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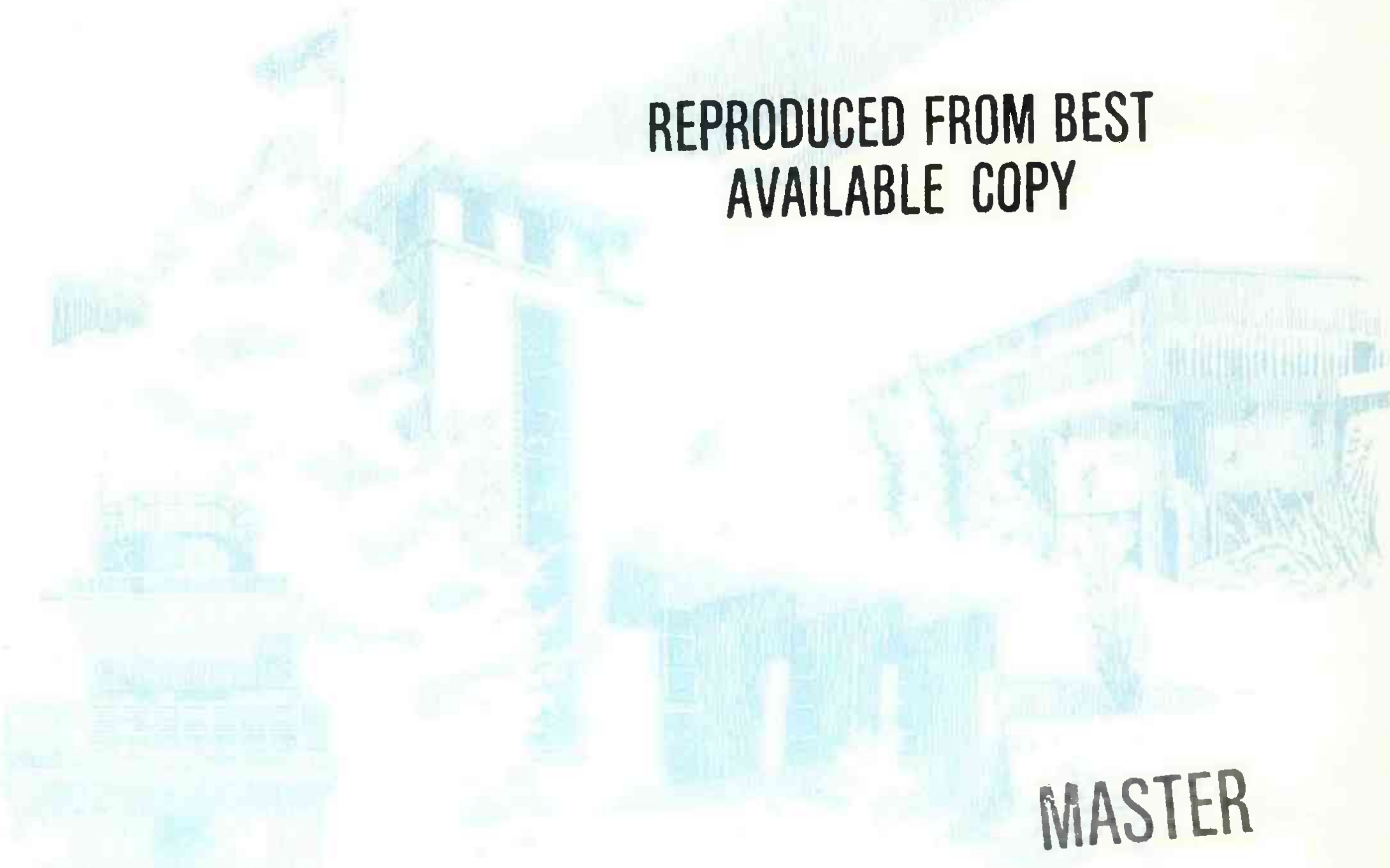

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A FALLING BALL RHEOMETER FOR OPAQUE, CONCENTRATED SUSPENSIONS

(SAND88 - 2308)

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ABSTRACT

With falling ball rheometry, we have measured the apparent relative viscosity of suspensions of large, neutrally buoyant, rigid rods in a viscous Newtonian fluid, while approximately maintaining the rods in a randomly oriented configuration. A new technique for measuring the time of flight of a ball between two positions is used. This computerized technique, based upon an eddy current detector, enables us to determine the position of a metallic (non-magnetic) ball falling through an opaque suspension, with high accuracy (less than 1.5% error). The rods for the suspensions had a nominal aspect ratio of 10 and experiments were carried out at a single volume fraction, 0.05. Two populations of rods were used having nominal diameters of 1.5875 mm and 3.175 mm. To within the errors of these experiments, suspensions from both populations had the same relative viscosity, with the overall average being 1.457. This viscosity was significantly different from that of a similar suspension (volume fraction = 0.05) of rods of nominal aspect ratio 20 and it agreed quite well with theoretical results for the viscosity of a dilute suspension of randomly oriented rods.

A. INTRODUCTION

It was recently shown that a technique historically used in the non-destructive evaluation of solids, x - ray radiography, could be coupled with high speed video to produce a powerful tool for probing the microstructural dynamics of suspensions [1-3]. This technique is capable of accurate studies of the mean properties of suspensions, and when combined with automated image analysis, it can provide microstructural information unobtainable by other means. This marriage of two technologies provides a paradigm for other possible experimental techniques which might readily be applied to the study of materials, which have only yielded to scientific investigation with much difficulty, such as highly concentrated, opaque suspensions.

As with x - ray radiography, eddy current techniques have been employed in a variety of applications associated with the testing of solids [4]. As described in the Experimental Section, when this measuring technology is combined with modern high speed electronics and a computerized data acquisition system, it can be applied to the study of fluid - like materials, regardless of their optical properties. In particular, an automated system can be developed for measuring viscosity by falling ball rheometry.

The particular systems studied using the eddy current technique were suspensions of randomly oriented rods in a Newtonian fluid. Our principal goal in these experiments was to determine the effect of aspect ratio, a_r, on the viscosity, μ, of the suspensions. Falling ball rheometry was previously shown to be a useful method for measuring properties of such suspensions [3,5,6], yielding accurate results which could be favorably compared with available theories [7-9]. One of our earlier studies focused on determining the effect of volume fraction, ϕ, on the relative viscosity, μ_r, for a fixed aspect ratio of particle, $a_r = 20$. Choosing one of the volume fractions used in those experiments, $\phi = 0.05$, in this paper we report upon measurements of the relative viscosity for two suspensions comprised of particles having $a_r = 10$. The difference between the particles was their dimensions. Our previous work [6], showed that when the diameter of the falling ball reached a critical size between the length and the diameter of the suspended particles, the apparent viscosities depended upon ball diameter, decreasing with decreasing ball diameter. The particles used in those studies were the same size, and the ball diameter was varied. Heretofore, there has been no effort to examine the effect of maintaining a constant ball size while varying the dimensions of the suspended rods.

B. EXPERIMENTAL

1. EDDY CURRENT MEASURING SYSTEM

The principal quantity measured in these experiments was the time required for a ball, falling under the influence of gravity in a cylindrical column, to pass across two reference points on the column. Knowing the distance between these points, it was possible to infer from such measurements the mean velocity of the falling ball. The technique is based upon a standard method used in non-destructive testing of solids known as the *eddy current test* [4]. A schematic of the test is shown in Figure 1. A sphere is placed in a time varying magnetic field, called the *primary* magnetic field. This induces eddy currents in the sphere which produces an additional time varying magnetic field that modifies the primary field. The field disturbance is detected by comparing the impedance of the coil containing the test sphere with the impedance of an identical, empty coil using the bridge shown in Figure 2. The two potentiometers are used to cancel out any differences in the two coils and balance the bridge in the absence of a test sphere. The primary field, which in these experiments oscillated at 100 kHz, is produced by an oscillator. The magnitude of this field is approximately 0.5×10^{-4} T, or slightly less than the earth's magnetic field. Hence, any additional forces acting on the falling ball due to the presence of the magnetic field are negligible. Since sensitivity to the sphere increases with frequency [4], a moderately high frequency is used.

The overall schematic of the experimental apparatus is shown in Figure 3. The two test coils are wrapped on a cylindrical glass column having a nominal outside diameter of 153 mm and a nominal inside diameter of 146 mm. Each coil consisted of fifteen strands of #22 diameter insulated magnet wire [Bel-sol, Beldon cable.]. These coils are sandwiched between two wraps of copper tape, or Faraday shields, which act to isolate the coils from electrostatic fields. In the absence of a sphere, the bridge is balanced to produce the null result using the potentiometers. As the sphere sediments through the column one of the coils acts as the reference coil while the sphere passes through the other coil. The change in impedance is detected by measuring the amplitude of the in - phase and quadrature parts of the signal from the bridge relative to the signal used to excite the primary magnetic field. This is done using a lock - in amplifier (Princeton Applied Research, Model 129A, Princeton, NJ) with the oscillator (Tektronix, Model FG 501A, Beaverton, OR) supplying the reference signal. The two outputs from the lock - in amplifier are acquired by a 12 bit analog - to - digital convertor (Metrabyte, Stroughton, MA) attached to an IBM AT (International Business Machines, Boca Raton, FL) microcomputer. Data acquisition and analysis software was written in BASIC employing CALL statements to routines supplied with the analog - to - digital convertor board.

