Vibrating Wire Viscometer with Wire Diameters of (0.05 and 0.15) mm: Results for Methylbenzene and Two Fluids with Nominal Viscosities at $T = 298$ K and $p = 0.01$ MPa of (14 and 232) mPa·s at Temperatures between (298 and 373) K and Pressures below 40 MPa

Mohamed E. Kandil and Kenneth N. Marsh*

Department of Chemical and Process Engineering, University of Canterbury, Christchurch, New Zealand

Anthony R. H. Goodwin

Schlumberger Technology Center, 125 Industrial Boulevard, Sugar Land, Texas 77478

A vibrating wire viscometer has been developed with an electrically insulating tensioning mechanism. It has been used with two wires, one of diameter 0.05 mm the other of diameter 0.15 mm, to measure the viscosity of methylbenzene and two reference fluids with viscosities of (14 and 240) mPa·s at $T = 298$ K and $p = 0.1$ MPa at temperatures in the range from (298 to 373) K and pressures below 40 MPa, where the viscosity covers the range from (0.3 to 100) mPa·s with an uncertainty of < 0.6 %. The results so obtained differ from literature values by < 1 %. Measurements with the 0.150 mm diameter wire at $T =$ 301 K demonstrate that the upper operating viscosity is 200 mPa·s with an uncertainty of about 3 %.

Introduction

Knowledge of viscosity is important to the chemical and petroleum industry. For the latter, the viscosity of the fluid in the reservoir determines the economic viability of the reservoir. There are numerous methods by which viscosity can be measured, and these have been reviewed in ref 1. Of those viscometers, vibrating wires are of simple construction and can be operated remotely over a wide range of temperature and pressure. Bessel[2] provided the physical description of a wire oscillating in a dense fluid in 1828. Stokes[3] in 1850 included the effect of viscosity on the vibrating wire. The development of the vibrating wire as a viscometer is summarized in Table 1. To our knowledge, all but three of these instruments have been used to measure viscosity in the range from (0.008 to 6) mPa·s with the three exceptions that are as follows: Charles et al.[38] reported measurements of the viscosity of glycol at about 53 mPa·s; Gourgouillon et al.[24] measured supercritical fluid-saturated polymer (PEG 400) with a viscosity of about 20 mPa·s; and Caetano et al.[17] reported measurements of diisodecylphthalate with a viscosity up to 120 mPa·s. In this work, a vibrating wire viscometer has been developed, and viscosities have been measured for standard reference fluids up to about 200 mPa·s, which is the upper operating viscosity. This vibrating wire viscometer has the following novel features: (1) an internal volume of 10 cm^3; (2) magnets located within the fluid; and (3) a mechanism for tensioning the wire that also electrically insulates each end. Item 2 reduces the separation between the magnets and, therefore, the dimensions of the magnet required to provide a particular field at the wire that ultimately reduces the diameter of the complete instrument. The viscometer has been used to measure the viscosity of methylbenzene and two reference fluids with nominal viscosities of (14 and 240) mPa·s at T= 298 K and p = 0.1 MPa at temperatures in the range from (298 to 373) K at pressure below 40 MPa. The measured viscosities range from (0.3 to 100) mPa·s. The results obtained differ from literature values by less than 1 %, and measurements show that, as anticipated from the working equations, increasing the wire diameter to 0.15 mm allows the measurements to be performed with fluids of viscosities up to 200 mPa·s with results within 2 % of the reference value. This difference is within the uncertainty of the measurement, which, at that viscosity, was estimated at 3 %.

Working Equations

A vibrating wire can be used to measure viscosity and density simultaneously: the resonance frequency of the wire is related to both the fluid density and wire tension, whereas the resonance line width is related to the ratio of viscosity to density. Several methods have been developed to optimize the response of a device to solely viscosity or both density and viscosity (with a compromise in the precision of both). To obtain both density and viscosity, the wire needs to be clamped at the top and a mass suspended at the lower end. This configuration significantly increases the sensitivity of the resonance frequency to the fluid density because the wire tension is related to the buoyancy of the suspended mass. To obtain measurements of density and viscosity, both the frequency and the half-width of the resonance must be stable and reproducible and the orientation of the suspended mass with respect to the gravitational field known. If only the viscosity is required, then the wire can be clamped at both ends. In this case, provided the vacuum resonance frequency is stable to within a few tens of Hertz, measurements of the resonance bandwidth suffice to determine the viscosity for a fluid when the density is known.

The theory that describes the motion of a wire within a fluid with either of the clamping arrangements above has been described previously.[4,5,29,46,48,50,51] Our instrument comprises a thin metallic wire clamped under tension

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* To whom correspondence should be addressed. E-mail: ken.marsh@canterbury.ac.nz. Tel: +64 3364 2140. Fax: +64 3364 2063.

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Table 1. Review of the Literature Reporting the Development of a Vibrating Wire to Determine Fluid Viscosity η along with Maximum Pressures p, Temperature Range T, Estimated Uncertainty u, Wire Material S, Wire Diameter D, Wire Length l, Clamped at One End with a Buoyant Mass BM, Clamped at Both Ends 2C, Clamped with a Constant Tension System CTS, and Operated in either Forced FR or Transient TR Mode.

