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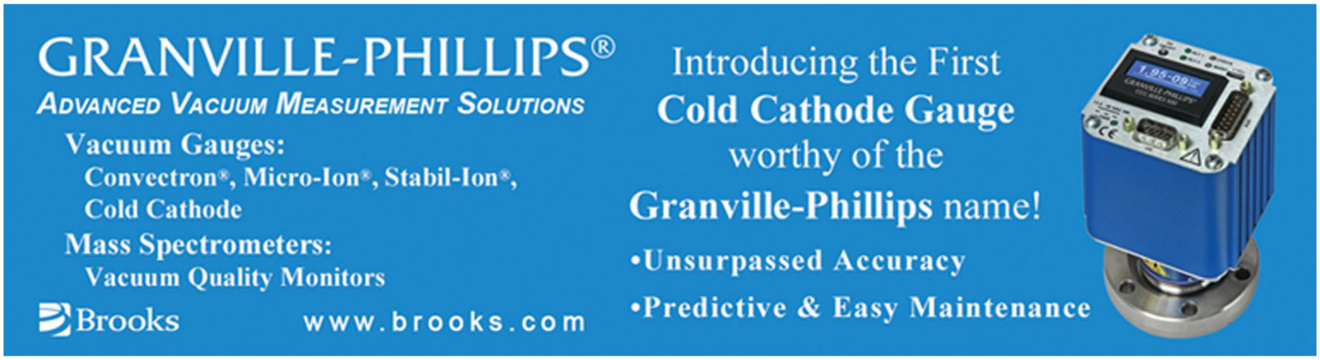

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Reliable method for determination of the velocity of a sinker in a high-pressure falling body type viscometer

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We present a new sensor configuration and data reduction process to improve the accuracy and reliability of determining the terminal velocity of a falling sinker in falling body type viscometers. This procedure is based on the use of multiple linear variable differential transformer sensors and precise mapping of the sensor signal and position along with the time of fall which is then converted to distance versus fall time along the complete fall path. The method and its use in determination of high-pressure viscosity of *n*-pentane and carbon dioxide are described. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1505100]

I. INTRODUCTION

The motivation for this work is linked to improving the reliability of a high-pressure viscometer that was developed in our laboratory.[1,2] Instrumentation for generating high-pressure viscosity is now of greater importance because of the need for transport property data for process design and development in supercritical fluid based technologies.[3] High-pressure viscosity data are also important in traditional processes such as lubrication, tertiary oil recovery and polymer processing among others.

We have in the past effectively used a special high-pressure viscometer for investigating the effect of pressure, temperature, composition and density on viscosity of simple fluids and polymer solutions.[4–8] Figure 1 is a schematic of the viscometer. The basic features of this viscometer are (a) a view cell with sapphire windows for observation, (b) a variable-volume part with a movable piston to bring about pressure or volume changes once the viscometer is loaded, (c) a fall-tube attachment that houses a cylindrical sinker with a hemispherical tip to assess the viscosity of the solution from the fall times of the sinker, and (d) a circulation loop to ensure the homogenization of the system contents.

The rationale for the suitability of this type of viscometer for use in high-pressure measurements and its reliability and limitations have been discussed in detail in our earlier publications.[1,2,4] In falling cylinder viscometers, the parameter measured is the time taken for a cylindrical sinker to fall a known distance in a fall tube filled with fluid. The relationship between the viscosity η of the fluid and the terminal velocity V_t of the sinker is given by

$$\eta=(1/V_t)[(\rho_s-\rho_f)K], \tag{1}$$

where ρ_s and ρ_f are the densities of the sinker and the fluid and K is an instrument constant which combines all the shape factors and is determined by measuring the terminal velocity of the sinker in fluids of known viscosity.[1] In the design of these viscometers, it is imperative that the fall of the cylindrical sinker is concentric in the fall tube. To achieve this, the ratio of the sinker-to-fall tube radii is maintained greater than 0.93. The most reproducible fall times and concentric falls in the fall tube are obtained with sinkers in which the leading edge is hemispherical. Reliability of the calibrations and ultimately the viscosity determinations depend on the accuracy with which fall times are measured and the accuracy with which the densities of the sinker and the fluid are determined at the experimental conditions. To determine the fall times (and the terminal velocities), common methods that are used are inductive, capacitive or optical methods. In our experimental system, we employ the inductive method using linear variable differential transformers (LVDTs).