2. STATIC CALIBRATION

A static calibration of the system was performed by suspending metallic balls of known diameter from a string and incrementally moving them longitudinally through the column. Measurements were made of the magnitude of the deviation from the null condition (no object present) as a function of the axial position of the ball in the column. In the vicinity of the coils, the smallest increment of movement possible was used, 1 mm. Away from the coils, larger increments were employed. Tests were performed using balls of various diameters, from 9.525 mm to 25.4 mm, and the effect of the ball not being on the axis of the column was explored.

Typical results are shown in Figure 4 for a brass ball of nominal diameter 9.525 mm. Three tests are shown in which the ball was moved along the axis of the column, one ball diameter off the column axis, and approximately 30 mm away from the wall of the column. In all cases, two peaks are observed corresponding to the balls' being in the planes of the coils. The distance between the two peaks was the same under all three conditions, 149 mm. The slight difference in the magnitude of the peaks can likely be attributed to slight non-uniformity in the induced magnetic field in the coils. The coils should ideally induce a magnitude field which is radially uniform. Figure 4 shows that such an assumption is approximately valid. There is some radial variation with the magnitude of the peaks exhibiting a local maximum at the center. Away from the center, the peak magnitude decreases slightly until near the coil where an increase is again observed. For all of the calibrations, which included balls of larger diameter, up to 25.4 mm, and made of one other material, stainless steel, the distance between peaks was found to be 149.0 ± 1.5 mm. It was concluded that even under the extreme case when the ball falls far off the axis, it is possible to calculate its average vertical velocity, v_a (in mm/s), by measuring the time of flight between the coils, t_f (in seconds), using:

$$v_a = \frac{149}{t_f} . \qquad [1]$$

3. SOFTWARE DESIGN

Computer software was designed to acquire the data from the lock - in amplifier and measure the time of flight of the ball as it passes between the two coils. The technique used to acquire the data is specific to the computer system used in these experiments, and will not be discussed further. It is sufficient to note that the net result of the data acquisition step was an array of 3000 voltage values containing points which were equally spaced in time. The strategy used to determine the locations of the two peaks, corresponding to the arrival of the falling ball at the center of the coils, is of general interest and it will be discussed further here.

Three techniques were used to determine t_f. The first involved the interaction of the user. After the data were acquired, they were plotted on the video display terminal. The data were searched numerically to ascertain the approximate location of the two peaks which, in general, are similar to those shown in Figure 4. Two markers were plotted along with the data, and the user would then visually check that the software had in fact determined the correct approximate location of the maxima. In case of a discrepancy between the visual observation of the maxima and the locations of the markers, the software allowed the user to move the markers to the approximate locations of the peaks.The time interval determined in this way constituted a first approximation to t_f.

The next stage of data analysis consisted of first performing a five point running mean average on the data in the neighborhood of the two maxima (200 points on both sides). This procedure smoothed the noise in the data resulting from noise in the electronics or, in some experiments, a lack of equipment resolution. For example, it is possible that the analog - to - digital convertor might yield the same digital representation of slightly different analog values, particularly around maxima where the signal is slowly varying. Subsequently a search of the smoothed data was performed to determine the maxima. Generally, the values obtained by these first two procedures differed by less than 3%.

The third technique consisted of fitting the data near the maxima determined after smoothing to parabolas, and then calculating the maxima of the parabolas. Various numbers of points, from 10 to 230, were routinely used for these fits. The difference in the numbers of points used for the curve fitting represented our efforts to ascertain the sensitivity of our measurements to the total number of points required for these fits. The difference between using, say, 50 and 100 points was that in the former case, all 25 points before and after the maximum would be used, while in the the latter case, all 50 points before and after the maximum would be used. In no instance did varying the number of points used for the fitting procedure represent a selection process by which spurious data points were eliminated. Except for the fewest (10 - 30) and most (170 - 230) numbers of points the results were independent (to within less than 1%) of the number of points used for the fit. This finding depended upon the total voltage excursion during an experiment, i.e., the steepness of the maxima.

A comparison of the three techniques was made by dropping balls of diameters ranging from 9.525 mm to 19.05 mm in a fluid and determining the time of flight between the coils. The results showed that the technique using the running mean and that using the fit to a parabola yield the same results. Over eleven experiments, the ratio of the mean velocities determined by these techniques was 1.000 with a 95% confidence limit of 0.008. From this study it was concluded that the two numerical data analysis techniques yield the same results and hence they could be used interchangeably.

4. SUSPENSIONS AND ANCILLARY EQUIPMENT

The particles and fluid were similar to those used in previous studies [3,5,6]. Two populations of particles were manufactured from continuous polymethyl methacrylate rods, having nominal diameters of 1.5875 mm and 3.175 mm. The rods were individually cut to have nominal aspect ratios of ten. A statistical study of 25 rods from each population showed that the 1.5875 mm diameter rods had a mean aspect ratio of 9.88 while the 3.175 mm diameter rods had a mean aspect ratio of 10.01. The standard deviations were 0.44 and 0.13, respectively. As with the earlier studies, these distributions were considered to be sufficiently narrow that the particles were monodisperse [3,5,6]. Further, both fiber flexibility, based upon the criterion of Forgacs and Mason [10], and rotary Brownian motion were negligible [5].

The fluid was a mixture of polyalkylene glycol (UCON-HB-9500 Union Carbide Corporation, Danbury, CT), 1,1,2,2 tetrabromoethane (Eastman Kodak Company, Rochester, NY), and Tinuvin 328 (Ciba - Geigy, Ashley, NY). Tetrabromoethane was added to the polyalkylene glycol to match the density of the mixture to that of the rods, 1181.8 kg/m^3 , at 20.00°C. A small quantity, ≤ 0.2 wt%, of an antioxidant (Tinuvin) was added to the mixture to prevent the breakdown and discoloration of the tetrabromoethane by ultraviolet light.

Before the suspensions were made, the rods were cleaned with soap and water, rinsed with deionized water, and dried. The suspensions were made by weighing out the required quantities of fluid and rods on a Mettler PC8000 balance. The balance was accurate to ±0.1 g, but was limited to a weight of 8 kg. The suspensions required 16-24 kg of fluid, resulting in an inaccuracy of the added weights of each suspension less than or equal to 0.3 g. The uncertainty in the volume fraction was determined to be less than 5×10^{-5} and is contained within the symbols representing the data points on all figures in this paper.

The suspensions were contained in a cylindrical glass column 501 mm high and 153 mm outside diameter and 146 mm inside diameter. The top of the column had a removable cover with guide tubes at the center to ensure that the falling ball was dropped along the center line. The guide tubes were fabricated from Teflon rods and had bore diameters slightly greater than the diameters of the falling balls.

There was a thin slot in the cover from the guide tube to the wall of the column for a stirrer. The stirrer, which was used to randomly orient the suspension, was a long brass rod with a handle on the top and eight short prongs on the bottom. We mixed the suspension thoroughly before each ball was dropped by briskly raising, lowering, and twisting the stirrer. Care was taken when stirring to avoid the introduction of air bubbles. A few colored test rods were placed in one suspension and their orientations were photographed after stirring. Using data from several such photographs, we applied the technique of Givler [11] for ascertaining the degree of randomness. In his analysis, orientation is measured by a scalar, f_p , which varyies between zero and one where zero represents a completely random orientation distribution. In these experiments, we found that this scalar tended toward zero as more data (photographs) were used in the analysis. In all, approximately 100 rods were used to determine that $f_p = 0.06$. From this we concluded that the suspensions were randomly oriented.

The cylindrical columns were placed in an insulated water tank controlled by a constant temperature circulator (Lauda Company, Brinkman Instruments, Westbury, NY). The columns were thermally equilibrated for at least 24 hours before any experiments were performed. The temperature of a suspension was measured with a thermocouple probe to within ±0.01°C (Instrulab Inc., Dayton, OH). The temperature was measured at the beginning of an experiment and after each set of 5 individual measurements. The probe allowed measurement of the temperature at different points in the suspension to ensure thermal homogeneity. Falling-ball experiments were performed when the measured temperature was within ±0.10°C of the desired set point (20.00°C).

The balls used in these experiments were brass ball bearings (Anti-Friction Bearing Manufacturers grade 200, Hoover Universal Company, Ann Arbor, MI). They were placed in the water bath and thermally equilibrated to the temperature of the suspension prior to the experiments. Balls having six different nominal diameters were used: 3.175, 6.35, 9.53, 12.7, 15.9, and 19.1 mm. The actual diameter of each ball was measured to ±0.003 mm and the weight of each was measured to ±0.0002 g.

