Operation of a Vibrating Wire Viscometer at Viscosities Greater than 0.2 Pa•s: Results for a Certified Reference Fluid with Nominal Viscosity at $T = 273$ K and $p = 0.1$ MPa of 0.652 Pa•s while Stagnant and a Fluid of Nominal Viscosity of 0.037 Pa•s while Flowing

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A vibrating wire viscometer formed from a tungsten wire with radius of about 0.125 mm has been used to measure the viscosity of certified reference fluid S60 at temperatures between (273 and 373) K at a pressure of 0.1 MPa where the viscosity varies from (0.0062 to 0.652) Pa s and the resonance quality factor *Q* lie between (20.8 and 1.02). Our results differ from the values cited by the supplier by less than ± 3 % over the whole range of *Q*, *T*, and *p*. We have also measured the viscosity of certified reference fluid S20, with η(293 K, 0.1 MPa) ≈ 37 mPa•s, while stagnant at pressures below 69 MPa and temperature between (298 and 423) K and also while flowing S20 at volumetric rates up to 66 $\mathrm{cm^3{\cdot}s^{-1}}$. The values obtained in stagnant S20 differed insignificantly from those while flowing and suggest there is no additional systematic error arising from flow in the viscosity obtained from an axially symmetric vibrating wire viscometer. The working equations for a vibrating wire have been used to estimate *Q* as a function of viscosity up to 1 Pa•s for wires with radii between (0.05 and 0.3) mm. The calculated *Q* compares favorably with the observed values and suggests a wire of radius about 0.2 mm can be used to measure the viscosity of fluids up to 1 Pa•s.

Introduction

This paper is concerned with a viscometer formed from a wire vibrating normal to its axis and held taut between two clamps.[1,2] The majority of the measurements reported with a vibrating wire and summarized up to early 2005 in ref 3 have been obtained with fluids of viscosity in the range (0.008 to 6) mPa•s. However, there are a few measurements at higher viscosities including those reported by Charles et al.[4] for the viscosity of glycol at about 53 mPa•s obtained with wires of nominal radius between (0.1 and 0.18) mm, Gourgouillon et al.[5] measured the viscosity of supercritical fluid-saturated polymer (PEG 400) with values of about 20 mPa•s with a wire of nominal radius of 0.1 mm, and Caudwell[6] measured the viscosity of hydrocarbon mixtures with values up to 16 mPa•s with a nominal wire radius of 0.05 mm. More recently, Caetano et al.[7] developed a vibrating wire viscometer with a wire radius of 0.1997 mm that was determined by calibration with water at $T = 293.15$ K and $p = 101.325$ kPa. They used the instrument to determine the viscosity of several fluids, including certified reference material S60, with viscosities in the range (0.5 to 135) mPa•s and stated the expanded ($k = 2$) uncertainty in their measurements was less than 0.6 % over this range.

Kandil et al.[3] have constructed a vibrating wire viscometer with a nominal wire radius of 0.075 mm and validated the operation of the instrument with certified reference materials N10 and N100 at viscosities in the range (2 to 100) mPa•s, with a standard uncertainty of less than 0.6 %, at temperature in the range (298 to 373) K. The upper operating viscosity of this instrument was determined with measurement on certified reference material N100 at a temperature of $T = 301$ K where $\eta \approx 200$ mPa•s, for which the $Q = 1.9$, and the measured viscosity had an uncertainty of about 3 % and the observed value differed from an interpolation of the cited values by about 2 %.

Lundstrom et al.[8] used a wire with $R \approx 0.0479$ mm to measure the viscosity of certified references fluids N10 and S20 at temperatures between (298 and 393) K at pressure below 55 MPa. The measurements with N10 were conducted at viscosities between (4.8 and 60) mPa•s that gave *Q* between 11 and 2, while for S20 the viscosity varied from (2.7 to 73) mPa•s and the *Q* decreased with increasing viscosity from 17 to 2. Sopkow et al.[9] measured the viscosity of N10 and S20 obtained from batches different to those used by Lundstrom et al.[8] At $p = 0.1$ MPa the viscosities of both N10 and S20 at temperatures of (293 and 313) K and 313 K, respectively, reported in ref 9 differ from those reported by Lundstrom et al.,[8] on different batches, by less than 1.2 % for N10 and 0.5 % for S20 that is within the combined uncertainty arising from the difference between the

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cited values for the lots and the uncertainty of the supplier's cited measurements for the reference fluids of ± 0.25 %. At pressures greater than 0.1 MPa, the results for N10 agreed with those of ref 8 within ± 4 % while for S20 the viscosities differed by ± 4.5 %. For both N10 and S20 these differences are about a factor of 2 greater than the expanded uncertainty of either series of measurements but are within the combined uncertainty of both measurements excluding the known but less than 1 % differences between the viscosities of the lots at $p = 0.1$ MPa. The differences in the densities of the different lots either cited by the supplier or obtained from the modified Tait equation[10] representation of the results of ref 8 (eqs 7 and 8 of ref 8 with coefficients of Table 5 of ref 8) are less than 0.6 % that is about 3 times the expanded uncertainty of 0.2 % for the measurements reported in ref 8; eqs 7 and 8 represented the densities of ref 8 with an average percentage deviation that never exceeded 0.04 %. The up to 0.6 % additional uncertainty in the density propagates to an uncertainty of less than 0.3 % in the viscosity determined from the vibrating wire. The assigned expanded uncertainty of the viscosity measurements with a wire clamped at both ends is ± 2 %.[7]

In the petroleum industry, measurements of the density and viscosity of petroleum reservoir fluids are required to determine the value of the produced fluid and production strategy. These thermophysical properties are also useful for the design of separators and process equipment and to control production processes. To measure the density and viscosity of petroleum fluids requires a transducer that can operate up to reservoir temperatures and pressures, typically below 473 K and 200 MPa, respectively, and to guide value and exploitation calculations with sufficient rigor, provide results with an accuracy of about ± 1 % in density and ± 10 % in viscosity. Necessarily, these specifications place robustness as a superior priority to accuracy for the design. There are numerous methods to measure the viscosity of fluids and these have been reviewed recently by Wakeham et al.[11] Some of the methods that are used to measure viscosity in the petroleum industry require calibration over the viscosity, temperature, and pressure range for which they are to be used. Therefore a calibrant is required.