The determination of the fall time of the sinker was initially based on the use of three independent LVDT coils positioned on a sleeve outside the fall tube. Figure 2 is a schematic of the signal from the three LVDT coils when the sinker passes through. The time corresponding to the zero voltage crossings in the linear response range of each LVDT coil is noted. From prior knowledge of the distance between the center points of each coil, which corresponds to the zero voltage reading positions, one can determine the velocity of the sinker between coils 1 and 2, or 2 and 3, or 1 and 3. From such data one can then assess if the sinker has reached its terminal velocity. If indeed the sinker reaches the terminal velocity prior to entering the coils, the velocity determined based on the distance traveled between any coil pair would not differ. This approach, even though it is simple and generates useful data, does not guarantee that terminal velocities are indeed reached in an unambiguous way in each measurement. Therefore, there is a need for an improved description of the fall time versus fall distance, rather than being limited to three-point measurements. To achieve this, we have adopted a simple approach in which we have measured the LVDT signals from each coil at 1 mm intervals as the sinker is manually moved along the fall tube. After a normalization procedure that is described in the following, a complete travel distance versus time plot can be generated in any

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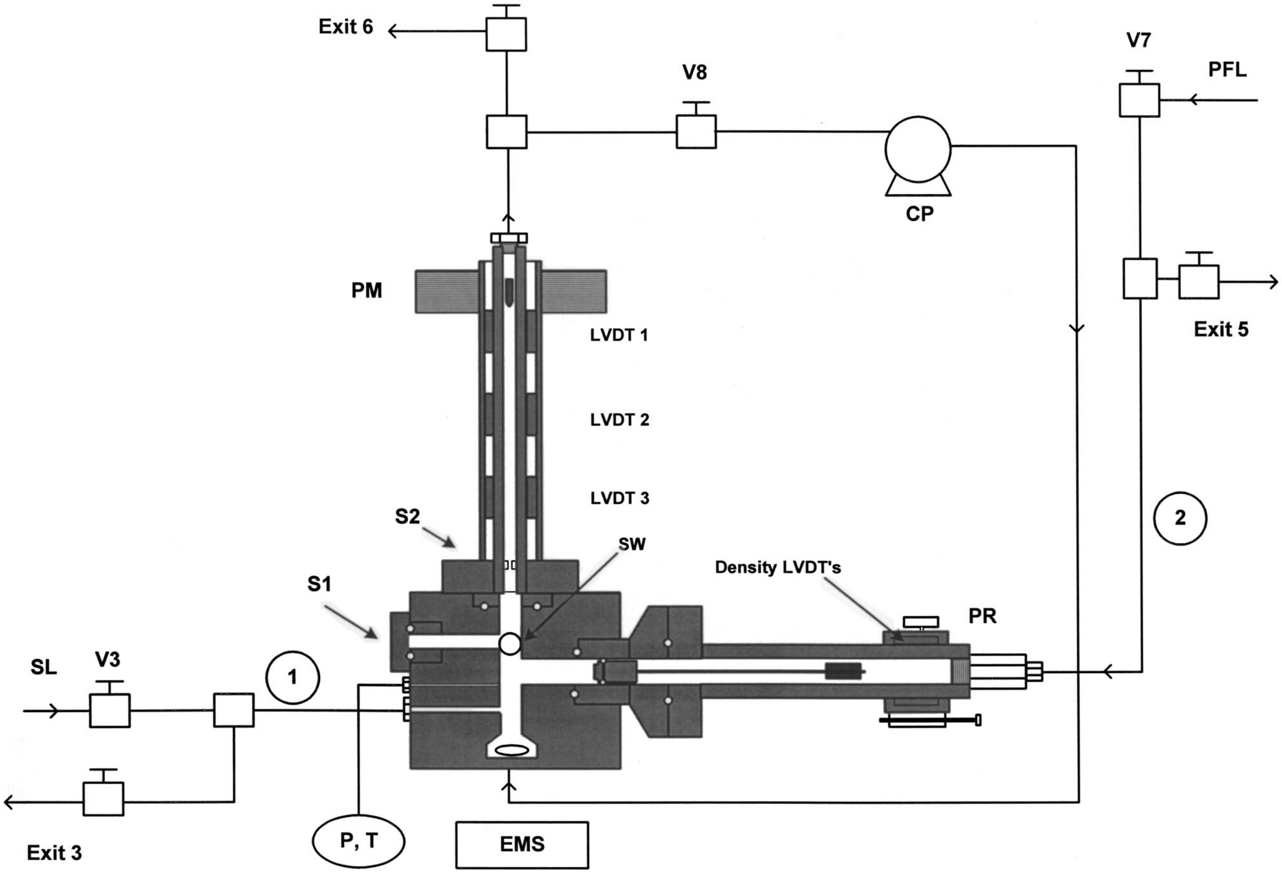

FIG. 1. Side view of the viscometer. Line 1=view cell inlet; line 2=piston inlet; SL=solvent line; PFL=pressurizing fluid line; CP=circulation pump; PM=pull-up magnet; S1=front loading part; S2=top loading part; SW=sapphire window; EMS=electromagnetic stirrer; PR=position readout unit; V=value; LVDT=linear variable differential transformer.

given experiment from which one can precisely determine if terminal velocities are indeed reached.

The new measurement approach gives an additional advantage. Again, like in the earlier measurement procedure, it is essential that the sinker pass through the full length of the fall tube for evaluation of the velocity. For viscous solutions this would require extremely long times. With the present procedure for complete documentation of sensor signal versus sinker position, assessment of terminal velocity can be made by monitoring the fall time starting from any position of the fall tube when working with highly viscous fluids.