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C. RESULTS

1. SUSPENDING FLUID

Prior to performing the measurements with the suspensions, falling ball experiments were performed using the suspending fluid in order to assess the accuracy of the eddy current system and to ascertain the viscosity of the suspending fluid itself. The data were analyzed according to the zero Reynolds number form of the relationship between the mean terminal velocity of the ball, v_a, and the viscosity, μ, which corrects for wall effects [12], viz.,

$$\mu = [1 - 2.104\,(d/D) + 2.09\,(d/D)^3 - 0.95\,(d/D)^5 + O(d/D)^6]\,\frac{d^2(\rho_s - \rho)g}{18 v_a} \qquad [2]$$

where d is the diameter of the falling ball, D is the diameter of the column containing the suspension, ρ_s is the density of the ball, ρ is the density of the fluid, and g is the gravitational constant. Equation [2] holds for the case when the ball falls along the center line of the cylindrical glass column [13], and when the characteristic Reynolds number, $N_{Re} = \frac{\rho v_a d}{\mu}$, was in the creeping flow regime. In the suspending fluid, the largest balls had Reynolds numbers greater than 0.1 but less than 0.14. However, to within the accuracy of the measurements, the low Reynolds number equation could also be used to analyze the data from these experiments.

Figure 5 presents the result of using Eq. [2] to analyze our data. When boundary effects are not properly considered, and the standard Stokes formula [14] for a sphere falling through an unbounded fluid is used to calculate the viscosity, the viscosity is found to be strongly dependent upon the ball diameter increasing by approximately 20% when the ball size increases by a factor of two. Applying Eq. [2] has two effects. Firstly, the dependency upon the ball diameter becomes negligible. There is a small decrease in the viscosity with b l diameter, but this is within the error of these experiments, as indicated by the 95% confidence bounds shown in Figure 5. Secondly, the average value of the viscosity, 11.42 Pa-s, becomes much closer to the value obtained previously for the same fluid, 11.95 Pa-s [5]. The slight difference being likely due to aging effects which occurred over the two year time span between the two sets of experiments.

Table 1 gives the comparison between the results obtained from the eddy current measurements and those obtained using an optical technique [3,5,6] to determine the velocity of the falling balls. For each ball diameter, five individual experiments, simultaneously employing both techniques, were performed and the data were analyzed using Eq. [2]. These data clearly show that the two techniques yield comparable results with the optical method consistently yielding a lower value of the viscosity. The average difference, 1.49%, was not considered to be sufficiently large as to warrant further investigation. As will be shown in the upcoming Sections, such a difference is generally smaller than the statistical variation in the suspension data. Due to the uncertainties in both techniques, particularly in obtaining an accurate mapping from camera coordinates to laboratory coordinates with the optical technique, neither result is more accurate than the other. Indeed, given the absolute nature of the calibration of the eddy current instrument, values obtained using it are likely to be the more accurate.

2. SUSPENSIONS

The results for suspensions of rods are presented in Table 2. Rather than examining the numerical values, it is useful to examine first the dependency of the velocities of the falling balls upon their diameters. At low Reynolds numbers, the velocity of a sedimenting ball in a Newtonian fluid of infinite extent increases as the square of its diameter. Figure 6 shows v_a versus d^2 for two suspensions both being composed of particles nominally having $a_r = 10$ at a volume fraction of 0.05. The difference between the two suspensions was the size of the rods. As can be seen from Figure 6, this factor made no systematic difference in the results. For the larger balls, the proportionality between the velocity and the square of the diameter breaks down as is easily seen by comparing the data with the straight line lying through the two data points for the smallest diameter balls. For the largest diameter balls, there are dramatic differences between the experimental and expected values, e.g., when d = 1.905 mm, this deviation reaches 33%.

Our earlier studies [3,5,6] indicated that such deviations could be assigned to wall effects of the Faxén type, as described by Eq. [2]. To test this with the present data, the velocities in Figure 6 were analyzed using Eq. [2] with the result being shown in Figure 7, and given in Table 2. Plotted along with these data is the best fit straight line which passes through the origin. Figure 7 demonstrates that the corrected velocities, i.e., the measured velocities divided by the term in square brackets in Eq. [2], are linear with the ball diameter squared. It is important to note that this result is independent of the diameter of the suspended particle for all but the 3.175 mm diameter balls.

D. DISCUSSION

1. RELATIVE VISCOSITY

The results for the suspending fluid demonstrate that the eddy current instrument, developed for this study, can be used to measure the viscosity accurately and reproducibly. For the randomly oriented suspensions, we have verified that in the mean they can be considered as Newtonian fluids. That is, the added drag on the ball due to the walls of the container are accurately described by Eq. [2], and a single viscosity coefficient characterizes the drag on the ball due to the suspension. As with our previous studies [3,5,6], such a conclusion applies to the very special case of a suspension of randomly oriented particles which are only slightly disturbed by the falling ball. Consistent with these earlier studies, we can therefore use the mean velocity data given in Table 2 and the viscosity data presented in Table 1 to calculate the average relative viscosities for each ball size. These data, along with their 95% confidence limits are also presented in Table 2. To within the uncertainties in these experiments, the relative viscosities (except possibly that found for the 3.175 mm diameter particles using the 3.175 mm diameter balls) are independent of the diameters of the falling balls. This is also shown in Figure 8, where we have plotted the relative viscosity as a function of the dimensionless ball diameter: d/d_{rods}. The form of this plot is typical of those found in our previous study [6]. For the largest ball diameters, there is no statistically significant variation in the average relative viscosity. For the smallest diameter ball used with the largest diameter rod, there appears to be a decrease in the relative viscosity from the average value obtained from the analysis of the data for the other balls. Caution must be exercised in drawing too much significance from this data point even though it represents ten individual experiments. Despite its mean being lower than all of the other data, the

95% confidence limits on this point overlap those of all the other data. Hence, it is not possible to conclude that for $d/d_{rods} \leq 1$, the relative viscosity decreases.

Consistent with our earlier work [3,5], we avoid complications arising from a dependency in the measured relative viscosity upon ball diameter by eliminating data from all but the four largest ball sizes and calculating the average relative viscosities. These results are presented in Table 2 under the heading of **composite.** As might be expected from the previous discussion, the results obtained using the two suspensions cannot be distinguished. The relative viscosity of the suspension of 1.588 mm diameter rods, which had an average aspect ratio of 9.88, was 1.433 ± 0.021 whereas the relative viscosity of the 3.175 mm diameter rods, $a_r = 10.01$, was 1.470 ± 0.032. The difference in aspect ratio might account for some of the variation. However, it is important to recall that the rods themselves are not uniform and that to within one standard deviation, they have the same aspect ratio.

While suspensions formed using the two populations of particles are found to have the same mean properties, a closer examination of the variations about the mean indicate some differences between them. Figure 9 shows the frequency distribution for data obtained using balls of diameter 19.05 mm in both suspensions. The data for the rods of smaller diameter are seen to be much more narrowly distributed. Ninety percent of these data have velocities between 6.2 and 6.6 mm/s. The data for the 3.175 mm diameter rods shows a more broader distribution, with less than 33% lying in that velocity range. A quantitative measure of the spread in the distributions is their variance, σ. For the suspension of 1.588 mm rods this was 1.17 whereas for the suspension of the larger rods, $\sigma = 6.09$. These results clearly demonstrate that there is not a measurable effect on the mean properties of the suspension when different sized rods are used. However, the variance, which can be extracted from mean velocity data is affected. More experiments would be required for each ball diameter to ascertain the details of the frequency distribution. At this time, these results are considered to be preliminary, but representative of a general trend found in all of the data gathered to date.

2. COMPARISON WITH THEORY

Our previous work led to the surprising conclusion that theories predicting the viscosity of suspensions in which Brownian forces are dominant could accurately predict the results from falling ball experiments using suspensions of large rods in which Brownian forces are negligible [5]. Such a result was anticipated by the work of Haber and Brenner [15]. They demonstrated that for bodies of revolution, early theories for suspensions, which described Brownian forces by specifying that particles in shearing flow remain randomly oriented, were consistent with later theories in which Brownian forces were rigorously modeled. An additional and equally surprising conclusion of our earlier study was that the suspensions were dilute, i.e., $\mu_r \propto \phi$, at volume fractions much higher than previously considered [16,17]. Rods having nominal aspect ratios of twenty had a linear viscosity - volume fraction relation for $\phi \leq 0.12$. This result was found to be consistent with recent molecular theories for rigid rodlike macromolecules [18-20] although at this time no rigorous basis for such a comparison has been established.

In the present study, these two conclusions provide a basis for comparing our results with the theory of Brenner for the rheology of dilute suspensions of rods subject to strong Brownian motion [5,7,15]. This comparison is given in Table 3 both in terms of each suspension and the overall averages from both suspensions. Examining first the similarity between the relative viscosity data and Brenner's prediction [7] quite good agreement is found. The largest error is for the suspensions of 1.588 mm particles, where the deviation is 2.1%. This comparison, however, masks some of the discrepancy between our results and Brenner's theory. A more exacting comparison is that between the experimental and theoretical values of the intrinsic viscosity, $[\mu]$. Since we have obtained relative viscosity data for a single concentration, it is not possible to apply the usual techniques of suspensions rheology to obtain $[\mu]$, i.e., plotting $(\mu_r - 1) / \phi$ versus ϕ and taking the limit as $\phi \to 0$. It is possible to use our data to calculate $[\mu]$ using

$$\mu_r = 1 + [\mu] \phi \qquad [3]$$

if we assume, consistent with our earlier findings [5], that our suspensions are dilute. The results of this calculation are found in Table 3 along with the predictions of Brenner [7]. In this form the agreement between experiment and theory is not as close as when the data were expressed in terms of μ_r , although, at worst, the discrepancy is only about 12%. Such a disagreement can readily be assigned to either experiments or the theory. While by the measure available to us, the suspensions tended to be random [11], no technique was available to determine whether this condition was met prior to each experiment. Brenner's theory was derived using approximation techniques drawn from slender body theory which are likely to break down for particles having aspect ratios of ten.