Here we report measurements of the viscosity of a reference fluid for viscosity, known as S60, with a vibrating wire of $R \approx 0.125$ mm at temperatures between (273 and 373) K over which the certified values of viscosity cover the range (0.0062 to 0.652) Pa•s. These measurements extend the operating range of the vibrating wire by more than a factor of 3 over those reported by Kandil et al.,[3] for a wire with $R \approx 0.0.75$ mm. We have used the vibrating wire viscometer working equations to estimate the quality factor of the resonance of the wire as a function of wire radius and viscosity of the fluid in which the wire is immersed. These results are in good agreement with our measured resonance quality factors as well as those reported by Lundstrom et al.,[8] Sopkow et al.,[9] and Kandil et al.[3] and suggest that a vibrating wire can be used to measure the viscosity up to 1 Pa s.

If a vibrating wire were inserted within a tubular that, for example, connected two vessels, and the fluid flowed between them then the viscometer could also be exposed to flowing fluid. In that case, one might speculate the shear experienced by an axially oriented vibrating wire subjected to a flowing viscous fluid to introduce at least noise in the observed $\boldsymbol{V}$ and also a systematic error in the determined viscosity. To experimentally determine the effect of flow, we have performed measurements of the viscosity of another certified material for viscosity S20, with nominal viscosity of 0.037 Pa•s at $T = 293$ K and $p = 0.1$ MPa, while flowing at volumetric flow-rates up to 66 $\text{cm}^3\cdot\text{s}^{-1}$; in the tube of internal diameter 5.5 mm this upper flow rate corresponds to a velocity of 0.28 $\text{m}\cdot\text{s}^{-1}$. Over these flow rates at a viscosity of about 0.04 Pa•s there was no observable systematic error in the viscosity obtained from the vibrating wire.

Theory

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 while the resonance line width is related to the ratio of the 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 since 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 the wire can be clamped at both ends. In this case, provided the resonance frequency is stable to better than the required resolution of the measurement over the time needed to acquire the resonance data knowledge of the band width suffice to determine the viscosity for a fluid when the density is known.

The wire is forced to vibrate by passing sinusoidal current I through the wire in the presence of a steady-steady magnetic field B perpendicular to the wire that results in a transverse oscillation. The force F experienced by the wire is given by

$$F = ILB \tag{1}$$

where L is the length of the wire. (Strictly we measure the potential difference and provided the current tends to zero this is equal the emf.) Movement of the wire creates a change in the magnetic flux through the voltage loop, generating an electromagnetic emf:

$$V_2 = \frac{\mathrm{d}\Phi}{\mathrm{d}t} = \frac{\mathrm{d}(\int B\cdot \mathrm{d}S)}{\mathrm{d}t} = B\frac{\mathrm{d}S}{\mathrm{d}t} \tag{2}$$

where Φ is the magnetic flux, S is the area of the wire loop which experiences the magnetic field, and t is the time. The derivative of the area S with respect to time can be approximated by dividing the maximum area created by the sinusoidal-like deflection of the wire by a quarter period (period denoted as T_0), resulting in

$$\frac{\partial S}{\partial t} \approx \frac{8AL}{\pi T_0} \tag{3}$$

where A is the maximum amplitude of the wire's deflection normal to its axis. The motional V_2 will then be this quantity multiplied by the magnetic field strength:

$$V_2 \approx B\frac{8AL}{\pi T_0} \tag{4}$$

The amplitude of the wire motion is proportional to the force and thus the field strength V_2 will be proportional to the square of the magnetic field:

$$V_2 \approx \frac{8IL^2B^2}{\pi T_0} \tag{5}$$

Equation 5 suggests small increases in the magnetic field can greatly increase the signal-to-noise ratio. Furthermore, the actual motional emf V_2 produced by a resonating wire would require multiplication of eq 5 above by the quality factor Q.

The fluid mechanics describing the motion of a thin metallic wire clamped under tension between two fixed points and immersed in a fluid have been presented elsewhere; we merely summarize some of the relevant conclusions here.[12,13] The net voltage $\boldsymbol{V}$ measured across the vibrating wire that is immersed in a fluid is given by the sum of the voltage $\boldsymbol{V}_1$ arising from the electrical impedance of the effectively stationary wire and $\boldsymbol{V}_2$ that represents the motional emf by

$$\boldsymbol{V} = \boldsymbol{V}_1 + \boldsymbol{V}_2 \tag{6}$$

where $\boldsymbol{V}_1$ is represented empirically by

$$\boldsymbol{V}_1 = a + ib + icf \tag{7}$$

and f is the frequency at which the wire is driven. In eq 7 a, b, and c are adjustable parameters determined by regression with the measured complex voltage and account for the electrical impedance of the wire and absorb the offset used in the lock-in amplifier to ensure the voltage signal is detected in the most sensitive range. $\boldsymbol{V}_2$, given by[12−17]

$$V_2 = \frac{\Lambda f i}{{f_0}^2 - (1 + \beta)f^2 + (\beta' + 2\Delta_0)f^2 i} \tag{8}$$

and relates the measured quantity to the fluid properties. In eq 8, 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.[1,2] analyzed the fluid mechanics of a wire of density ρ_s vibrating in a fluid of density ρ and determined that β and β' are given by

$$\beta = k\frac{\rho}{\rho_s} \tag{9}$$

and

$$\beta' = k'\frac{\rho}{\rho_s} \tag{10}$$

where k and $k'$are given by

$$k = -1 + 2\mathscr{I}(A) \tag{11}$$

and

$$k' = 2\mathscr{R}(A) \tag{12}$$

In eqs 11 and 12, A is a complex quantity given by

$$A = i\left\{1 + \frac{2K_1(\sqrt{\Omega i})}{\sqrt{\Omega i}K_0(\sqrt{\Omega i})}\right\} \tag{13}$$

where

$$\Omega = \frac{2\pi f \rho R^2}{\eta} \equiv \frac{N_{\mathrm{Re}}}{\epsilon} \tag{14}$$

In eq 13, K_0 and K_1 are modified Bessel functions of zeroth and first order, and Ω (the ratio of linear inertial to viscous forces) is related to the Reynolds number N_{Re} that characterizes the flow around the cylindrical wire of radius R in a fluid of viscosity η where N_{Re} equals $\upsilon R\rho/\eta$, υ being the average fluid velocity. In equation eq 14, ϵ is a dimensionless displacement (A/R) for a periodic oscillation.