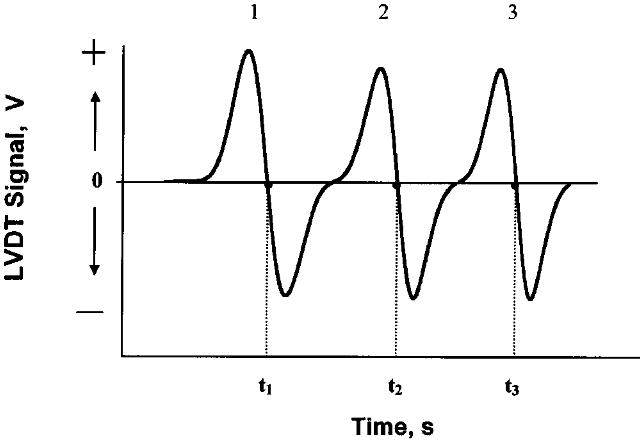

FIG. 2. Schematic of the signal from the three LVDT coils as the sinker passes through the fall tube.

II. DESCRIPTION OF THE LVDT SYSTEM FOR TERMINAL VELOCITY DETERMINATION

Figure 3 shows the new LVDT assembly. In this design, we now use one long LVDT washer, made of brass, that accommodates the three coils. This is in contrast to the three separate washers that were being used previously. The total length of the washer is 11.10 cm, whereas the coil lengths are about 3.53 cm. The outer diameter is 1.91 cm and each LVDT coil has one primary and two secondary coils as shown in Fig. 3. The outlets from the first secondaries are introduced as the inlets for the second secondaries at each LVDT coil. The outlets from the second secondaries of the first and the second LVDT coils are introduced as the inlets for the first secondaries of the second and third LVDT coils, respectively. Likewise, the outlets of the primaries of the first and the second LVDT coils are introduced as the inlets of the primaries of the second and third LVDT coils, respectively. Finally, the inlet of the first LVDT coil and the outlet from the third LVDT coil are connected for the primary and the secondary coils.

The primary of the first LVDT coil is excited by a sine wave signal generator and the output signal from the secondaries are sent to a LVDT signal conditioner unit. A balanced

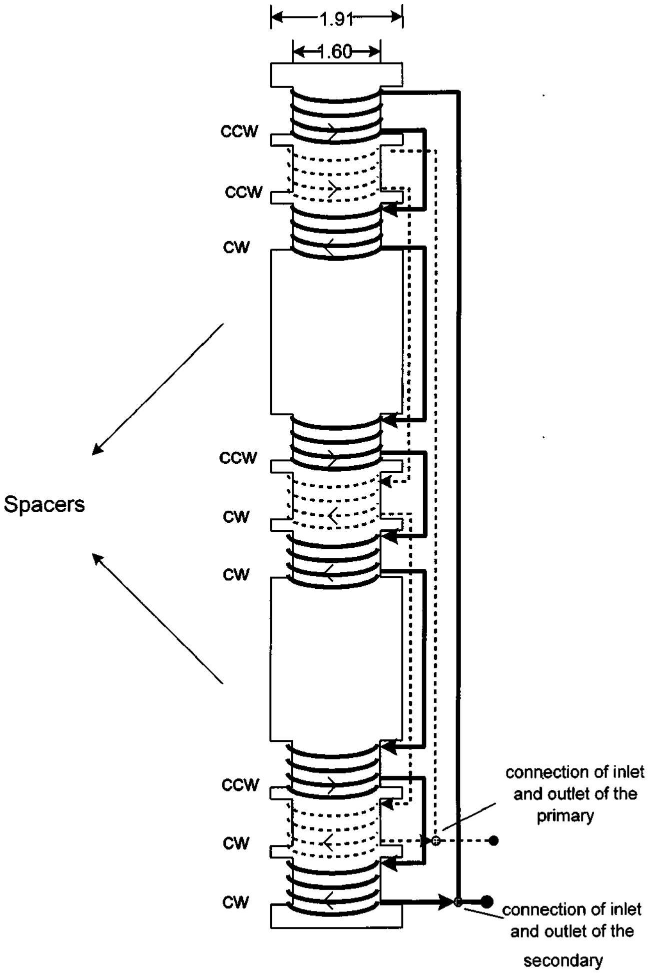

FIG. 3. Schematic of the windings of the viscosity LVDTs. Primary coils =dashed lines; secondary coils=solid lines; CW=clockwise winding; CCW =counterclockwise winding.

demodulator chip is used in the signal conditioner to produce a dc voltage, which corresponds to the position of the ferromagnetic sinker as it passes through the LVDT coils. The output of the LVDT signal conditioner is sent both to a dc ammeter to monitor the fall of the sinker visually on the control panel, and to a computer through an analog/digital (A/D) interface which stores the "voltage and real time" readings.

The first step in the new procedure is to relate the voltage readings from each LVDT to the positions of the sinker at any given time during the fall. For this determination, the ferromagnetic sinker is attached to the end of a long glass rod (longer than the length of the fall tube). The position when the glass rod is all the way in the tube is assigned as the initial point. Then the rod is pulled up in 1 mm increments, and the distance from the initial point as well as the corresponding LVDT voltage readings are recorded. A voltmeter connected to the ends of the LVDT coils is used to read the voltage. This procedure is repeated in the reverse direction: now the all-the-way-out position of the rod while a voltage reading could still be recorded is assigned as the initial position, and the rod is moved down into the tube. Again the distance and the voltage readings are determined in 1 mm increments. The distance versus voltage readings determined in this manner are shown in Fig. 4.