T_{min} to T_{max} K	p MPa	fluid	η_{min} to η_{max} mPa·s	±100·u	S	D µm	l mm	clamp	mode	comments	ref
										theory	4
										theory	5
240 to 455	165	natural gas mixtures	0.009 to 0.016	1	W	7			TR		6
210 to 370	0.1	methylbenzene	0.3 to 2.7	0.5	W			BM	TR		7
210 to 370	30	methylbenzene	0.3 to 2.7	0.5	W	100	56	BM	TR		8
303 to 348	250	heptane	0.2 to 1.8	3	W	100	56	BM	TR		9
303 to 348	300	heptane	0.2 to 2	3	W	100	56	BM	TR		10
298	70	hexane	0.298 to 0.529	0.5	W	100	54	CTS	TR		11
303 and 323	80	benzene, methylbenzene, *m*-xylene	0.421 to 0.897	0.5	W	100	54	CTS	TR		12
213 to 373	250	methylbenzene		2.5						reference correlation	13
		1,1,1,2-tetrafluoroethane (R134a)								IUPAC round-robin	14 15
303 and 323	70	methylbenzene, heptane	0.421 to 0.897	0.5	W	100	54	CTS	TR		16
288 to 308	0.1	diisodecylphthalate	120	1	W	200	60	2C	TR		17
213 to 298	20	methylbenzene	0.554 to 3.3	1.5	W	100	40	CTS	TR		18
298 to 473	200	dodecane and octadecane	0.22 to 6.7	2	W	100	32	BM	FR		19
298 to 393	75	mixtures of methane and decane	0.086 to 1.648	3	W	100	28	BM	FR		20
303 to 423	50	mixture of hexane and 1-hexanol	0.2 to 6.7	4	W	100	28	BM	FR		21
298 to 383	100	pentane	0.1 to 0.43	2.5	W	100	28	BM	FR		22
222 to 348	80	methylbenzene	0.3 to 2.2	3	W	100	40	BM	FR		23
313 to 348	25	supercritical CO_2-saturated PEG400	20	4	W	100	40	BM	FR		24
198 to 348	100	2,2,4-trimethylpentane (isooctane)	0.289 to 6.16	3	W	100	40	BM	FR		25
199 to 298	100	1,1,1,2-tetrafluoroethane (R134a)	0.205 to 0.824	2.5	W	100	40	BM	FR		26
197 to 348	40	cyclohexane, 1,1,1,2-tetrafluoroethane, 2,2,4-trimethylpentane	0.2 to 3	2	W	100	40	BM	FR		27
260 to 300	100	CO_2(l)	0.06 to 0.24	0.5	W	100	56	BM	FR		28
										theory	29
298 and 323	10	methylbenzene	0.422 to 0.601	0.4 (FR) 0.8 (TR)	W	100	40	BM	FR/TR		30
231 to 343	5.1	R32, R125	0.06 to 0.25	1	W	7.5	140	BM	TR		31
223 to 343	4.8	R134a, R32, R125	0.010 to 0.015	2	W	7.5	140	BM	TR		32
303 to 348	250	methylbenzene, pentane, hexane, octane, and decane	0.18 to 5.9	0.5	W	100	56	BM	TR		33
235 to 343	50	R134a	0.11 to 0.24	0.6	W	100	56	BM	TR		34
298 to 423	20	C_3H_4 (g)	0.008 to 0.073	3		25	90	BM	TR	chromel wire	35
298 to 423	20	Ar, Kr, C_3H_8	0.022 to 0.042	3		25	90	BM	TR	chromel wire	36
298 to 500	40			0.2						theory	37
80 to 323		H_2O, glycol, CS_2, O_2(l), Ar(g)	0.024 to 53	0.13	W	100	50	2C	TR		38
223 to 323	779	Ar	0.11 to 0.33		W	50	20	2C	TR		39
273	1000	CH_4	0.010 to 0.415		W	50	20	2C	TR		40
220 to 280	450	CO_2(l)	0.5	0.5	W	50	20	2C	TR		41
174.45	470	Ar	0.61	2.5	W	50	15	2C	TR		42
243 to 393	3.5	1,1-difluoroethane (R152a)	0.012 to 0.339	2.8	W	50	15	2C	TR		43
				2						theory	44
174	471	Ar								comments	45
										theory	46
2.18 to 1.1		^3He (l)	0.026	5	W	25	50	2C	TR		47
										theory	48
1.8 to 3.2	2.5	^4He	0.023 to 0.069			80	50	2C	TR	316 wire	49
										theory	50
										theory	51
0.0013		^3He				300				NbZr wire	52

between two fixed supports and immersed in a fluid. The wire is placed in a magnetic field, and it is driven in steady-state transverse oscillations by passing an alternating current. The complex voltage $\boldsymbol{V}$ across the wire is given by

$$\boldsymbol{V} = \boldsymbol{V}_1 + \boldsymbol{V}_2 \tag{1}$$

where $\boldsymbol{V}_1$ is the voltage arising from the electrical impedance of the effectively stationary wire and $\boldsymbol{V}_2$ represents the motional emf. $\boldsymbol{V}_1$ is represented empirically by

$$\boldsymbol{V}_1 = a + \mathbf{i}b + \mathbf{i}cf \tag{2}$$

where f is the frequency at which the wire is driven and a, b, and c account for the electrical impedance of the wire and absorb the offset used in the lock-in amplifier to ensure that the voltage signal is detected in the most sensitive range. In eq 2, a, b, and c are adjustable parameters determined by regression with the measured complex voltage. $\boldsymbol{V}_2$, given by[4,5]

$$\boldsymbol{V}_2 = \frac{\Lambda f \mathbf{i}}{f_0^2 - (1+\beta)f^2 + (\beta' + 2\Delta_0)f^2\mathbf{i}} \tag{3}$$

relates the measured quantity to the fluid properties. In eq 3, Λ is an amplitude, f_0 is the resonance frequency of the wire in vacuum, Δ_0 is the internal damping of the wire, β is the added mass arising from the fluid displaced by the wire, and β' is the damping due to the fluid viscosity.