Equations 8 through 14 were obtained with the following assumptions: (i) 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 $(R/L) = 0.0006$ for a 0.050 mm diameter wire and 0.0018 for a 0.150 mm diameter wire; (ii) the isentropic compressibility κ_S of the fluid is negligible; (iii) 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 $R_f/R = 150$ for the 0.05 mm diameter wire and 50 for the 0.15 mm diameter wire) then uncertainty in viscosity arising from the location of the boundary relative to the wire is less than 0.5 %; and (iv) the amplitude of oscillation $\epsilon \ll R$, in our case ϵ/R is estimated < 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.

Experimental Procedures

Vibrating Wire Viscometer. The wire holder and pressure vessel used for the measurements reported here have been described by Lundstrom et al.[8] In ref 8 a wire of $R \approx 0.048$ mm was used. For this work, the wire was replaced with tungsten of nominal radius 0.125 mm. To tension the wire, the wire holder was positioned vertically, the wire slid through the clamps and secured at the upper while a 0.6 kg mass was attached to the wire protruding beneath the lower clamp. This mass applied a constant force of about 6 N to the wire, which was secured in the bottom clamp after an elapsed time of about 24 h. Prior to commencing measurements, the vibrating wire was aligned within the magnetic flux so as to preferentially excite the highest frequency orthogonal mode of the fundamental oscillation that arises from the non circular cross section of the wire. This was achieved by rotating the entire pressures vessel housing within the magnetic fluid as described by Lundstrom et al.[8] The tensioned wire was cycled between temperatures of (298 and 473) K until the resonance frequency at $T = 298$ K varied by less than a few Hz between each thermal cycle.

A sinusoidal voltage was generated by a lock-in amplifier (Stanford Research Systems model 850), with a maximum output of 5 V ac, and was passed through a 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 (5 and 5000) mV to maintain the amplitude of the wire motion, estimated from the applied force, magnetic flux, and viscosity to be less than 10 % of its radius. Thus, in vacuum a driving current of 5 μA was used and as much as 50 mA (aided by power amplifier) was used when the wire was submerged in methylbenzene, S20, or S60. The W wire $\{c_p(\mathrm{W}, 298\ \mathrm{K}) \approx 133\ \mathrm{J{\cdot}kg^{-1}{\cdot}K^{-1}}$ and $\rho(\mathrm{W}, 298\ \mathrm{K}) \approx 19300\ \mathrm{kg{\cdot}m^{-3}}\}$ of diameter 0.25 mm and mass $4.9{\cdot}10^{-5}$ kg has a resistance of order 1 Ω, and when immersed in S60 $\{c_p(298\ \mathrm{K}) \approx 2\ \mathrm{kJ{\cdot}kg^{-1}{\cdot}K^{-1}}$, $\rho(298\ \mathrm{K}) \approx 840\ \mathrm{kg{\cdot}m^{-3}}$, and $\kappa \approx 120\ \mathrm{mW{\cdot}m^{-1}{\cdot}K^{-1}}\}$, with the highest current of 5 mA, over the 200 s acquisition time the fluid temperature was estimated to rise by 1 mK {assuming all electrical energy dissipated into the fluid volume enclosed by the wire holder (about $1.1{\cdot}10^{-6}\ \mathrm{m^3}$)}. The resulting error in viscosity at $T = 313$ K, where $\eta = 46.3689$ mPa·s and $\mathrm{d}\eta/\mathrm{d}T \approx -2.3\ \mathrm{mPa{\cdot}s{\cdot}K^{-1}}$, would be ≈ 0.005 %. The temperature rise resulting from the wire motion within the fluid was always negligible.

Thermostat, Thermometer, and Pressure Gauge. The fluid was displaced into an evacuated pressure vessel, and the pressure of it was controlled with a positive displacement pump (Isco model 65D) with a maximum operating pressure of 138 MPa. For the viscosity measurements obtained while flowing S20 the pressure between (0.07 and 138) MPa was determined with the strain gauge transducer fitted to the positive displacement pump with an uncertainty of ± 0.69 MPa. The viscometer was placed in an air thermostat (Sigma Systems model M33M) capable of operating over the desired temperature range of (273 to 373) K. The temperature of the outer surface of the pressure vessel was determined with a K-type thermocouple with an uncertainty of ± 0.1 K. The values obtained from this thermometer differed by less than 0.3 K from those indicated by the thermostat temperature controller and this difference places an upper bound on the uncertainty in our temperature measurements. Thermal equilibrium was assumed to occur when the viscosity observed from three or more sequential measurements differed by less than the estimated standard uncertainty of ± 1 %.

Material and Measurements. Three fluids were used for the measurements. Methylbenzene from Riedel de Haën with a stated mole fraction purity greater than 0.997 containing a mole fraction of less than 0.00001 of water was used to determine the wire radius. No further chemical analysis or purification of this material was attempted. The two certified reference materials for viscosity S20 and S60 {with nominal viscosities of (0.037 and 0.140) Pa·s, respectively, at $T = 293$ K and $p = 0.1$ MPa} were obtained from Cannon Instruments with assigned lot numbers of 04201 and 5301, respectively. The supplier measured the kinematic viscosity for both S20 and S60 at temperatures between (273 and 373) K using long-capillary Master viscometers according to ASTM D2164. The supplier also provided densities over the same temperature range measured in accordance with ASTM D1480. For S60, Cannon also provided interpolation equations for the kinematic viscosity. For both S20 and S60 the expanded uncertainty in the kinematic viscosity was ± 0.25 % relative to water, for which the uncertainty at $T = 298$ K and $p = 0.1$ MPa is ± 0.25 %, and the uncertainty in the density was ± 0.02 %. When these uncertainties are combined in quadrature the expanded uncertainty in the dynamic viscosity is ± 0.35 % assuming no additional uncertainty arises from the step-up procedure.[18] However, measurements of the viscosity of reference materials between different laboratories have been reported with discrepancies of less than ± 0.1 %.[19]

The methods used to determine the resonance frequency of the wire were similar to those described by Lundstrom et al.,[8] and only significant differences are described here. The frequency source of the lock-in was amplified and split into the following two paths each of equal resistance: (i) through a 100 Ω resistor in series with the vibrating wire (that has a resistance of about 4 Ω when combined with the resistance of the 4 m long leads) so as to provide an approximately constant current source at each frequency and (ii) through a variable resistor with resistance about that of the wire. The lock-in amplified the difference between these two signal source, and this arrangement essentially nulls the signal arising from the resistance of the wire.