Combining the data for the intrinsic viscosity given in Table 3 with the results of our earlier study for rods having an aspect ratio of twenty [3], it is possible to begin to construct a curve which describes the dependency of the relative viscosity upon the aspect ratio for a fixed concentration. Figure 10 shows such a plot for the two sets of data which are currently available. We have also shown the predictions of Brenner's theory and, to represent the $a_r = 1$ data, we have used the experimental results for spherical particles. We view the inclusion of this point as a convenient reference. It is possible that the intrinsic viscosity of a suspension of rods having $a_r = 1$ will be different from the results for spheres.

E. CONCLUSIONS

The computerized data acquisition and analysis system for falling ball experiments developed around the eddy current detection technique has been shown to produce accurate results. This device provides excellent data on the averaged properties of both transparent and opaque materials, and can be implemented at minimal cost, less than $10,000. Further, it is likely that a more advanced system could be used to determine the time dependent motion of the falling sphere, and, hence more detailed microstructural information.

The present studies on suspensions add to the growing body of evidence that falling ball rheometry offers unique insights into the properties of suspensions of rods [3,5,6]. Even when the measuring device, viz., the falling ball, is roughly the same size as the suspended rods, the relative viscosity determined by averaging the results of many experiments is the same as that measured using balls which are much larger than the suspended particles. The suspension behaves as if it were a Newtonian liquid having an effective viscosity in excess of that of the suspending fluid and which is greater than the viscosity measured for suspensions of rods using conventional rotational rheometry [21,22]. The large difference between the relative viscosities measured by the falling ball technique and those measured using shearing flows [5] can likely be ascribed to the orientation induced by the prescribed shearing flow. With the falling ball technique, the rods are only slightly perturbed by the passage of the ball through the suspension , and an initially isotropic suspension remains substantially so during a measurement. This conclusion is supported by the agreement between our results and an existing rigorous theory for the viscosity of a suspension of randomly oriented rods [7]. The data are consistent with the discovery of the extended dilute regime [5]. However, it is readily admitted that no firm conclusions can be drawn on this point without obtaining more data over a wider range of concentrations.

Obtaining an accurate representation of the surface defined by the relative viscosity, volume fraction, and aspect ratio, i.e., $\mu_r (a_r,\phi)$, has eluded many previous investigators [17]. Recent results would indicate that some progress has been made for shearing flows [16,21,22], however, all existing studies lack a rigorous theoretical benchmark against which data can be evaluated. The present study provides more evidence that the falling ball technique is a powerful method for obtaining accurate and theoretically consistent results for suspensions of randomly oriented rods. It appears that the technique could be broadened by inducing some mean orientation in the rods prior to dropping the ball and use the technique to measure the viscosity of oriented suspensions, obtaining results comparable to those measured in shearing or extensional flows.

F. ACKNOWLEDGEMENTS

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G. LIST OF REFERENCES

1. L. A. Mondy, A. L. Graham, A. Majumdar and L. E. Bryant, Jr., *Int. J. Mult. Flow,* **12,** 497 (1986).

2. L. A. Mondy, A. L. Graham and J. L. Jensen, *J. Rheol.,* **30,** 1031 (1986).

3. A. L. Graham, L. A. Mondy, M. Gottlieb and R. L. Powell, *Appl. Phys. Letts.,* **50,** 127 (1987).

4. R. C. McMaster, *Nondestructive Testing Handbook Vol. II,* Ronald Press Co., New York, NY, 1963.

5. W. J. Milliken, M. Gottlieb, A. L. Graham, L. A. Mondy and R. L. Powell, *J. Fluid Mech.* to appear (1989).

6. W. J. Milliken, L. A. Mondy, M. Gottlieb, A. L. Graham, and R. L. Powell, submitted for publication (1989).

7. H. Brenner, *Int. J. Mult. Flow,* **1,** 195 (1974).

8. R. G. Cox, *J. Fluid Mech.,* **44,** 791 (1970).

9. R. Simha, *J. Phys. Chem.,* **44,** 25 (1940).

10. O. L. Forgacs and S. G. Mason, *J. Coll. Sci.,* **14,** 457 (1959).

11. R. C. Givler, *Quantitative Description of In Situ Fiber Orientation State with Image Processing Techniques,* **SAND86-0075,** Sandia National Laboratories, Albuquerque, NM (1986).

12. T. Bohlin, *Trans. Roy. Inst. Tech.* (Stockholm), No. 155 (1960).

13. L. A. Mondy, A. L. Graham, J. Miller, N. Wagner and W. Cook, submitted for publication (1989).

14. G. K. Batchelor, *An Introduction to Fluid Dynamics*, Cambridge University Press, Cambridge, UK (1967).

15. S. Haber and H. Brenner, *J. Coll. Intf. Sci.*, **97**, 496 (1984).

16. S. M. Dinh and R. C. Armstrong, *J. Rheol.*, **28**, 207 (1984).

17. E. Ganani and R. L. Powell, *J. Composite Matl.*, **19**, 194 (1985).

18. G. T. Keep and R. Pecora, *Macromol.*, **18**, 1167 (1985).

19. J. J. Magda, H. T. Davis, and M. Tirrell, *J. Chem. Phys.*, **85**, 6674 (1986).

20. I. Bitsanis, H. T. Davis, and M. Tirrell, *Macromol.*, in press (1988).

21. T. Kitano, T. Kataoka, and Y. Nagatsuka, *Rheol. Acta*, **23**, 20 (1984).

22. E. Ganani and R. L. Powell, *J. Rheol.*, **30**, 995, (1986).

LIST OF FIGURES AND TABLES

FIGURES

suspended particles; (◆) 1.588 mm diameter suspended particles. Also shown is the least squares best fit straight line passing through the origin

8. Relative viscosity as a function of the dimensionless ball diameter (see Table 2). (□) 1.588 mm diameter rods ; (◆) 3.175 mm diameter rods. The error bars represent the 95% confidence limits on the data.

9. Frequency distribution of velocities of 19.05 mm balls for suspensions of 1.588 mm diameter rods (hatched bars) and 3.175 mm diameter rods (solid bars). While both suspensions yield the same value of average velocity, the variance about the mean is different with $\sigma = 1.17$ for the 1.588 mm rods and $\sigma = 6.09$ for the 3.175 mm rods.

10. Relative viscosity as a function of aspect ratio at a volume fraction of 0.05 for suspensions of randomly oriented rods. Shown are the values obtained in this study and our earlier experimental results [3] (■), Brenner's prediction [7] for a particle having $a_r = 10.01$, the average of that for the two populations of particles used in these experiments (◆), and the relative viscosity of a suspension of spheres having $\phi = 0.05$ (●) (to gauge approximately the behavior as $a_r \rightarrow 1$).

TABLES

1. Comparison of the viscosity of the suspending fluid using two different techniques to measure the terminal velocity of the falling balls.

2. Summary of Data for Suspensions of Rods.

3. Comparison of the intrinsic viscosities measured in the these experiments with the theoretical results of Brenner [7].

Ball Diameter (mm)	Viscosity (Pa-s) Eddy Current	Optical	Percent Difference[1]
9.525	11.5 ± 0.09[2]	11.36 ± 0.02	1.23
12.7	11.47 ± 0.11	11.32 ± 0.11	1.32
15.875	11.37 ± 0.09	11.17 ± 0.11	1.78
19.05	11.34 ± 0.01	11.17 ± 0.01	1.51
Average	**11.42 ± 0.03**	**11.26 ± 0.03**	**1.49**

Table 1. Comparison of the viscosity of the suspending fluid using two different techniques to measure the terminal velocity of the falling balls.

[1] [Viscosity $_{eddy\ current}$ - Viscosity $_{optical}$] x 100 / Average Viscosity.

[2] Ninety five percent confidence limit.