Retsina et al.[4,5] analyzed the fluid mechanics of a wire of density ρ_s moving in a fluid of density ρ and determined that β and β' are given by

$$\beta = k\frac{\rho}{\rho_s} \tag{4}$$

and

$$\beta' = k'\frac{\rho}{\rho_s} \tag{5}$$

where k and k' are given by

$$k = -1 + 2\mathrm{Im}(\boldsymbol{A}) \tag{6}$$

and

$$k' = 2\mathrm{Re}(\boldsymbol{A}) \tag{7}$$

where Im and Re identify the imaginary and real components, respectively. In eqs 6 and 7, $\boldsymbol{A}$ is a complex quantity given by

$$\boldsymbol{A} = \mathbf{i}\left\{1 + \frac{2K_1(\sqrt{\Omega\mathrm{i}})}{\sqrt{\Omega\mathbf{i}}K_0(\sqrt{\Omega\mathrm{i}})}\right\} \tag{8}$$

where

$$\Omega = \frac{2\pi f \rho R^2}{\eta} \tag{9}$$

In eq 8, K_0 and K_1 are modified Bessel functions of zeroth and first order, and Ω is related to the Reynolds number that characterizes the flow around the cylindrical wire of radius R in a fluid of viscosity η.

Equations 4 through 9 were obtained with four assumptions. (1) The radius of the wire R must be small in comparison with the length L of the wire (typically on the order of 0.001); in our case, this ratio was $(R/L) = 0.0006$ for the 0.05 mm diameter wire and 0.0018 for the 0.15 mm diameter wire. (2) The isentropic compressibility κ_S of the fluid is negligible (for methylbenzene $\kappa_S = (\rho u^2)^{-1} \approx 6 \times 10^{-10}$ Pa and was estimated assuming the speed of sound $u = 1304\ \mathrm{m{\cdot}s^{-1}}$ and density $\rho = 862\ \mathrm{kg{\cdot}m^{-3}}$ reported by Vyas and Nautiyal[53]). (3) The ratio of the inner radius of the body containing the fluid R_f to that of the wire R is greater than 45 for Reynolds numbers <100 (in our case, $R_f/R = 100$ for the 0.05 mm diameter wire and 50 for the 0.15 mm diameter wire) so that the uncertainty in viscosity arising from the location of the boundary relative to the wire is less than 0.5 %.[54,55] (4) The amplitude of oscillation $\epsilon \ll R$; in our case, ϵ/R is estimated to be <0.03. All of these conditions must be satisfied in any implementation of the method, and in the remainder of this paper, we will show that is so for this viscometer.

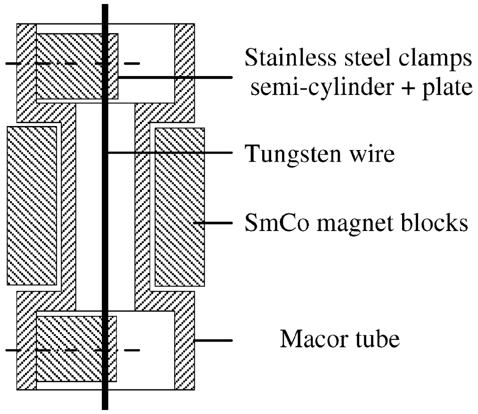

Figure 1. Schematic cross section of the vibrating wire viscometer showing the holder constructed from MACOR, the SmCo magnets, and the nonmagnetic wire clamps.

Apparatus and Experimental Procedure

The vibrating wire viscometer is, in principle, an absolute device that, in theory, requires no calibration. However, in practice, the wire radius and internal damping cannot be determined to sufficient accuracy by independent methods, hence they are determined by calibration. A measurement of the resonance bandwidth in vacuum is used to obtain the internal damping Δ_0, whereas a measurement in a reference material, such as methylbenzene, at known temperature and pressure is used to obtain the wire radius R.

The vibrating wire, shown schematically in Figure 1, has a wire that is about 40 mm long and a nominal diameter of either (0.05 or 0.15) mm. The wire used was tungsten, which despite the surface roughness is the preferred material because both Youngs' modulus E ($\approx$ 411 GPa) and density ρ_s ($\approx 19\,300\ \mathrm{kg{\cdot}m^{-3}}$) are high relative to the values for other materials. The former provides a stable resonance, whereas the latter provides sensitivity to the fluid around it through the ratio ρ/ρ_s in eqs 4 and 5. The effect of surface roughness is negligible provided the amplitude of vibration is small and the Reynolds number is less than 100. The wires used in this experiment were obtained from Goodfellow (Cambridge, U.K.) with a mass fraction purity of > 99.95 %. The wire was cold drawn; consequently, the cross section had elliptical rather than circular symmetry, which resulted in the resonance appearing as a doublet. Further discussion of the effect of both different wire materials and surface roughness on the measurement of viscosity can be found in ref 37.

The position at which the wire is clamped needs to be geometrically well defined because rotation can give rise

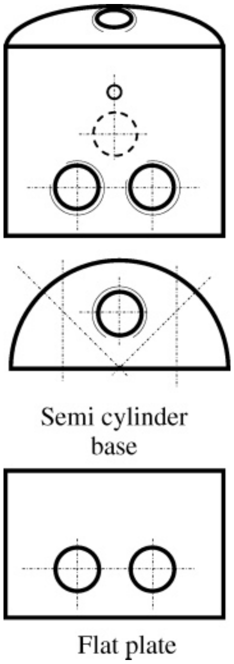

Figure 2. Schematic of the two clamp parts fabricated from nonmagnetic austenitic stainless steel type 316. (Top) side view of the semicylindrical base. (Middle) end view of the semicylindrical clamp base. (Bottom) plate that is screwed to semicircular part clamping wire between.

to additional resonance frequencies. Therefore, clamps are used that restrict, as much as possible, motion to one plane that is perpendicular to the magnetic field. Each clamp, shown in Figure 2, was fabricated from austenitic stainless steel grade 316 as two pieces, one semicylindrical with a diameter of 8 mm and a height of 8 mm and the other a flat plate. Both clamping surfaces were polished. To restrict the motion to one plane, the wire was clamped within a slot. The magnetic field was then aligned so as both to preferentially excite one of the two components arising from noncircular symmetry and to vibrate the wire in a plane perpendicular to the slot. This configuration was preferred to define the wire length clearly.