Results

Calibration of Internal Damping under Vacuum and Radius with Methylbenzene. When the wire was exposed to a pressure of less than 1 Pa, we assumed $p = 0$ and determined the internal damping of the vibrating wire $\Delta_0 = 1.97\ 10^{-4}$. The complex voltage $V_2(f)$ obtained over the frequency range of the fundamental mode of the vibrating wire immersed in methylbenzene (shown in Figure 1) at a viscosity of 0.590 mPa·s that was obtained from the correlation reported by Assael et al.[20] The measured $\boldsymbol{V}(f)$, with a specified density of 874 kg·m^{-3}, were represented by eqs 6 to 14 with $a = -1.40177 \cdot 10^{-5}$, $b = 3.09998 \cdot 10^{-7}$, $c = -1.33634 \cdot 10^{-10}$, $d = -1.29169 \cdot 10^{-9}$, $\Lambda = 0.000401517$, $f = 752.845$ Hz, and $\eta = 0.587$ mPa·s, which is 0.5 % below the value obtained from ref 20. Figure 2 shows the measurements performed with methylbenzene at pressures up to 55.2 MPa to determine the wire radius. A radius of (0.1253 mm) was chosen so as to minimize the difference between the measured and calculated $\boldsymbol{V}$ over the whole pressure range.

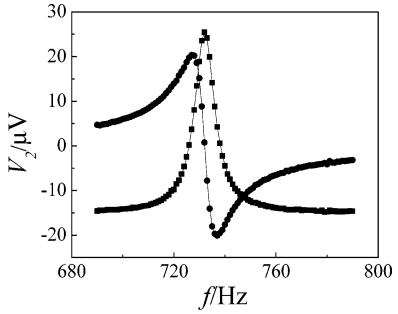

Figure 1. Motional complex voltage $\boldsymbol{V}(f)$ the frequency f of the fundamental mode of the vibrating wire immersed in methylbenzene at a viscosity of 0.590 mPa·s obtained from the correlation reported by Assael et al.[20] The measured $\boldsymbol{V}(f)$ for $R = 0.1253$ mm with a specified density of 874 kg·m^{-3} were represented by eqs 6 to 14 with $a = -1.40177 \cdot 10^{-5}$, $b = 3.09998 \cdot 10^{-7}$, $c = -1.33634 \cdot 10^{-10}$, $d = -1.29169 \cdot 10^{-9}$, $\Lambda = 0.000401517$, $f = 752.845$ Hz, and $\eta = 0.587$ mPa·s, which is 0.5 % below the value obtained from ref 20. ■, in-phase voltage; ●, quadrature voltage; —, in-phase {Re($\boldsymbol{V}$)} voltage obtained from eqs 6 to 14 with the parameters listed above; and —, quadrature {Im($\boldsymbol{V}$)} voltage obtained from eqs 6 to 14 with the parameters listed above.

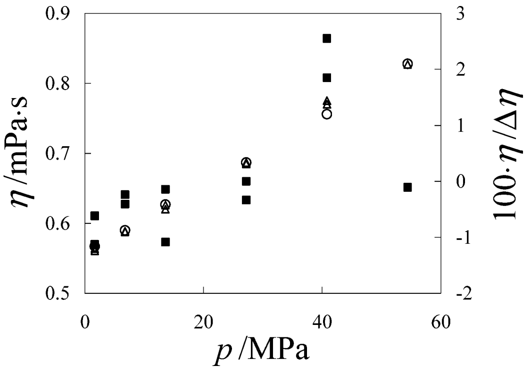

Figure 2. Calibration of the VW with methylbenzene under pressures ranging from ambient to 55.2 MPa. The measured viscosities and expected viscosities are represented with △ and ○, respectively, with magnitudes indicated on the left axis. The fractional deviation ($\Delta\eta/\eta = \{\eta(\text{exptl}) - \eta(\text{calcd})\}/\eta(\text{calcd})$) is plotted with solid squares and the magnitudes are indicated on the right axis.

The uncertainty in R is a major source of error in our measurements of viscosity, which have an estimated expanded uncertainty of ± 2 %.[3,7] Fortunately, for measurements with liquids the working equations are insensitive to the value of Δ_o and there is no need to determine this parameter with high precision. No corrections were applied to both the wire radius and density to account for variations in temperature and pressure from that of the calibration. Neglecting these variations has been estimated to introduce and additional uncertainty in viscosity of less than 0.1 %.[21] Our measurements of temperature have an uncertainty of no more than 0.3 K that when combined with, in the worst case, $(\partial\eta/\partial T)p \approx -2.3$ mPa·s·K^{-1} for S60 at $T =$

Table 1. Viscosity η_{exptl} Obtained from the Vibrating Wire Viscometers along with the Viscosity η_{ref} and Density ρ_{ref} Provided by the Supplier of Certified Reference Fluid S60 and the Relative Deviations of η_{exptl} from η_{ref} at Temperatures T and Pressures p = 0.1 MPa with Expanded Uncertainties for η_{exptl} (Confidence Interval of 0.995)

T/K	f_0/Hz	Q	ρ_{ref}/kg·m^{-3}	η_{ref}/Pa·s	η_{exptl}/Pa·s	100 $\Delta\eta/\eta$
373.15 ± 0.1	651.4	20.85	825.5	0.006219	0.00631 ± 0.00013	1.51
363.15 ± 0.1	648.7	18.06	831.8	0.007944	0.00803 ± 0.00016	1.14
353.15 ± 0.1	646.6	15.36	838.1	0.010434	0.01052 ± 0.00021	0.84
343.15 ± 0.1	645.4	12.78	844.4	0.014166	0.01430 ± 0.00029	0.93
333.15 ± 0.1	643.8	10.31	850.7	0.02000	0.02033 ± 0.00041	1.63
323.15 ± 0.1	640.6	8.12	856.9	0.02961	0.03018 ± 0.00060	1.95
313.15 ± 0.1	633.6	6.08	863.2	0.04637	0.04744 ± 0.00095	2.30
303.15 ± 0.1	618.1	4.34	869.5	0.07776	0.0802 ± 0.0016	3.17
293.15 ± 0.1	588.6	2.93	875.8	0.14167	0.1434 ± 0.0029	1.19
288.15 ± 0.1	571.9	2.35	878.9	0.19838	0.1985 ± 0.0040	0.08
283.15 ± 0.1	546.0	1.81	882.1	0.2857	0.2832 ± 0.0057	−0.88
278.15 ± 0.1	514.9	1.41	885.0	0.4243	0.4195 ± 0.0084	−1.13
273.15 ± 0.1	448.7	1.02	888.3	0.6524	0.666 ± 0.013	2.10