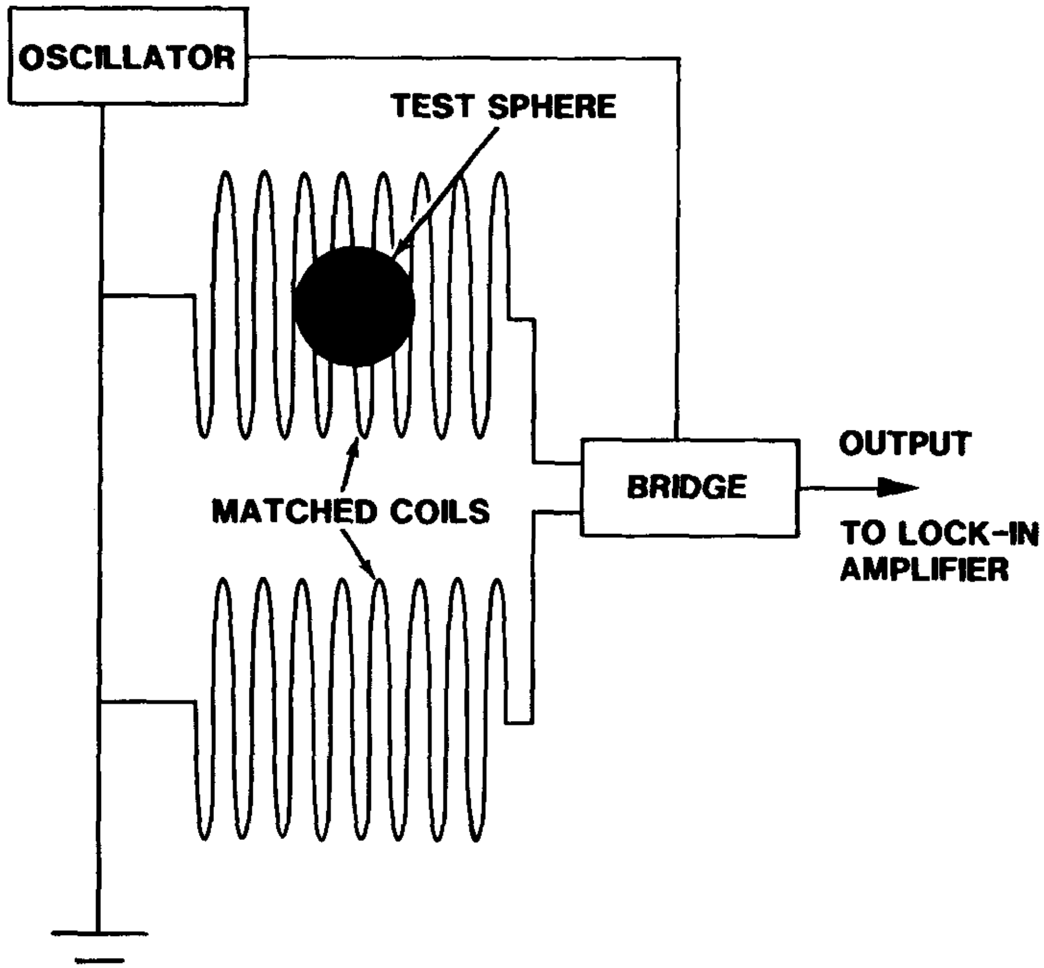
Suspended Rod Dia. (mm)	Ball Diameter (mm)	Number of Observations	Velocity (mm/s)	Corrected Velocity (mm/s)	Relative Viscosity
1.588	9.525	10	18.418±0.72[1]	21.45	1.463±0.058
1.588	12.7	10	31.506±1.22	38.8	1.435±0.058
1.588	15.875	10	47.642±1.22	62.2	1.419±0.038
1.588	19.05	10	63.817±2.	88.54	1.393±0.019
1.588	**Composite**	**40**			**1.433±0.021**
3.175	3.175	10	2.564±0.25	2.69	1.334±0.142
3.175	6.35	14	8.980±0.54	9.92	1.429±0.088
3.175	9.525	9	18.961±1.39	22.08	1.425±.102
3.175	12.7	19	30.760±1.16	37.88	1.480±0.052
3.175	15.875	18	46.643±2.29	60.89	1.460±0.11
3.175	19.05	28	62.34±2.36	86.49	1.483±0.058
3.175	**Composite[2]**	**74**			**1.470±0.032**

Table 2. Summary of Data for Suspensions of Rods.

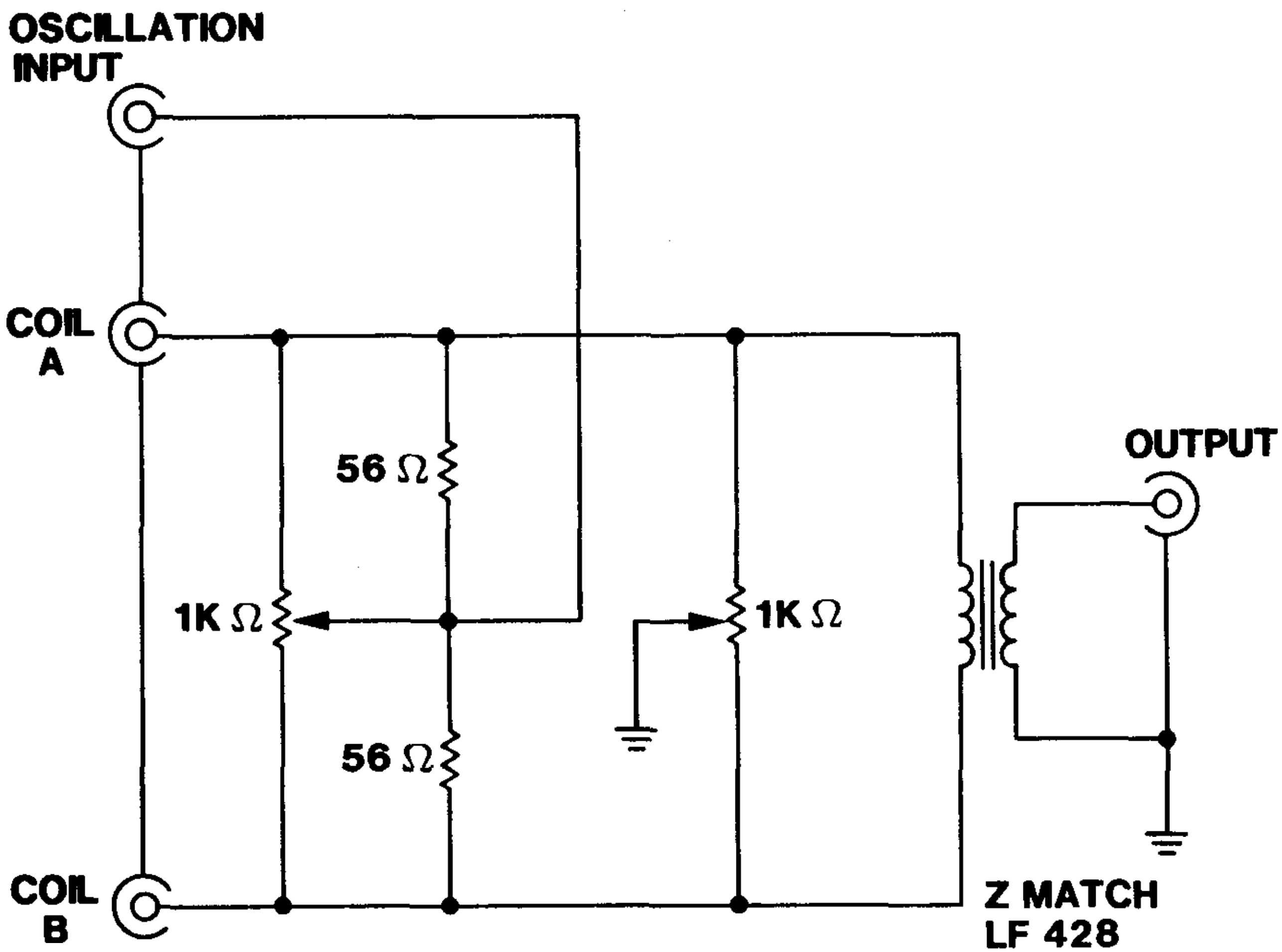
[1]Errors refer to 95% confidence limits
[2]Composite (average) includes data from four largest ball sizes only.

Rod Diameter (mm.)	**μ_r (Measured)**	**μ_r (Theory)**	**Percent Difference**	**$[\mu]$ (Measured)**	**$[\mu]$ (Theory)**	**Percent Difference**
1.588	1.433	1.495	2.1	8.66	9.85	12.9
3.175	1.470	1.50	1.0	9.4	10.	6.2

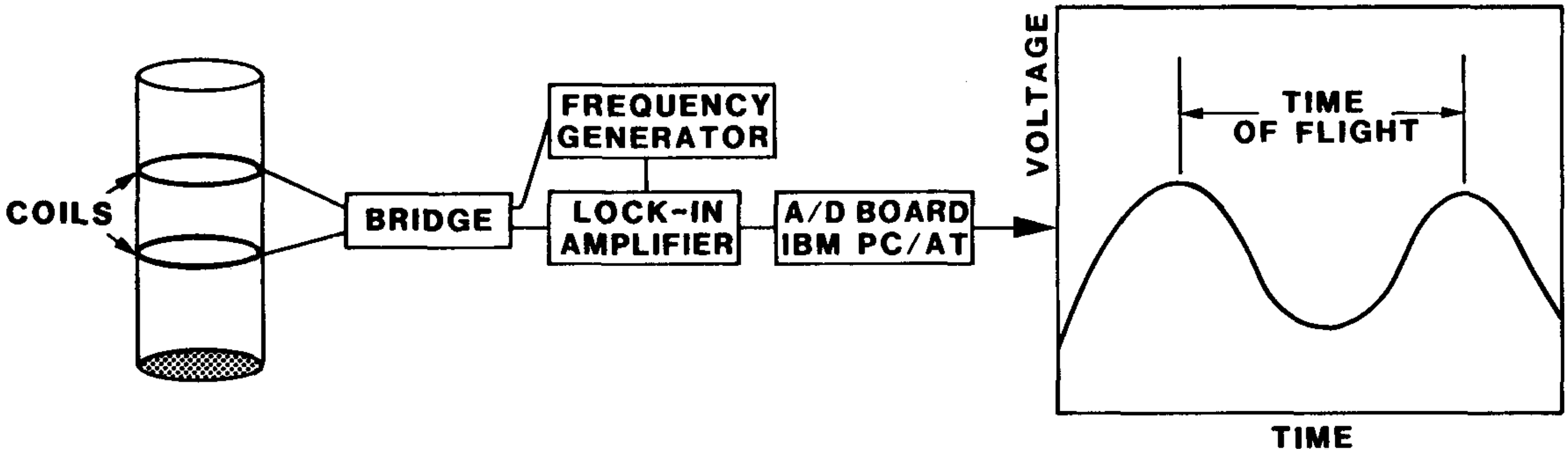
Table 3. Comparison of the intrinsic viscosities measured in the these experiments with the theoretical results of Brenner [7].