The wire was clamped between the two plates at one end with two stainless steel screws. The other end of the wire passed through the second open clamp and was attached to a mass for about 24 h, after which time the lower clamp was carefully tightened and the mass removed. The selection of the tension mass was based on the desired fundamental resonance frequency f_r that was estimated, neglecting the wire stiffness, from

$$f_{\mathrm{r}} \approx \frac{1}{2L}\sqrt{\frac{mg}{\rho_{\mathrm{s}}\pi R^2}} \tag{10}$$

where m is the mass, g is the local acceleration of free fall, and L is the wire length. The tension mass was selected both to determine the fundamental resonance frequency and maintain the tension between 20 % and 50 % of its tensile yield (about 720 MPa). The resonance frequency was estimated to be about 1500 Hz from eq 10 for $m \approx 500$ g and the tungsten (W) wire diameter = 0.15 mm.

The two clamps were separated from each other by a tube fabricated from MACOR (machinable glass ceramic) obtained from Wesgo Ceramics (Hayward, CA). The electrical resistivity of this material is $>10^{16}\ \Omega\cdot$m, and the linear thermal expansion coefficient is $13 \times 10^{-6}\ \mathrm{K}^{-1}$, which is about 4 times that of tungsten at $T = 298$ K. Two tubes were fabricated, both having an outer diameter of 10 mm; one had an inner diameter of 5 mm that was used with the 0.05 mm diameter wire, and the other had an inner diameter of 7.5 mm that was used with the 0.15 mm diameter wire to maintain adherence to assumption 3.[54,55] At each end of the tube was a step, shown in Figure 1, that was used to locate and retain the wire clamp.

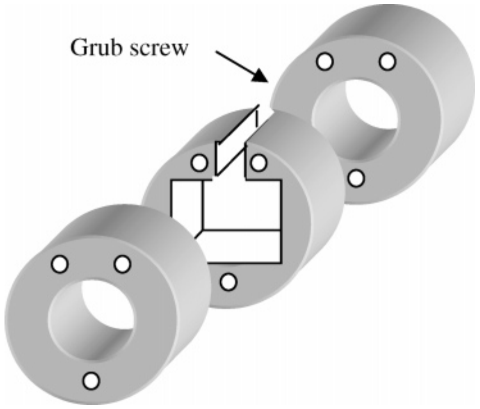

Figure 3. Isometric projection of the rare-earth permanent magnet holders, constructed from type 413 stainless steel, consisting of one center piece with a rectangular cutaway to accommodate magnets and two end rings to clamp it with the tube and wire holder.

The magnetic field was provided by two rectangular magnets, each with a length $L_B = 31$ mm located symmetrically along the wire of length L so that the ratio $L/L_B = 1.3$ was sufficient to suppress the second and third harmonics.[37] The magnets were supplied by Magnet Sales & Services (U.K.), fabricated from $\mathrm{Sm_2(Co, Fe, Cu, Zr, Hf)_{17}}$ (known commonly as Sm2Co17 or 2:17) for which the Curie temperature is about 1098 K and the maximum operating temperature is about 623 K with a temperature dependence of the field strength B_r of $(1/B_r)(\mathrm{d}B_T/\mathrm{d}T) \approx -0.0004\ \mathrm{K}^{-1}$. This material was preferred because, compared with other magnet materials, the upper operating temperature is higher (by about 100 K) and the rate at which the field decreases with increasing temperature is lower. In this arrangement, the magnetic field was exposed to the fluid and exerted a field of about 0.3 T on the wire, so the largest force applied at a current of 3 mA was about 36 μN. The magnet blocks were encapsulated in an envelope assembly constructed from three martensitic stainless steel grade 413 rings, shown in Figure 3, that consists of one center piece, with a rectangular cut-away, to accommodate the two magnet blocks and two end rings that clamp it to both the tube and wire holder. The grub screw shown in Figure 3 was used to lock the wire holder after tuning the system to minimize one, usually the lower, resonance frequency.

The assembled vibrating wire was placed in a pressure vessel, shown in Figure 4, fabricated from nonmagnetic austenitic stainless steel grade 316 with a maximum operating pressure of up to 60 MPa at the highest operating temperature. The fluid flowed into and out of the apparatus through two ports located at the bottom and top of the vessel, respectively. Electrical connections through the pressure vessel to the wire were made with a Conax Buffalo electrical feedthrough. The vessel was placed in an oil-filled stirred fluid bath whose temperature was controlled with a precision of ±0.01 K.