373 K, gives rise to an additional uncertainty in the measured viscosity of 2.55 %.

Measurements with S60. The viscosity of S60 obtained with the vibrating wire is listed in Table 1 along with the measured resonance frequency and quality factor as well as the certified values of both viscosity and density cited by the supplier at temperatures between (273 and 373) K at a pressure of 0.1 MPa. Figure 3 shows the complex voltage $\boldsymbol{V}_2(f)$ obtained when the vibrating wire was immersed in S60 at viscosities of 0.652 Pa·s (lower two curves are the in-phase and quadrature voltages) and 0.0062 Pa·s (upper two curves are the in-phase and quadrature voltages for this viscosity). For both viscosities, $\boldsymbol{V}_2(f)$ is measurable and distinguishable from the noise floor. The uncertainties, listed in Table 1, are at a confidence interval of 0.995 (k = 2) and were obtained by combining in quadrature uncertainties arising from the uncertainty of the instrument with dη/dT for viscosity; the estimated expanded uncertainty does not include the error from the fit to V_2 that increased with increasing viscosity and decreasing Q to be in the worst case 3.17 %. For the viscosity obtained from the vibrating wire viscometer the standard uncertainty is, based on the work reported in refs 3 and 7, assumed to be ± 1 %. The most significant and quantifiable contribution to the uncertainties arises from dη/dT. The derivative was estimated from the cited values and the δT listed in Table 1. For S60 the contribution to $\delta\eta$ from δT lies between (0.017 to 4.6) Pa·s {about (0.3 to 0.7) %} that decreased with increasing temperature. Clearly, for our measurements the uncertainty with which the temperature is measured is significant only at the lowest temperature studied of 273.15 K. In the absence of a chemical analysis for S60, the contribution to the uncertainty arising from the uncertainty in composition was assumed to be 0.

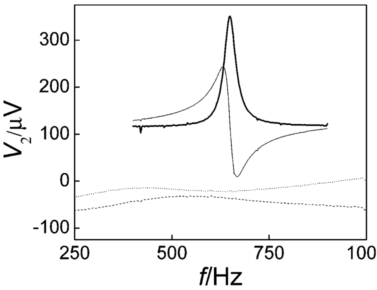

Figure 3. Motional complex voltage $\boldsymbol{V}_2(f)$ over the frequency f of the fundamental mode of the vibrating wire immersed in S60. Top: $\boldsymbol{V}_2(f)$ obtained with a cited viscosity of 0.0062 Pa·s with a Q of about 21. Thick solid line, in-phase {Re($\boldsymbol{V}_2$)} voltage; thin line, quadrature {Im($\boldsymbol{V}$)} voltage. Bottom: $\boldsymbol{V}(f)$ obtained with a viscosity of about 0.652 Pa·s with a Q of 1.02. dashed line, in-phase {Re($\boldsymbol{V}$)} voltage; and dotted line, quadrature {Im($\boldsymbol{V}_2$)} voltage.

The viscosities from Table 1 are shown relative to the cited values in Figure 4 for certified reference material S60 where the error bars are our estimated expanded uncertainty. The dashed lines are the expanded (confidence interval of 0.95) uncertainty in the viscosity of the certified material of 0.35 %. At p = 0.1 MPa the viscosities of Table 1 for S60 are, as shown in Figures 3, in reasonable agreement deviating by less than 3.2 % and typically within our expanded uncertainty (about 2 %) from the cited values albeit with a systematic undulation, and surprisingly even the result η(273.15 K, 0.1 MPa) = (0.666 ± 0.013) Pa·s lies only 2.09 % above the cited value. However, all but two of the measurements, at temperatures of (278.15 and 283.15) K, show positive differences from the cited values and the specific source of this systematic difference has not been identified, although it is plausible that they arose from either an unidentified error R or a chemical impurity, such as the presence of S20 or methylbenzene in the apparatus. No measurements were conducted to confirm either of these postulates. Caetano et al.[7] have measured, with a vibrating wire viscometer, the viscosity of S60 supplied by Paragon Scientific, UK, with a batch number of U0465, that is different to that used here. The measurements reported in ref 7 have a cited expanded uncertainty of 0.8 % and differed from their suppliers cited values by between (−0.3 and 0.1) %. The viscosities reported by Caetano et al.[7] lie, as shown in Figure 3, between −(3.7 and 1.3) below the certified values for our batch of S60

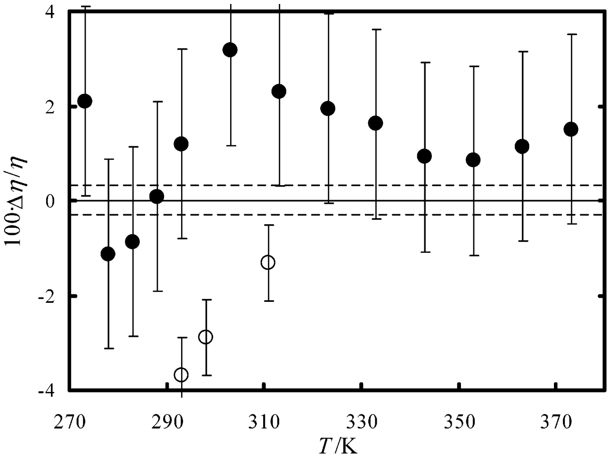

Figure 4. Fractional deviation $\Delta\eta/\eta$ = {η(exptl) − η(calcd)}/η(calcd) as a function of temperature T of the viscosity obtained with the vibrating wire η(exptl) from the value η(calcd) obtained from an interpolating equation provided by the supplier and based on their measurements. ▲, certified reference fluid S60 with error bars for our expanded uncertainty; ○, ref 7 with claimed uncertainty of 0.8 %; and - - -, the expanded uncertainty in the values cited by the supplier of certified reference fluid S60.

and have a similar trend to our deviations albeit with a ordinate offset.