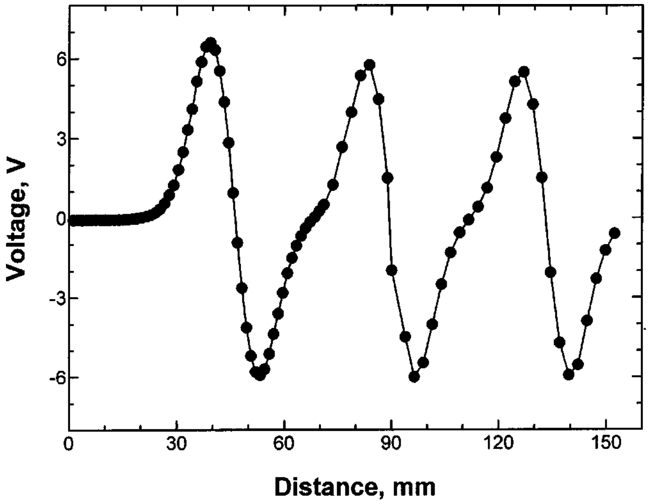

FIG. 4. LVDT signal in volts vs sinker position in the fall tube.

The second step is then to generate the LVDT signal in real time when the sinker falls through a given fluid. In the present experiments, we generated such data with *n*-pentane and also with carbon dioxide. Loading of the viscometer and operational procedures were the same as before.[1,2]

Once the system is loaded and thermal equilibrium is reached, several consecutive fall time measurements are taken at the desired temperatures and pressures. Sufficient time, typically 30 min, is allowed between consecutive repeat measurements to achieve stability in the system. This is needed because the pull-up magnet used to bring the sinker back to the top position after each fall time measurement (see Fig. 1) may lead to a change in temperature if sufficient time is not allowed. Figure 5 shows a typical computer output, which is obtained for pentane at 100 °C and 20.97 MPa.

Having the voltage versus distance (Fig. 4) and voltage versus time (Fig. 5) data, one can then generate the distance versus time plots along the fall tube. However, the different voltage readings (that arise from the different natures of the fluid used and the differences in temperature) in those ex-

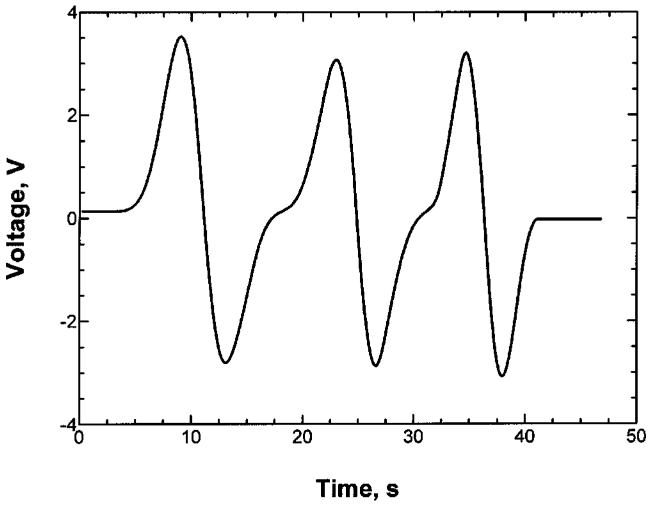

FIG. 5. LVDT response as a function of the fall time for the sinker in *n*-pentane at 100 °C and 20.97 MPa.

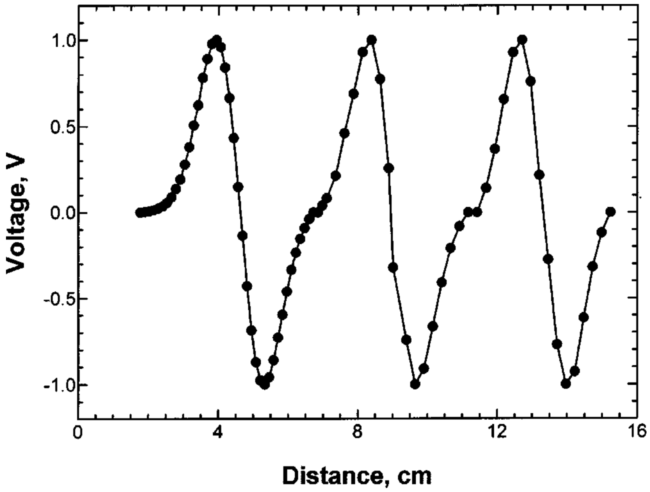

FIG. 6. Normalized plot of the LVDT response with the sinker position.