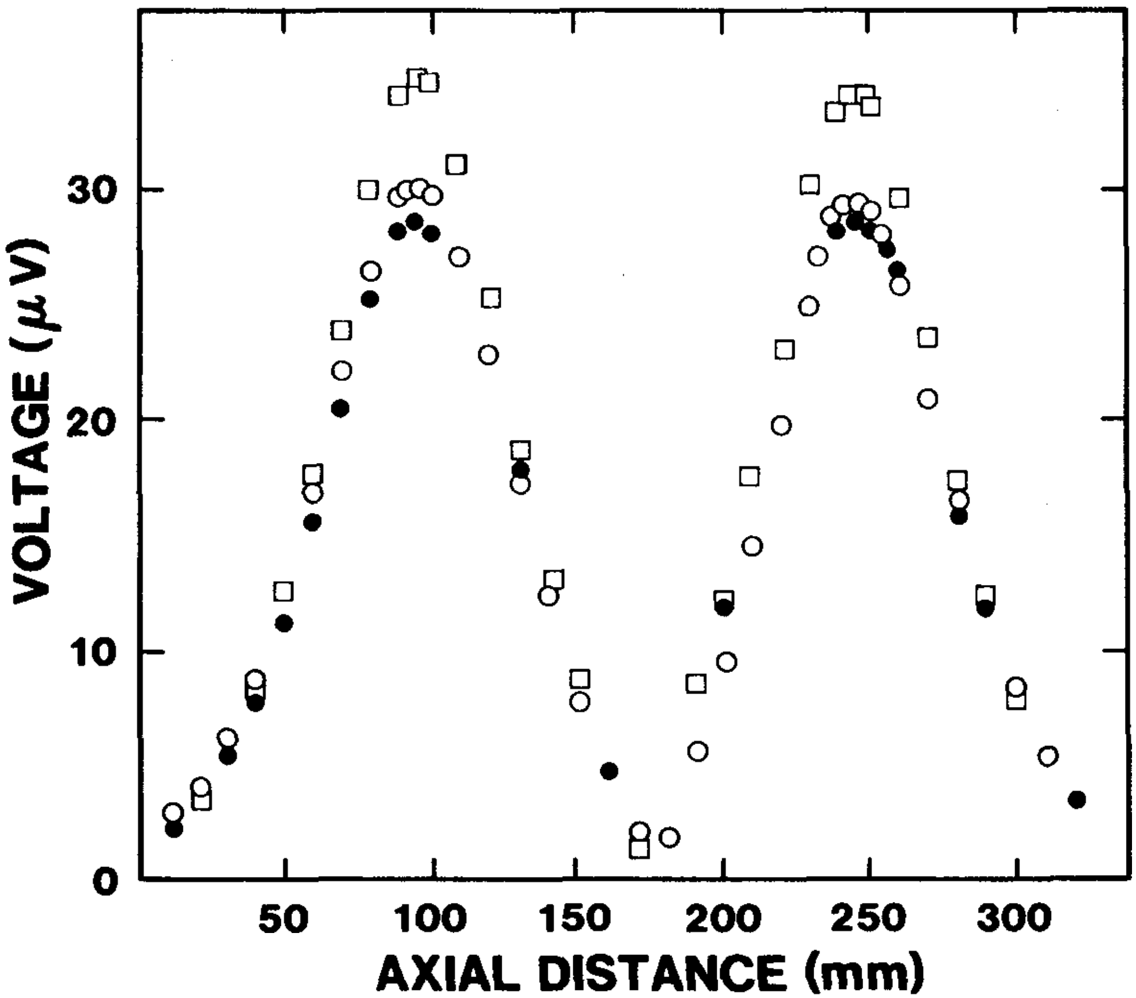
1. Schematic of the principle of the eddy current device. The oscillator excites the primary magnetic field in two identical coils. In the absence of the ball, the bridge connecting the coils is balanced. When the ball is placed near to one of the coils, an imbalance occurs due to the eddy currents induced in the sphere by the primary magnetic field.



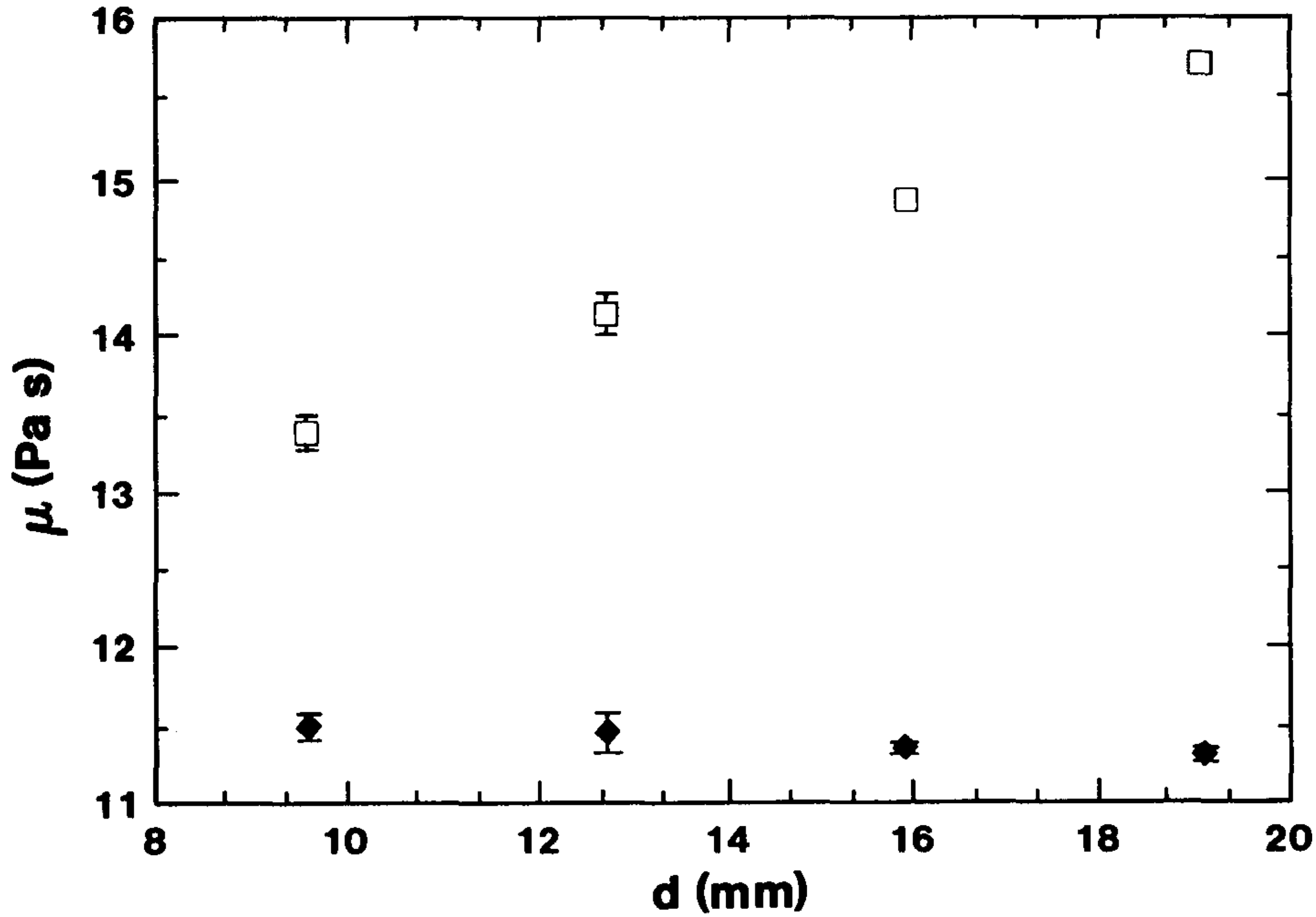
2. Bridge circuit used for eddy current measurements.