Upper Operating Viscosity. The quality factor of the resonance Q is given by

$$Q = f_0/(2g_0) \tag{15}$$

where f_0 is the resonance frequency and g_0 is half the resonance line width at $2^{-1/2}$ times the maximum amplitude obtained at the resonance frequency; 2 g is also referred to as the 3 dB resonance line-width. The physical factors that determine the Q of the wire consist of the following: (1) viscosity of the fluid; (2) the density of the wire material; (3) the wire radius; and (4) the wire tension and therefore the resonance frequency. From a practical perspective items for the VW (1), (2), and (3) dominate the Q. In practice the precision of the Q measurement can be increased through regression such as with the acoustic function provided by Mehl and defined by[22]

$$V_2(f) = \frac{A_0 f}{f^2 - (f_0 + ig_0)^2} \tag{16}$$

In eq 16, A_0 is a complex constant determined by nonlinear regression. We will henceforth use f_0 and g_0 determined from the best fit of the data to eq 16 to calculate the quality factor Q for eq 15.

The upper operating viscosity of a vibrating wire viscometer is essentially determined by two factors: (i) the practical limitation that the resonance quality factor Q is large enough to provide a signal sufficiently distinguishable from the noise floor and (ii) the requirement to comply with the assumptions used to derive the working eqs 6 to 14. The second constraint can be summarized by

$$\epsilon \ll \Omega \ll \frac{1}{\epsilon^2} \tag{17}$$

where Ω is given by eq 14 and its value depends on the arbitrary selection of the bounds and ensures that the design conforms to the theoretical development of the working equation. In eq 17 ϵ is defined as the dimensionless amplitude of oscillation, that is, the transverse displacement divided by the wire radius. The second constraint normally provides a larger upper limit to the viscosity, and provided it is satisfied, the first constraint on Q becomes the critical one.

The Q of a tungsten wire with density $\rho_s = 19300\ \mathrm{kg{\cdot}m^{-3}}$ tensioned between two rigid clamps so that $f_0 = 1.2$ kHz has been calculated from eqs 6 to 17 as a function of viscosity up to 1 Pa·s for R of (0.05, 0.75, 0.1, 0.125, 0.15, 0.2, and 0.3) mm. Because Q is essentially inversely proportional to the fluid viscosity, the values of Q^{-1} obtained from the analysis are shown in Figure 5 as a function of viscosity η. Figure 5 also includes an arbitrarily selected lowest measurable $Q = 2$ along with values obtained from this work for S20 and those from Kandil et al.,[3] Lundstrom et al.,[8] and Sopkow et al.[9] that include measurements at $Q < 2$. The values of Q were estimated from a subsequent analysis with the function reported by eq 16. The measured Q are in reasonable agreement with those calculated from the theory because, as Table 1 shows, the measured resonance frequency is not a constant, it decreases with increasing viscosity. The method used to estimate Q form the measured $\boldsymbol{V}(f)$ is not intended for low Q resonances. Figure 5 also suggest that a wire of radius of 0.2 mm is required in order to obtain a Q of 2 for a vibrating wire viscometer immersed in a fluid of viscosity 1 Pa·s. Finally we note that if a stainless steel wire of density $\rho_s = 8000\ \mathrm{kg{\cdot}m^{-3}}$ were used instead of tungsten, this would decrease the upper operating viscosity as determined by $Q = 2$ by almost a factor of 2.

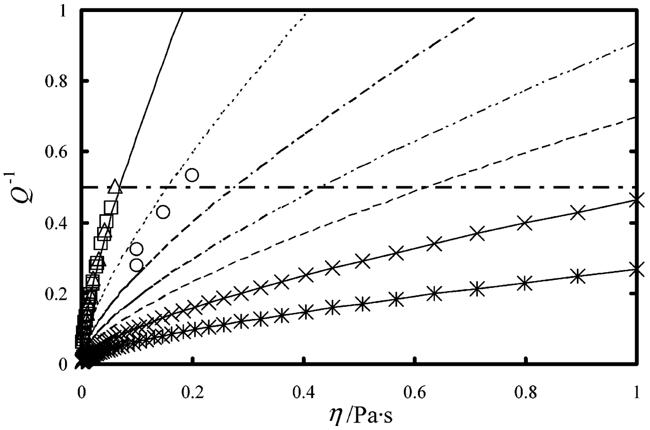

Figure 5. Resonance quality factor Q of a tensioned tungsten wire vibrating at a frequency $f_0 = 1.2$ kHz immersed in a fluid with $\rho = 850\ \mathrm{kg{\cdot}m^{-3}}$ as a function of viscosity η and wire radius R. —, $R = 0.05$ mm; - - -, $R = 0.075$ mm; — - —, $R = 0.1$ mm; — - - —, $R = 0.125$ mm; — —, $R = 0.15$ mm; —×—, $R = 0.2$ mm; —∗—, $R = 0.3$ mm; at $Q = 2$; ○, ref 3 with $R \approx 0.0747$ mm for certified reference fluid N100 at temperatures between (301 and 313) K and $p = 0.1$ MPa; △, ref 8 with $R \approx 0.0479$ mm for certified references fluids N10 at temperatures between (298 and 333) K and pressures in the range (0.1 to 55) MPa; □, ref 8 with $R \approx 0.0479$ mm for certified references fluids S20 at temperatures between (298 to 393) K.

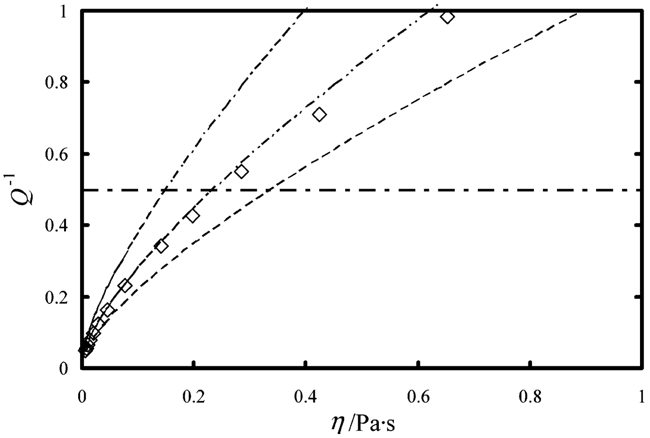

Figure 6. Resonance quality factor Q of a tensioned tungsten wire vibrating at a frequency $f_0 = 650$ kHz immersed in a fluid with $\rho = 850\ \mathrm{kg{\cdot}m^{-3}}$ as a function of viscosity η and wire radius R. — - —, $R = 0.1$ mm; — - - —, $R = 0.125$ mm; — —, $R = 0.15$ mm; - - -, at $Q = 2$; ◇, this work with $R \approx 0.125$ mm for certified reference fluid S60 at temperatures between (273 and 373) K and $p = 0.1$ MPa.