The temperature of the pressure vessel was determined with a four-wire industrial 25 Ω platinum resistance thermometer inserted in a well located in the pressure vessel. This thermometer had been calibrated against a 25

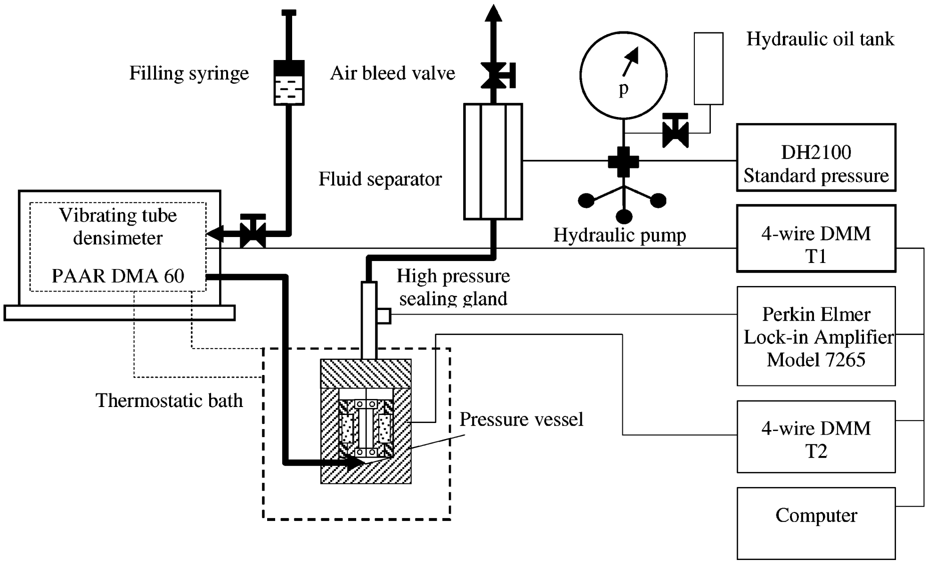

Figure 4. Experimental setup for viscosity measurements, including a high-pressure densimeter for potential simultaneous measurements.

Ω standard platinum resistance thermometer that had been calibrated on ITS-90. The resistance was determined with a multimeter and converted to temperature with an uncertainty, including the calibration, of about ±0.01 K.

Pressure was generated with a hydraulic pump and measured, in the range from (0.1 to 40) MPa, with a transducer that was calibrated against a force balance dead-weight gauge (Desgranges et Huot model 21000) with an uncertainty of ±0.05 MPa.

A sinusoidal voltage generated by a lock-in amplifier (PerkinElmer model 7265), with a maximum output of 5 V ac, was passed through a variable resistance of about 1 kΩ connected in series to the tungsten wire so as to provide a constant current source. The voltage was varied between (0.005 and 2000) mV and the resistance was varied between (0.1 and 5) kΩ to maintain the amplitude of the wire motion, estimated from the applied force and viscosity, to be less than 10 % of its radius. Thus, for a wire with a diameter of 0.15 mm in vacuum a driving current of 5 μA was used. A 0.1 mA current was required when immersed in methylbenzene; 0.5 mA, when submerged in N10; and 3 mA, when the wire was surrounded by N100, For the 0.05 mm diameter wire immersed in methylbenzene, a driving current of 0.4 mA was used. The W wire $\{c_p(\mathrm{W}, 298\ \mathrm{K}) \approx 133\ \mathrm{J{\cdot}kg^{-1}{\cdot}K^{-1}}$ and $\rho(\mathrm{W}, 298\ \mathrm{K}) \approx 19\,300\ \mathrm{kg{\cdot}m^{-3}}\}$ of diameter 0.15 mm and mass 14×10^{-6} kg has a resistance of order 1 Ω, and in vacuum, a current of 5 μA results in a temperature increase of < 3 μK over the 200 s data acquisition time. When the 0.15 mm diameter wire was immersed in N100 $\{c_p(298\ \mathrm{K}) \approx 2\ \mathrm{kJ{\cdot}kg^{-1}{\cdot}K^{-1}}$, $\rho(298\ \mathrm{K}) \approx 840\ \mathrm{kg{\cdot}m^{-3}}$, and $\kappa \approx 120\ \mathrm{mW{\cdot}m^{-1}{\cdot}K^{-1}}\}$, with the highest current of 3 mA, over the 200 s acquisition time, we estimated that the fluid temperature would rise by 0.7 mK {assuming that all electrical energy is dissipated into the fluid volume enclosed by the wire holder (about $1.8 \cdot 10^{-6}\ \mathrm{m^3}$)}, and the resulting error in viscosity at $T = 313$ K, where $\eta = 100$ mPa·s and $\mathrm{d}\eta/\mathrm{d}T \approx -3.8\ \mathrm{mPa{\cdot}s{\cdot}K^{-1}}$, would be ≈ 0.003 %. For both the other fluids and the 0.05 mm diameter wire, the estimated error in viscosity arising from self-heating is ≪ 0.003 %. The temperature rise resulting from the wire motion within the fluid is always negligible.

The frequency generated by the synthesizer was stepped over the resonance frequency of the wire, and the in-phase and quadrature voltages $\boldsymbol{V}$ that included the motional emf $\mathbf{V}_2$ were determined with the lock-in amplifier over the frequency range $(f_\mathrm{r} \pm 5g)$, where g is half the resonance line width at a frequency 0.707 times that of the maximum amplitude. The frequency sweep started at $(f_\mathrm{r} - 5g)$ with positive frequency steps to $(f_\mathrm{r} + 5g)$ and then with negative increments to $(f_\mathrm{r} - 5g)$ and took about 200 s for acquisition. Prior to acquiring $\mathbf{V}$, the majority of the contribution arising from drive voltage $\boldsymbol{V}_1$ was removed by setting the lock-in offset voltage at $f < (f_\mathrm{r} - 5g)$. The temperature was measured at each acquisition frequency and then averaged.

