50 процентов результаты верифицировали экстраполяции низкого напряжения трения, как это сообщалось ранее при измерении обычной вязкости, и это наводит на мысль, что в таких пределах концентрации и при низком трении, поведение суспензии нейтральной плавучести является подобным поведению эквивалентной ньютоновской среды.