periments require that the data be normalized before these assignments can be made. Our normalization procedure is to assign +1 and −1 to the positive and negative apexes in the voltage readings for each LVDT coil and express all other readings accordingly in a given coil. This then converts data in Figs. 4 and 5 to the corresponding normalized figures given in Figs. 6 and 7 from which one can then generate the sinker position (travel distance) versus time plot shown in Fig. 8(a). The slope of the data in Fig. 8(a) would by definition give the velocity of the sinker in any given section of the fall tube. A perfectly linear variation would mean constant velocity throughout the fall tube. Figure 8(a) shows a high degree of linearity especially over the fall distance corresponding to the second and third coils. By fitting a linear regression line to the data, a terminal velocity for the sinker is assigned. This regression line is demonstrated in the Fig. 8(a) based on data corresponding to the third coil. Distance versus time data for the sinker passing through *n*-pentane at 50 °C and 20.70 MPa with the assigned terminal velocity is presented in Fig. 8(b) as another example. A similar procedure is carried out to determine the terminal velocities of carbon dioxide when it was used as the calibration fluid. The distance versus time data for the sinker passing through carbon dioxide at 37 °C and 20 MPa and at 97 °C and 40 MPa are given in Figs. 9(a) and 9(b), with the fitted regression lines to assign the terminal velocities.

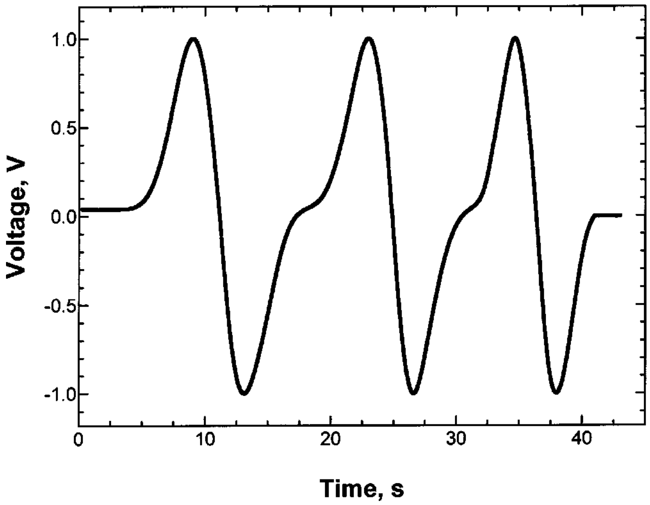

FIG. 7. Normalized LVDT response vs fall time in *n*-pentane at 100 °C and 20.97 MPa.

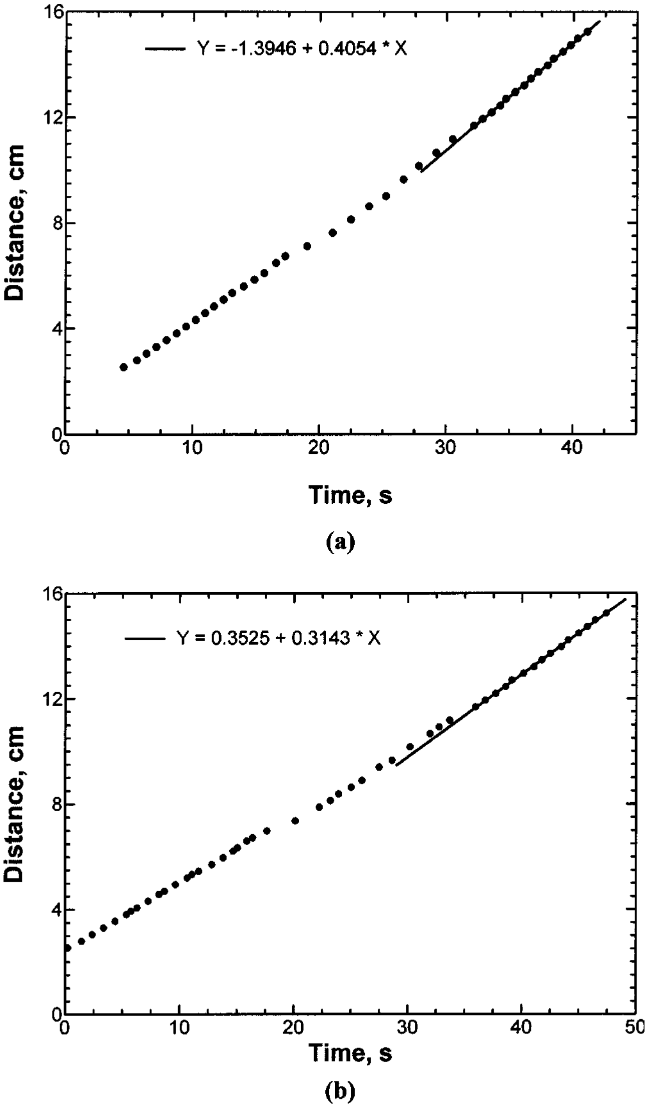

FIG. 8. Sinker fall distance vs time in *n*-pentane (a) at 100 °C and 20.97 MPa (b) at 50 °C and 20.70 MPa. The terminal fall velocity is given by the slope of the regression line.

III. CALIBRATION AND VISCOSITY MEASUREMENTS

The hydrodynamics of falling body viscometers and the governing equations have previously been described in the literature.[1,2] The basic equation is given by Eq. (1). Once the instrument constant is determined, then the viscosity of a given fluid can be calculated from measurements of the terminal velocity and the fluid density.