Three fluids were used for the measurements. Methylbenzene from Merck BDH Ltd had a mass fraction, determined by gas chromatography using both thermal conductivity and flame ionization detectors, of > 0.99917. There were two major impurities, benzene of mass fraction of 0.00031 and water of mass fraction 0.00023. The two viscosity calibration standards N10 and N100 (nominal viscosities of (14 and 240) mPa·s at $T = 298$ K and $p = 0.1$ MPa) were obtained from Paragon Scientific Ltd (U.K.) with assigned lot numbers 531606 and 131202, respectively. The supplier measured the kinematic viscosity for both N10 and N100 at temperatures between (293 and 373) K using long-capillary Master viscometers according to ASTM D 2164. The supplier also provided density values at all temperatures measured in accordance with ASTM D 1480. The uncertainty in the kinematic viscosity was ±0.25 % relative to water, and the uncertainty in the density was ±0.02 %.

The complex resonance frequency was measured with the wire immersed in methylbenzene at temperatures of (298, 323, and 348) K; for the 0.05 mm diameter wire, the measurements were performed at pressures in the range from (0.1 to 11) MPa, whereas for the 0.15 mm diameter wire measurements were made at pressure in the range from (0.11 to 40) MPa. Over these temperature and pressure ranges, the resonance frequency increased with increasing temperature and declined with increasing pressure to cover the range from (1.85 to 2.41) kHz for the 0.05 mm diameter wire and the range from (0.80 to 1.55) kHz for the 0.15 mm diameter wire; both values are within the range estimated from eq 10. The resonance quality factor $Q\ \{= f/(2g)\}$ for the 0.05 mm diameter wire varied from 23 at $\eta(298\ \mathrm{K}, 10\ \mathrm{MPa}) \approx 0.6$ mPa·s to 37 at $\eta(298\ \mathrm{K}, 0.1\ \mathrm{MPa}) \approx 0.33$ mPa·s, whereas for the 0.15 mm diameter wire Q varied from 41 at $\eta(298\ \mathrm{K}, 40\ \mathrm{MPa}) \approx 0.75$ mPa·s

Table 2. Viscosity η of Methylbenzene Measured with the 0.05 mm Diameter Vibrating Wire as a Function of Temperature T and Pressure p along with Values of Density ρ Determined from the Correlation of Assael et al.[13]

T/K	p/MPa	ρ/kg·m^{-3}	η/mPa·s
298.15	0.1	862.6	0.5550[a]
	2.11	864.1	0.5649
	4.10	865.7	0.5743
	6.10	867.2	0.5850
	8.11	868.7	0.5937
	10.11	870.1	0.6024
323.15	0.1	839.2	0.4188
	2.08	841.0	0.4290
	4.11	842.8	0.4348
	6.00	844.4	0.4409
	8.12	846.2	0.4493
	10.05	847.9	0.4550
	12.21	849.6	0.4637
	15.00	851.9	0.4736
348.15	0.1	815.2	0.3331
	2.12	817.3	0.3382
	12.11	827.1	0.3664
	15.21	830.0	0.3759

[a] Calibration point.

Table 3. Viscosity η of Methylbenzene Measured with a 0.15 mm Diameter Vibrating Wire as a Function of Temperature T and Pressure p along with Values of Density ρ Determined from the Correlation of Assael et al.[13]

T/K	p/MPa	ρ/kg·m^{-3}	η/mPa·s
298.15	0.1	862.6	0.5551[a]
	10	870.1	0.6003
	20	877.1	0.6445
	30	883.6	0.6953
	40	890.0	0.7510
323.15	0.1	839.2	0.4223
	10	847.8	0.4573
	20	855.8	0.4945
	30	863.2	0.5313
	40	870.4	0.5718
348.15	0.1	815.2	0.3285
	10	825.2	0.3583
	20	834.4	0.3875
	30	842.7	0.4165
	40	850.7	0.4483

[a] Calibration point.

to 90 at η(298 K, 0.1 MPa) ≈ 0.33 mPa·s; at a certain viscosity, the ratio of Q obtained from the 0.15 mm diameter to that of the 0.05 mm diameter wire is about 2.4. Solely on the basis of these values of Q, we suggest that the upper operating viscosity of the 0.15 mm diameter wire is greater than that of the 0.05 mm diameter wire. For both N10 and N100, measurements were performed with the 0.15 mm diameter wire at a pressure of 0.1 MPa and at temperatures corresponding to the temperatures at which the density and viscosity were measured by the supplier. For N10 and N100, Q decreased from 36, with N10 at 373 K and η(373 K, 0.1 MPa) ≈ 2 mPa·s, to 3.2, with N100 at η(313 K, 0.1 MPa) ≈ 99 mPa·s. Measurements were also performed with N100, where η(305.8 K, 0.1 MPa) ≈ 149 mPa·s and Q was found to be 2.3 and η(305 K, 0.1 MPa) ≈ 200 mPa·s and Q was found to be 1.9.

For each fluid temperature and pressure, the measured $\mathbf{V}$ was replicated by adjusting a, b, and c of eq 2, f_0 of eq 3, and η of eq 9 using R and Δ_0 from the calibration and the appropriate density. This numerical procedure separates complex voltages $\mathbf{V}_1$ and $\mathbf{V}_2$ utilizing the different frequency dependence of eqs 2 and 3, respectively.