The measurements of viscosity reported in this article for certified reference fluid S60 were obtained with a tungsten wire $R \approx 0.125$ mm tensioned so that $f_0 = 650$ Hz. The analyses above were repeated with this f_0 for R of (0.1, 0.125, and 0.15) mm and the Q^{-1} as a function of η. The results (shown in Figure 6) demonstrate that decreasing f_0 by about factor of 2 decreases Q^{-1} by a relative factor of about 0.5 at a viscosity. Thus, decreasing f_0 decreases the available operating viscosity range of a vibrating wire viscometer. Again, the experimental and calculated Q are in reasonable agreement.

The sensitivity of a vibrating wire to viscosity is given by $Q{\cdot}\mathrm{d}Q^{-1}/\mathrm{d}\eta$, and as shown in Figures 5 and 6, this quantity decreases with increasing R. The resonance frequency is an insignificant factor in the sensitivity of a vibrating wire to the viscosity of the fluid in which it is immersed.

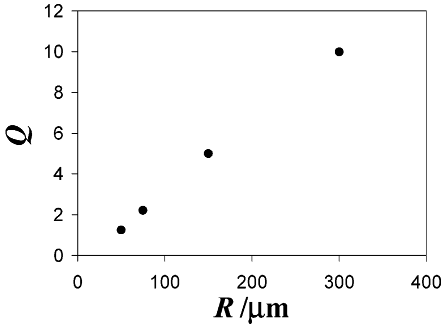

Figure 7. Resonance quality factor Q from Figure 6 for wires of several radii immersed in a fluid of viscosity 200 mPa•s. Note that the linearity of $Q(R)$ is consistent with the scaling law described in the text (eq 18).

In Figure 7, we show the linear relationship between the calculated values of Q as a function of wire radius R from the data presented in Figure 5 for a fluid of viscosity 200 mPa•s. This, perhaps surprising, linearity can be understood from some simple scaling law arguments presented here. For a driven harmonic oscillator experiencing dampening, the Q is given by[23]

$$Q = \frac{\sqrt{MK}}{b} \tag{18}$$

where M is the effective mass, K is the spring constant, and b is the frictional constant. The effective mass per unit length of the vibrating wire is proportional to the wire radius squared and is only weakly affected by the fluid in which it is immersed because the resonance frequency varies little as a function of fluid density or viscosity. After rewriting the right hand side of eq 18 as $\sqrt{(K/M)} \times M/b$, we note that the resonance frequency is approximately $\sqrt{(K/M)}$ and is held constant regardless of radius by varying wire tension. The drag b has been calculated by Stokes to be roughly proportional to the wire radius so that $Q \sim M/b \sim R^2/R \sim R$. This relationship can then be used to select the appropriate wire radius for a given viscosity range.

Measurements in Stagnant and Flowing S20. We have measured the viscosity of stagnant S20 at a temperature of 297 K and pressure of 20.68 MPa and obtained a viscosity of 0.055 Pa•s. This value can be compared with viscosity reported by Kandil et al.[24] for the same batch of S20 used for these measurements. To do so, the smoothing equation reported in ref 28 was used at a temperature about 3 K below the temperature of their lowest measurement. This extrapolated value of viscosity is 5 % below our result and within the combined expanded uncertainty of the measurements. We can also compare our measurement with the viscosity measurements reported by Lundstrom et al.,[8] albeit values determined for a different batch of S20 than that used here. Extrapolation of their best fit equation to a temperature also about 3 K below the lowest at which measurements were made gave a viscosity that is 9 %.

However, for our application, we are interested in the effect of fluid flow rate on the precision with which viscosity can be obtained from the vibrating wire. In the absence of measurements one might naively speculate the shear experienced by an axially oriented vibrating wire subjected to a flowing viscous fluid would introduce noise to the measured signal and also a systematic error to the measured viscosity. To addresses this speculation, we measured the $\boldsymbol{V}(f)$ of the fundamental mode of the wire immersed in S20 at flow rates of (22, 44, and 66) $\mathrm{cm^3{\cdot}s^{-1}}$. The $\boldsymbol{V}(f)$ obtained for stagnant and while flowing at a volumetric rate of 66 $\mathrm{cm^3{\cdot}s^{-1}}$ are shown in Figure 8; the tube has an inner diameter 0.0055 m, and at a volumetric flow rate of 66 $\mathrm{cm^3{\cdot}s^{-1}}$ has a velocity of 0.28 $\mathrm{m{\cdot}s^{-1}}$ producing a Reynold's number of about 200, suggesting that we are in the transitional regime between laminar and turbulent. Clearly, there is no significant additional noise arising from flowing fluid thus dispelling the first myth. However, as Figure 8 shows, the in-phase component is shifted to higher voltages in flowing fluid than experienced in stagnant. Fortunately, this background shift is accommodated by parameter a of eq 7 and is insignificant to the determination of η from $\boldsymbol{V}(f)$. The measurements were repeated with methylbenzene for which volumetric flow rate is turbulent and similar variations were observed. Because this shift in $\boldsymbol{V}(f)$ is observed solely in Re($\boldsymbol{V}$) (the in-phase component), one plausible explanation for its origin arises from a temperature change that affects the resistance of the tungsten wire. Such a temperature change was observed.

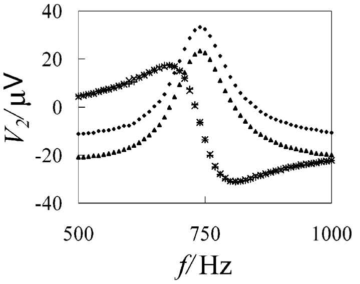

Figure 8. $\boldsymbol{V}(f)$ determined over the frequency f of the fundamental mode while the vibrating wire was immersed in stagnant S20 and S20 flowing with $\dot{n} = 66\ \mathrm{cm^3{\cdot}s^{-1}}$. ▲, in-phase {Re($\boldsymbol{V}(f)$)} voltage for $\dot{n} = 0$; ◆, in-phase {Re($\boldsymbol{V}(f)$)} voltage for $\dot{n} = 66\ \mathrm{cm^3{\cdot}s^{-1}}$; +, quadrature {Im($\boldsymbol{V}(f)$)} voltage for $\dot{n} = 0$; and ∗, quadrature {Im($\boldsymbol{V}(f)$)} voltage for $\dot{n} = 66\ \mathrm{cm^3{\cdot}s^{-1}}$. Note that the quadature data for the stagnant and flowing fluid lie on top of one another.