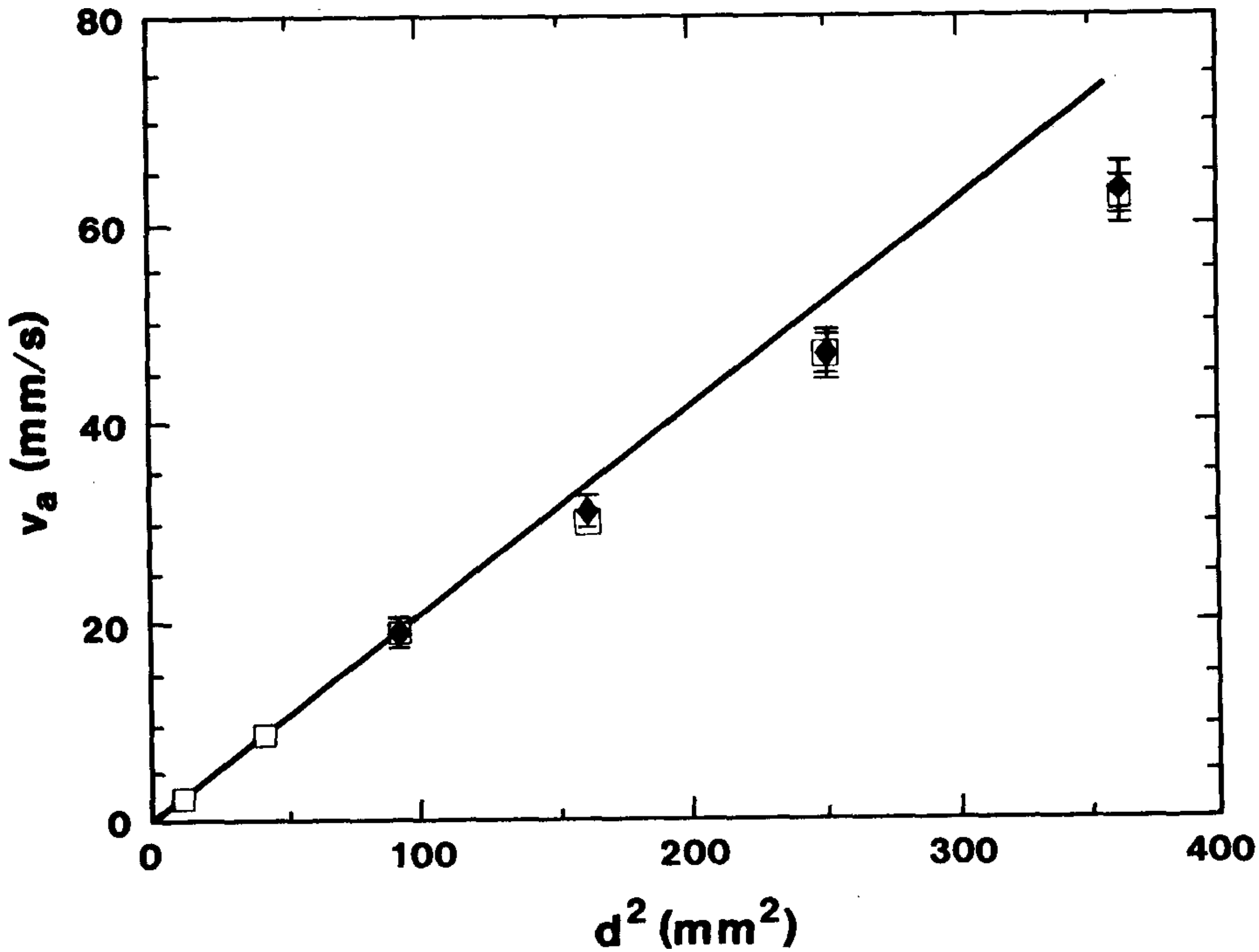
3. Schematic of system used for eddy current measurements of ball's terminal velocity in falling ball experiments.



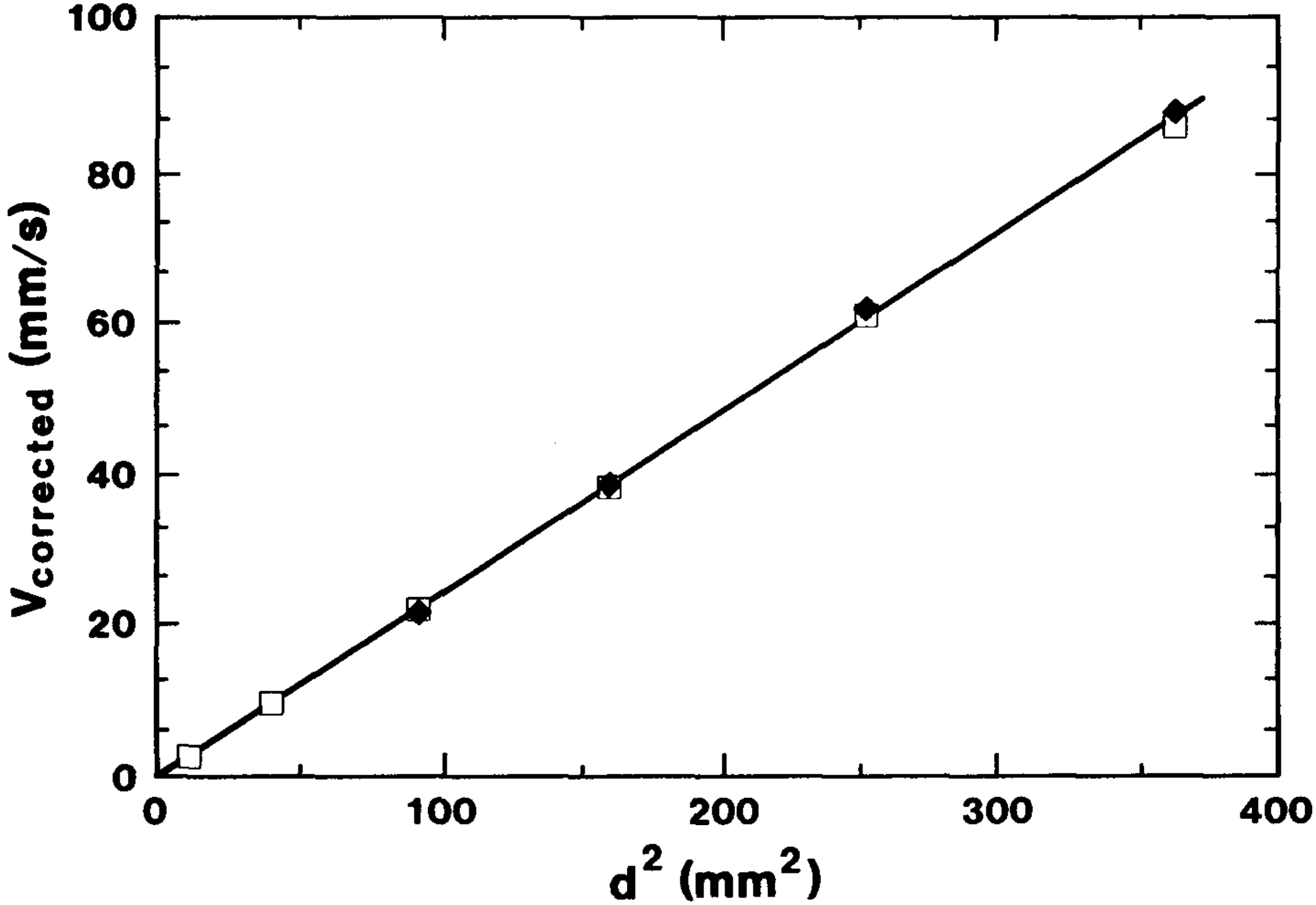
4. Calibration of eddy current device. Shown is the voltage versus the distance along the column for a 9.525 mm diameter brass ball as it is moved axially at three different radial positions: (○) along the center line; (●) one ball diameter off the center line; and (□) approximately 30 mm away from the wall of the column. Note that the zero distance point is arbitrary.



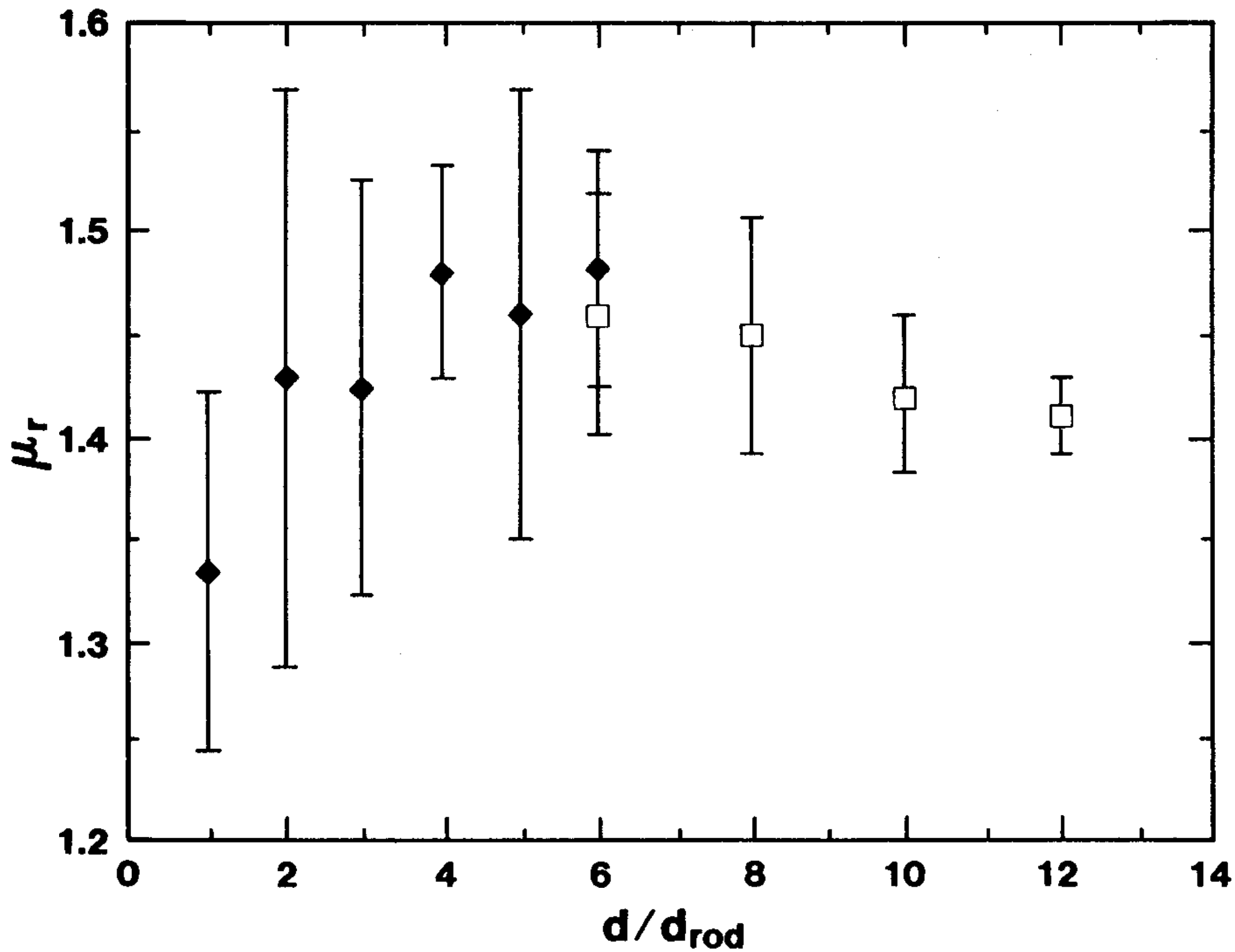
5. Viscosity of the suspending fluid as a function of the diameter of the falling balls. When Stokes's formula [14] for a ball falling through an unbounded medium is used to convert velocity to viscosity (□), the viscosity is found to increase with increasing ball diameter. When Eq. [2] is applied (◆), the viscosity is determined to be substantially unaffected by the presence of the container walls (see Table 1).