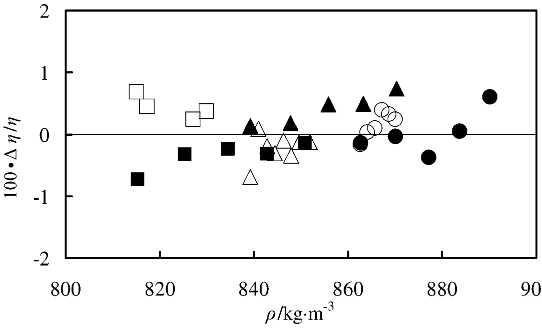

Figure 5. Fractional deviation $\Delta\eta/\eta = \{\eta(\text{exptl}) - \eta(\text{calcd})\}/\eta(\text{calcd})$ of the viscosity for methylbenzene η(exptl) measured as a function of T and p from the correlation of Assael et al.[13] η(calcd) as a function of density ρ. ○, T = 298.15 K with wire diameter ≈ 0.05 mm; ●, T = 298.15 K with wire diameter ≈ 0.15 mm; △, T = 323.15 K with wire diameter ≈ 0.05 mm; ▲, T = 323.15 K with wire diameter ≈ 0.15 mm; □, T = 348.15 K with a wire diameter ≈ 0.05 mm; and ■, T = 348.15 K with wire diameter ≈ 0.15 mm. The scale of the ordinate axis is equal to the cited uncertainty of the correlation.[13]

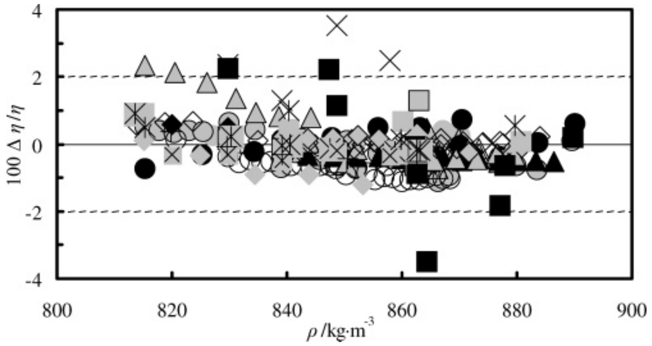

Figure 6. Fractional deviation $\Delta\eta/\eta = \{\eta(\text{exptl}) - \eta(\text{calcd})\}/\eta(\text{calcd})$ of the viscosity η(exptl) for methylbenzene from the correlation reported by Assael *et al.*[13] η(calcd), at T from (298 to 348) K and p from (0.1 to 40) MPa, as a function of density ρ. ●, our results with wire diameter ≈ 0.15 mm; gray filled circle, our results with wire diameter ≈ 0.05 mm; gray filled circle with black outline, ref 23; ○, ref 8; △, ref 18; ▲, ref 12; gray filled triangle with black outline, ref 33; gray filled triangle, ref 58; ◇, ref 59; ◆, ref 60; gray filled diamond with black outline, ref 61; gray filled diamond, ref 62; □, ref 64; gray filled square, ref 65; gray filled square with black outline, ref 66; ■, ref 67; gray filled square containing cross, ref 68; gray filled square containing ×, ref 69; cross, ref 70; ×, ref 71; +, ref 72. The dashed lines are the estimated uncertainty of ref 13.

Measurements of $\mathbf{V}$ in vacuum were used to obtain Δ_0, whereas measurements in methylbenzene at a temperature of 298.15 K and a pressure of 0.1 MPa were used to obtain R using the viscosity and density of methylbenzene obtained from Assael et al.[13] These measurements gave R = 0.0246 mm and $\Delta_0 = 4.9 \times 10^{-5}$ for the nominally 0.05 mm diameter wire, whereas for the nominally 0.15 mm diameter wire R = 0.0746 mm and $\Delta_0 = 12.2 \times 10^{-5}$.

Results and Discussion

The viscosities of methylbenzene obtained with both the (0.05 and 0.15) mm diameter wires are listed in Tables 2 and 3, respectively, and are shown as deviations from the correlation of Assael et al.[13] in Figure 5. Our results, shown in Figure 5, lie within ±1 % of the values in ref 13. Figure 6 shows the same deviations for all of the literature values listed in Table 5, except for ref 66 because they lie outside our temperature and pressure ranges. The literature values were obtained with experimental techniques that utilize

Table 4. Experimental Viscosity η_{exptl} of Two Standard Fluids N10 and N100 Determined with a Vibrating Wire Viscometer with Wire of Nominal Diameter of 0.15 mm, at $p = 0.1$ MPa along with Fractional Deviations $\Delta\eta = \eta_{\mathrm{exptl}} - \eta_{\mathrm{calcd}}$ from the Supplier Values η_{calcd} as a Function of Temperature

fluid	T/K	η_{exptl}/mPa·s	$100\Delta\eta/\eta_{\mathrm{calcd}}$
N10	373.15	2.088	0.000[a]
	323.15	6.13	0.459
	313.15	8.25	0.258
	298.15	14.1	0.531
N100	373.15	10.5	−0.694
	333.15	38.8	0.036
	323.15	60.7	0.878
	313.15	99.0	0.013

[a] Calibration point.

different principles, and thus suffer from quite different sources of systematic error, and lie within ±4 % of the values reported in ref 13.

For the two standard fluids, N10 and N100, a nominal wire diameter of 0.15 mm was used, and the results obtained with these fluids are listed in Table 4 along with the relative differences, shown in Figure 7, that never exceed ±1 %. The viscosities were obtained with $R = 0.0747$ mm determined from measurements with the standard fluid N10 at $T = 373$ K where $\eta \approx 2$ mPa·s. This value was 0.1 μm greater than that determined with methylbenzene, and had we used the lower value, the viscosities would have differed by < 0.7 % from those reported in Table 4. The measurements with N100 at a temperature of $T = 301$ K gave $\eta \approx 204.2$ mPa·s, which is 1.9 % above the value cited by the supplier, whereas at $T = 305.8$ K the vibrating wire gave $\eta \approx 146.5$ mPa·s, which lies 2.2 % below a value interpolated from the supplier's values. These differences are within the uncertainty of the measurements, which, at these viscosities, was estimated to be 3 %. These results were used to determine the upper operat-