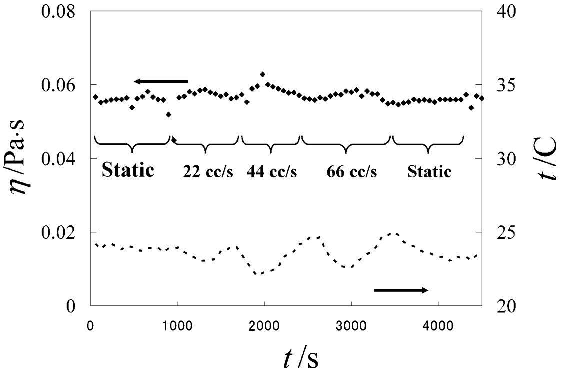

Figure 9. Viscosity η, shown on the left-hand ordinate, and temperature T, shown on the right-hand ordinate, determined while S20 was either stagnant or flowed over the vibrating wire at a flow rate $\dot{n}$ during a time t at a pressure of about 20.68 MPa.

The viscosity obtained from these measurements are shown in Figure 9 along with the observed temperature variations; the variations in viscosity at a flow rate vary with the changes in the measured temperature as anticipated from the $\eta(T, p)$ reported by Kandil et al.[24] The most striking feature of Figure 9 is the consistency of the measured viscosity over the whole range of flow rates investigated. The fluctuations in the measured viscosity are more likely a result of the variation in temperature, also shown in Figure 9, which was cyclic with a peak-to-peak variation of about 2 K. The temperature was measured on the outside of the tube and the viscometer was inside. The tube has a finite thermal conductivity and a small temperature gradient was imparted and it is not at all unreasonable for the measured viscosity to be out-of-phase with the temperature variations. The observed temperature fluctuations might have arisen from the energy imparted to the system by the mechanical pump used to flow the fluid through the vibrating wire and interconnecting tube. Based solely on these observations, we conclude that the flowing at rate less than 66 $\text{cm}^3 \cdot \text{s}^{-1}$ fluid with $\eta \approx 0.05$ Pa•s neither introduces significant noise to the measurement nor a systematic error in the measured η.

These results can be understood when the magnitude of the forces involved are estimated. If we assume the perturbation to flow from the thin wire is insignificant, then at a flow rate Q_f the pressure drop per unit length $\Delta p/L$ can be calculated for laminar flow in a circular pipe from

$$\frac{\Delta p}{L} = \frac{8\eta Q_f}{\pi R^4} \tag{19}$$

The stress on the wire will be largest at the highest flow rates of the most viscous fluid. In our case $Q_f = 66 \cdot 10^{-6}\ \text{m}^3 \cdot \text{s}^{-1}$, $R = 0.00275$ m, $\eta = 0.055$ Pa•s, and $\Delta p/L = 162{,}000\ \text{Pa} \cdot \text{m}^{-1}$, which we will use to approximate dp/dx in eq 20. The net force on the axially centered wire can be calculated from the stress τ as a function of radius r assuming laminar flow in a pipe-like annulus using[25]

$$\tau = \left(\frac{\mathrm{d}p}{\mathrm{d}x}\right)\frac{r}{2} + \left(\frac{\mathrm{d}p}{\mathrm{d}x}\right)\frac{{r_2}^2 - r^2}{4r \ln (r_2/r_1)} \tag{20}$$

In eq 20, r_1 and r_2 are the inner and outer radii, respectively, with values of $(75 \cdot 10^{-6}$ and $2.75 \cdot 10^{-3})$ m, respectively. To calculate the stress on the wire surface, we set r equal to r_1 resulting in $\tau = 1125$ Pa. The shear force on the wire parallel to the flow is then this shear stress multiplied by the surface area giving a value of about 10^{-2} N. However, this force is almost 3 orders of magnitude smaller than the tension on the wire (typically 10 N). The measured maximum amplitude of the resonance is about 50 μV. Based on this voltage, eq 4 can be used to estimate the maximum amplitude of the wire deflection at the wire center with the result of 1 μm. Decomposing the shear force to the transverse and axial components by a zeroth order approximation for a wire of length 50 mm the magnitude of the transverse force is about $5 \cdot 10^{-7}$ N for either the upstream or downstream half of the wire, each of which has been approximated as forming a triangle with respect to the pipe axis. However, the shear forces on the upstream and downstream portion of the wire tend to cancel each other out and further minimize the effect of flow. For the measurements reported in this article the wire was excited with a current of 25 mA within a magnetic field of about 0.4 T so that the Laplace force was about $5 \cdot 10^{-4}$ N, which is 3 orders of magnitude larger than the transverse-directed force produced by the shear on the wire, allowing the sensor to operate even under flow.

These calculations have assumed the wire is precisely aligned with the axis of the cylindrical tube in which it resides. To prove this we performed another series of experiments in which the wire was not axially symmetric with the cylinder by between (1 and 2)°. This off-axis location resulted in a signal-to-noise ratio lower than shown in Figure 8 even for a fluid with a viscosity of 1.2 mPa s at a flow rates of 44 $\text{cm}^3\ \text{s}^{-1}$ (data not shown). In this example, the uncertainty in the measured viscosity increased to 10 %. A reasonable estimate of the best alignment possible of the wire with the tube axis would be 0.25°. Assuming again a shear force of 10^{-2} N, the transverse force experienced by the misaligned wire would be about 10^{-4} N, which is the same order of magnitude as the Laplace force generated by the actuation current and magnetic field. Thus flow would be expected to degrade the signal-to-noise ratio. These back-of-the-envelope calculations imply the mechanical arrangement that holds the wire within the tube must do so with high axial symmetry. Our measurements were, in hind sight, obtained, perhaps fortuitously, with a wire aligned with the cylindrical axis of the tube.

Finally, the introduction of turbulence at much higher Reynolds numbers could add noise to the measurement as the forces experienced by the wire would not be symmetrically balanced and one could expect the nonuniformity to introduce perturbations to the motion of the wire, resulting in a spectrum with a higher noise level, lowering the precision of the measured viscosity.

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