The procedure for determining the instrument constant involves measuring the terminal velocity in fluids of known viscosity and density. Then a K value can be calculated from Eq. (1). This is repeated for different fluids of different viscosity or for a given fluid at different temperature and pressure conditions for which viscosities and densities are

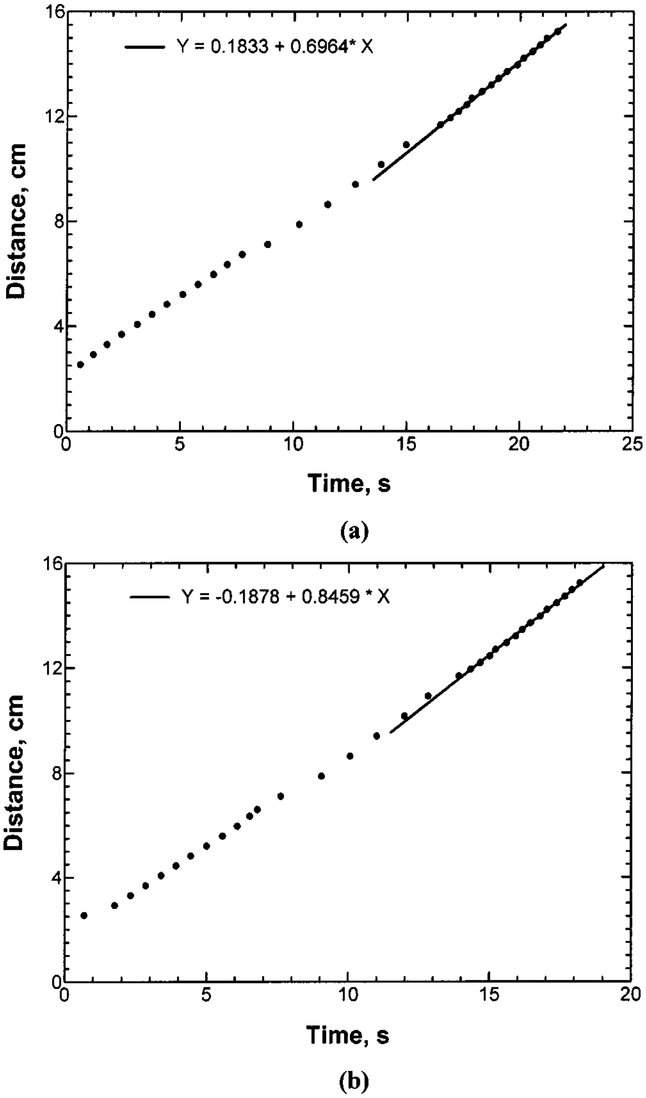

FIG. 9. Sinker fall distance vs time in carbon dioxide (a) at 37 °C and 20 MPa (b) at 97 °C and 40 MPa. The terminal fall velocity is given by the slope of the regression line.

known. By this procedure an average value of K that is applicable in a range of T/P conditions and viscosities can be assigned. The T/P dependence of K is considered to correct for any dimensional changes in the system, and the viscosity dependence of K is considered to extend the range of use of Eq. (1) to the non-Newtonian regime. In this study, we have conducted such calibrations using pentane and also carbon dioxide as calibration fluids.

A. Evaluation of the instrument constant using *n*-pentane as the calibration fluid

The densities of *n*-pentane and the fall times for the sinker were determined at 50, 75, 100 and 125 °C at pressures up to 50 MPa. The density values were determined from the initial mass of pentane loaded to the viscometer, the total internal volume of the viscometer (42.6 cm^3) when the movable piston is in its all-the-way-out position (in the variable volume part of the viscometer), and the exact piston position and the corresponding internal volume at any given temperature and pressure.

TABLE I. Density of *n*-pentane.

T (°C)	P (MPa)	Value in Ref. 4 (ρ/g/cm^3)	Value in present study (ρ/g/cm^3)	Difference (%)
50	44.76	0.6417	0.6358	0.91
	34.41	0.6328	0.6262	1.05
	27.75	0.6257	0.6200	0.91
	20.70	0.6178	0.6125	0.87
	15.39	0.6113	0.6075	0.61
	10.23	0.6041	0.6009	0.53
75	45.75	0.6232	0.6210	0.35
	44.92	0.6229	0.6313	1.35
	36.45	0.6135	0.6122	0.21
	28.39	0.6041	0.6042	0.01
	27.62	0.6041	0.6027	0.23
	20.72	0.5946	0.5934	0.21
	15.45	0.5870	0.5864	0.11
100	45.69	0.6055	0.6042	0.22
	38.21	0.5973	0.5956	0.17
	20.85	0.5730	0.5730	0
	15.90	0.5645	0.5647	0.03
125	45.08	0.5886	0.5883	0.06
	42.03	0.5826	0.5847	0.28
	34.61	0.5731	0.5736	0.10
	29.97	0.5626	0.5666	0.70
	24.47	0.5595	0.5607	0.22
	17.08	0.5447	0.5440	0.13
	10.85	0.5274	0.5308	0.64

Table I provides the density values determined in the present study along with the densities that were determined using this instrumentation in an earlier study. The reproducibility is extremely high with the differences being less than 1%. Based on the earlier comparisons in connection with such density measurements,[4] the reliability of these measurements is within 1% of the literature values for *n*-pentane density.