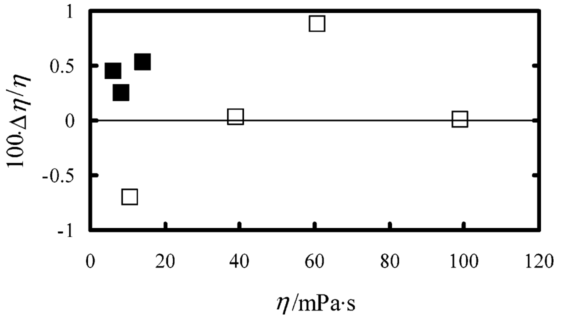

Figure 7. Fractional deviation $\Delta\eta/\eta = \{\eta(\mathrm{exptl}) - \eta(\mathrm{calcd})\}/\eta(\mathrm{calcd})$ of the viscosity for two fluids with η(298 K, 0.1 MPa) ≈ 14 mPa·s and η(298 K, 0.1 MPa) ≈ 240 mPa·s determined with the vibrating wire of diameter 0.15 mm η(exptl) from the values provided by the supplier η(calcd) as a function of viscosity η(calcd). For each fluid, the viscosity increases with decreasing temperature in the range from (298 to 373) K. □, η(298 K, 0.1 MPa) ≈ 240 mPa·s; ■, η(298 K, 0.1 MPa) ≈ 14 mPa·s.

Table 5. Measurements of the Viscosity η_{exptl} of Methylbenzene Reported in the Literature and Obtained with a Vibrating Wire (VW), Oscillating Disk (OD), Falling Body (FB), Surface Light Scattering (SLS), Rolling Ball (RB), Capillary (CP), and Torsional Vibrating Crystal (TVC) Viscometer Over a Temperature Range T at Maximum Pressures p with Estimated Uncertainty u Along with the Source of the Density and the Maximum Deviations ($\Delta\eta = \eta_{\mathrm{exptl}} - \eta_{\mathrm{calcd}}$) of η_{exptl} from the Viscosity Calculated with the Correlation Reported in Reference 13 η_{calcd} at Temperatures T between (298 and 348) K and Pressure p < 40 MPa that Cover our Range

T_{min} to T_{max} / K	p / MPa	$100\Delta\eta$ / η_{calcd}	±100·u[a]	method	density obtained from	ref
222 to 348	80	1.1	3	VW	measured simultaneously with buoyant mass	23
210 to 370	0.1	0.64	0.5	VW	ref 73	7
210 to 370	30	1.12	0.5	VW	ref 13	8
213 to 298	20	0.71	1.5	VW	ref 13	18
303 and 323	70	0.52	0.5	VW	ref 74	12
303 to 348	250	2.34	0.5	VW	ref 74	33
298 to 373	200	[b]	0.5	TVC	ref 74	56
298 to 348	110	[b]	2	TVC	ref 57	57
298 and 323	375	0.65	2	FB	ref 58	58
298 to 423	30	0.66	0.5	OD	ref 74	59
263 to 383	0.1	0.57	1	SLS	ref 75	60
255 to 323	400	0.26	1	FB	pycnometer	61
298 to 368	0.1	1.19	0.5	CP	pycnometer	62
353 to 463	0.1	out of our range	Not stated	RB	not stated	63
288 to 373	0.1	0.69	0.5	CP	pycnometer and volumeter, ref 64	64
298 to 373	500	0.91	4	FB	ref 65	65
298 to 323	500	1.29	5	FB	ref 65	66
298 to 363	100	3.49	1	FB	Anton-Paar DMA 45 and 512 at p < 40 MPa and extrapolated to $p \approx 100$ MPa, ref 67	67
298 to 348	0.1	0.92	0.5	CP	ref 74	68
218 to 378	0.1	0.48	1.4	CP	ref 13	69
298 to 373	200	0.99	0.5	TVC	ref 74	70
303 to 333	0.1	3.51	Not stated	CP	pycnometer, ref 71	71
293 to 353	0.1	0.51	0.2	CP	pycnometer, ref 72	72
298 to 348	40	0.69	0.6	VW	ref 13	this work, 0.05 mm wire
298 to 348	40	0.74	0.6	VW	ref 13	this work, 0.15 mm wire

[a] Uncertainties (u) cited by authors of the original work. [b] Only smoothing equation provided.

ing viscosity and are not reported in Table 4 or shown in Figure 7.

The values of f_0, the vacuum frequency of the wire determined from the regression analysis to eq 3, varied linearly with temperature for the 0.15 mm diameter wire of $\mathrm{d}f_0/\mathrm{d}T \approx 4\ \mathrm{Hz{\cdot}K^{-1}}$, whereas for the 0.05 mm diameter wire we observed $\mathrm{d}f_0/\mathrm{d}T \approx 9\ \mathrm{Hz{\cdot}K^{-1}}$. These values are about a factor of 2 less than the result anticipated solely from the variation in wire tension that would arise from the difference in linear thermal expansion coefficients between tungsten and Macor. The values of f_0 determined from the regression analysis to eq 3 decreased with increasing pressure with $\mathrm{d}f_0/\mathrm{d}p \approx -11\ \mathrm{Hz{\cdot}MPa^{-1}}$. We have not performed experiments to identify the source of this observed variation; plausible sources include the rotation of one wire clamp relative to the other. Had we fixed f_0 in the analysis to a value determined at $p = 0$ for each temperature, the viscosities determined would have deviated from the literature values by up to 50 %. Clearly, allowing f_0 to float in the regression is important for the accurate determination of viscosity and was included in our analysis.

Conclusions

This work demonstrates that vibrating wires can be used to measure viscosities up to 100 mPa·s with an uncertainty of < 1 % and that the upper limit of the 0.15 mm diameter wire is 200 mPa·s with an uncertainty of about 3 %.

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