6. Ball velocity as a function of the square of the ball diameter for the suspensions of rods (see Table 2). (□) 3.175 mm diameter suspended particles; (◆) 1.588 mm diameter suspended particles. The straight line passes through the smallest diameter balls and the origin.



7. Ball velocity corrected for wall effects as a function of the square of the ball diameter for the suspensions of rods (see Table 2). (□) 3.175 mm diameter suspended particles; (◆) 1.588 mm diameter suspended particles. Also shown is the least squares best fit straight line passing through the origin



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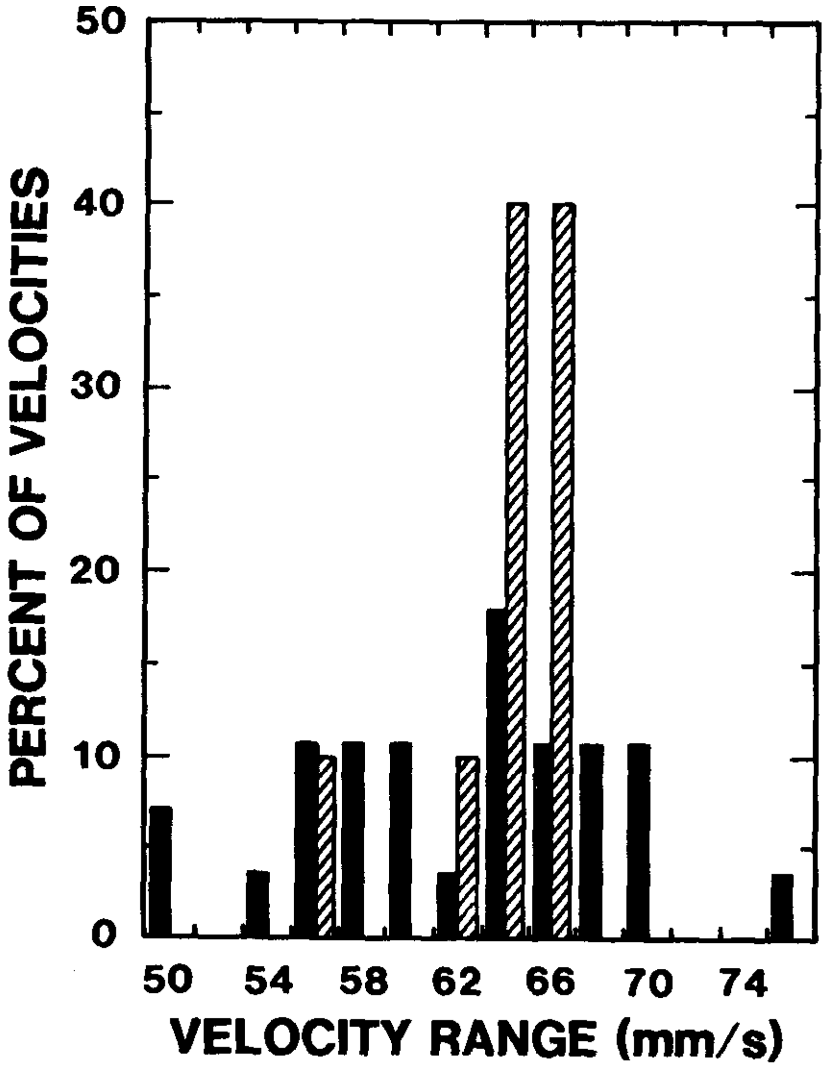

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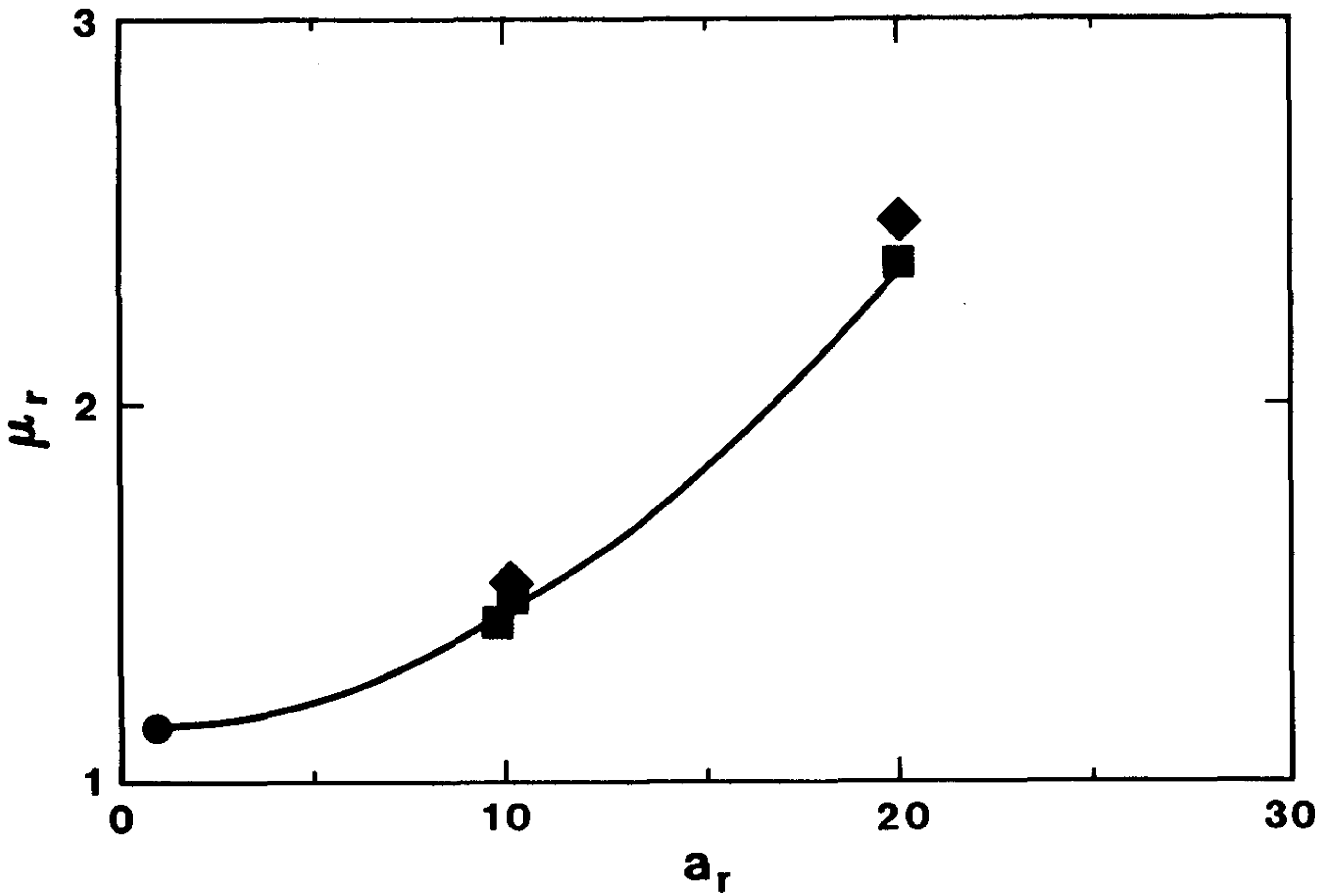

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