For the viscosity reference values for *n*-pentane to be used in calibrations we also used the values reported from an earlier study conducted in our laboratory. These values have previously been extensively evaluated against all available *n*-pentane viscosity data and were found to be within 3% of the value in the literature.[4]

At the pressure and temperature conditions for which we had had reference viscosity data, we have determined the density of the fluid and the sinker terminal velocities. These are shown in Table II. The K values that are determined from these measurements are also tabulated at each pressure and temperature. Table II includes results of several repeat measurements at given temperature and pressure conditions to illustrate the repeatability of terminal velocity determinations. At each temperature an average value of K applicable in the given pressure range has been assigned. Table II shows that K values do not show much variation with pressure, but show slight sensitivity to temperature. Based on these data an overall average value of 0.0199 has been assigned for the instrument constant. This is particularly applicable in the temperature range from 50 to 125 °C and pressure range from 10 to 46 MPa.

TABLE II. Instrument constant using *n*-pentane as the calibration fluid. η=viscosity value from Ref. 4; ρ=solvent density, measured; V_t=terminal velocity, determined; K=instrument calibration constant.

T (°C)	P (MPa)	η (mPa s)	ρ (g/cm^3)	V_t (cm/s)	K
50	10.23	0.1988	0.6009	0.3393	0.0198
	15.39	0.2072	0.6075	0.3318	0.0203
	20.70	0.2197	0.6125	0.3143	0.0204
	27.75	0.2348	0.6200	0.2925	0.0203
	34.41	0.2483	0.6262	0.2752	0.0203
	44.76	0.2717	0.6358	0.2513	0.0203
				Average	0.0202
75	10.62	0.1664	0.5790	0.4129	0.0201
	15.52	0.1783	0.5854	0.3928	0.0205
	20.72	0.1971	0.5929	0.3700	0.0214
	15.56	0.1783	0.5864	0.3923	0.0205
	15.53	0.1783	0.5864	0.3920	0.0205
	20.75	0.1971	0.5935	0.3736	0.0216
	20.70	0.1971	0.5936	0.3737	0.0216
	28.39	0.2012	0.6042	0.3352	0.0199
	28.39	0.2012	0.6041	0.3426	0.0203
	28.34	0.2012	0.6042	0.3346	0.0198
	36.49	0.2147	0.6152	0.3147	0.0200
	36.39	0.2147	0.6152	0.3150	0.0200
	36.45	0.2147	0.6152	0.3147	0.0200
	45.11	0.2333	0.6310	0.2971	0.0206
	45.18	0.2333	0.6314	0.2959	0.0205
				Average	0.0204
100	15.81	0.1479	0.5645	0.4382	0.0189
	15.81	0.1479	0.5645	0.4337	0.0187
	15.90	0.1479	0.5645	0.4329	0.0186
	15.90	0.1479	0.5645	0.4378	0.0188
	15.90	0.1479	0.5647	0.4326	0.0186
	15.90	0.1479	0.5647	0.4369	0.0188
	15.85	0.1479	0.5647	0.4342	0.0187
	15.85	0.1479	0.5647	0.4391	0.0189
	20.86	0.1596	0.5724	0.4144	0.0193
	20.86	0.1596	0.5724	0.4118	0.0192
	20.86	0.1596	0.5724	0.4078	0.0190
	20.86	0.1596	0.5724	0.4143	0.0193
	20.97	0.1596	0.5730	0.4054	0.0189
	20.97	0.1596	0.5730	0.4089	0.0190
	20.86	0.1596	0.5730	0.4128	0.0192
	20.86	0.1596	0.5730	0.4159	0.0194
	28.31	0.1759	0.5835	0.3856	0.0199
	28.28	0.1759	0.5835	0.3858	0.0199
	38.14	0.1999	0.5973	0.3512	0.0206
	38.14	0.1999	0.5973	0.3560	0.0209
	38.11	0.1999	0.5973	0.3531	0.0207
	38.11	0.1999	0.5973	0.3570	0.0210
	45.50	0.2032	0.6041	0.3377	0.0202
				Average	0.0197
125	17.11	0.1269	0.5440	0.4981	0.0183
	30.02	0.1470	0.5666	0.4256	0.0182
	30.02	0.1470	0.5666	0.4373	0.0187
	34.62	0.1595	0.5737	0.4120	0.0192
	42.03	0.1795	0.5843	0.3885	0.0204
				Average	0.0190

Overall average K (T=50–125 °C; P=10–46 MPa) 0.0199

B. Evaluation of the instrument constant using carbon dioxide as the calibration fluid

TABLE III. Instrument constant using carbon dioxide as the calibration fluid.

T (°C)	P (MPa)	η (mPa s)	ρ (g/cm^3)	V_t (cm/s)	K
37	20.0	0.0796	0.8696	0.6005	0.0187
	30.1	0.0956	0.9264	0.6964	0.0177
				Average	0.0182
52	20.0	0.0671	0.8821	0.8306	0.0179
	30.0	0.0835	0.8855	0.6990	0.0187
	40.0	0.0963	0.9397	0.6167	0.0194
				Average	0.0187
77	30.7	0.0667	0.8809	0.8602	0.0184
	40.0	0.0787	0.8584	0.7344	0.0184
				Average	0.0184
97	40.0	0.0678	0.8875	0.8459	0.0184
				Average	0.0184

Overall average K (T=37–97 °C; P=20–40 MPa) 0.0184

Table III shows literature data for the viscosity of carbon dioxide, along with densities and the terminal sinker velocities that were measured in the present system in the temperature range from 37 to 97 °C and pressures in the range from 20 to 40 MPa. An overall average value of 0.0184 is obtained for the instrument constant from these measurements.

C. Influence of *K* on viscosity evaluations

The K value determined from CO_2 as the calibration fluid is different than that obtained from *n*-pentane as the calibration fluid. This suggests that for accurate viscosity determinations one should use the calibration constant applicable in the viscosity range of interest. Nonetheless, we have evaluated the influence of using K_{pentane} in determining the viscosity of CO_2, and K_{CO_2} in determining the viscosity of *n*-pentane.

If the terminal velocities and the density values in Table III are used along with $K=0.0199$, the calculated viscosities for CO_2 show an average deviation of about 7.3% from the literature values given in Table III.

Similarly, if the terminal velocities and the density values given in Table II are used with $K=0.0184$, then the

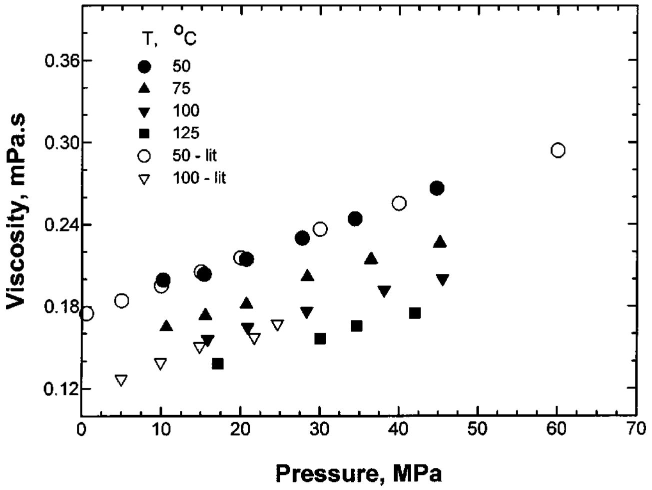

FIG. 10. Variation of the viscosity of *n*-pentane, calculated using K_{pentane} (closed symbols), with pressure and comparison with literature values (open symbols). The literature data at 50 °C are from Ref. 9 and the data at 100 °C are from Ref. 10.

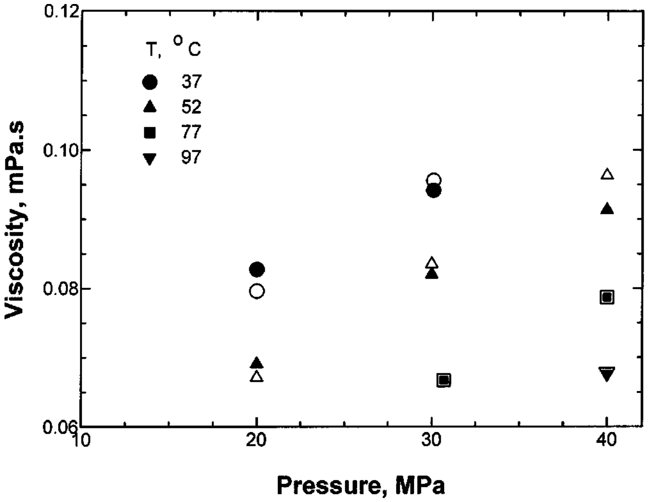

FIG. 11. Variation of the viscosity of carbon dioxide, calculated using K_{CO_2}, (closed symbols), with pressure. The literature viscosity data (open symbols) at the same temperatures are from Ref. 11.

calculated viscosities for *n*-pentane show an average deviation of about 7.6% from the literature values given in Table II.

Using the average K value based on *n*-pentane, recalculation of pentane viscosities at each T/P condition given in Table II leads to an average error of about 3.4%. Figure 10 shows the variation of viscosity values, using the K_{pentane} calibration constant, with pressure at selected temperatures. Figure 10 also includes independent measurements from the literature at nominal temperatures of 50 and 100 °C.[9,10] As is shown, there is excellent agreement at 50 °C. The minor difference at 100 °C possibly arises from the fact that the literature data for the viscosity at this temperature are based on density values estimated from correlation instead of direct measurements like in the present study.

Using the average K value for CO_2, recalculation of CO_2 viscosities at each T/P condition given in Table III leads to an average error of about 1.95%. Figure 11 shows the variation of viscosity values, using the K_{CO_2} calibration constant, with pressure at selected temperatures. Figure 11 also includes measurements from the literature at the same temperatures.[11] The agreement with the literature is very good.

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