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# Torsional Piezoelectric Crystal Viscometer for Compressed Gases and Liquids[1]

**D. E. Diller[2] and N. V. Frederick[2]**

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A torsional piezoelectric crystal viscometer for compressed gases and liquids at temperatures to 600 K and a pressures to 70 MPa has been developed. Several torsional crystals were prepared from swept (electrolyzed) quartz to obtain a good performance at high temperatures. Measurements of the bandwidth of the crystal resonance curve were automated using an impedance analyzer. The viscometer was tested on compressed gaseous argon and methane at temperatures to 500 K and at pressures to 50 MPa. The measurements differ from accurate wide-range correlating equations by less than 2%.

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**KEY WORDS:** argon; compressed gases and liquids; correlating equation; crystal resonance curve bandwidth; high pressures; high temperatures; methane; swept quartz; torsional crystal viscometer.

## 1. INTRODUCTION

Transport properties data and models are required for processing gases and liquids at high temperatures and pressures. There are few viscometers available for measurements at both high temperatures and high pressures. This report describes a new torsional crystal viscometer for both gases and liquids at temperatures up to 600 K and at pressures up to 70 MPa. The viscometer has been tested extensively on compressed gaseous argon and methane, and the data have been compared with accurate, wide-range correlating equations [1, 2].

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[2] Thermophysics Division, National Institute of Standards and Technology (formerly National Bureau of Standards), Boulder, Colorado 80303, U.S.A.

## 2. METHOD

The torsional piezoelectric crystal method is described in Refs. 3 and 4. We have computed all viscosities reported here using the data reduction equation published previously [4],

$$\eta = \frac{\pi f_{\text{fluid}}}{\rho} \left[\frac{M}{S}\right]^2 \left[\frac{\Delta f_{\text{fluid}}}{f_{\text{fluid}}} - \frac{\Delta f_{\text{vac}}}{f_{\text{vac}}}\right]^2 \tag{1}$$

where $\rho$ is the density of the fluid, $f$ is the resonant frequency, $M$ is the mass of the crystal, $S$ is the surface area of the crystal, and $\Delta f$ is the bandwidth of the crystal resonance curve. $\Delta f_{\text{fluid}}$ is measured with the quartz crystal surrounded by the test fluid, and $\Delta f_{\text{vac}}$ is measured with the cell evacuated with a typical oil diffusion pump.

The torsional crystal viscometer method has several advantages over other methods. It can be used at extreme temperatures and pressures, it does not require calibration with a fluid of known viscosity, it can be easily automated, and it permits measurements on fluids very close to equilibrium.

### 2.1. Apparatus

This apparatus is an adaptation of a low-temperature apparatus, described in Ref. 4, to higher temperatures.

#### 2.1.1. Transducer

A schematic drawing of the transducer (end view) is shown in Fig. 1. This transducer is very similar to the transducer described in Ref. 4. Only

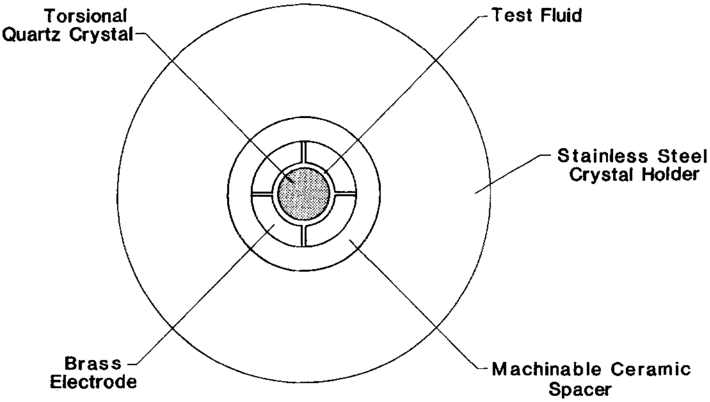

**Fig. 1.** Torsional crystal transducer (end view).

**Table I.** Quartz Crystal Parameters and Resonant Frequencies

| | Crystal D | Crystal 2 |
|---|---|---|
| Mass, g | 0.9662 | 0.8004 |
| Length, cm | 5.0076 | 4.2984 |
| Diameter, cm | 0.3046 | 0.2995 |
| Resonant frequency, Hz | 39,537. | 45,870 |
| Preparation | "Unswept" | "Swept" |

important differences between them are discussed here. Most of the differences involve materials which were selected for use at high temperatures.

*2.1.1.1. Quartz Crystals.* For measurements on gases, $Q_{\mathrm{vac}}^{-1} = \Delta f_{\mathrm{vac}}/f$ should be as small as possible. $Q_{\mathrm{vac}}^{-1}$ for quartz increases rapidly with temperature at temperatures above 450 K, making quartz unsuitable for use as a high-temperature viscometer. However, when natural quartz is swept or electrolyzed, $Q_{\mathrm{vac}}^{-1}$ is nearly independent of temperature to 846 K [5]. Two

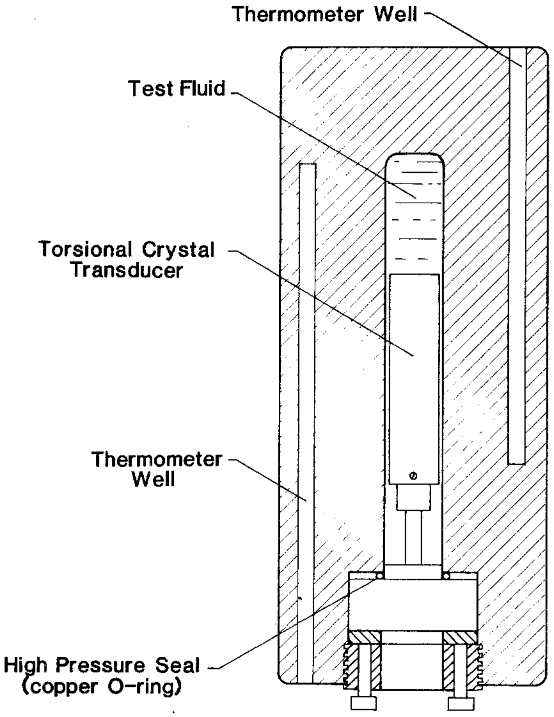

**Fig. 2.** High-pressure cell.

crystals, an unswept crystal and a swept crystal, were selected from crystals prepared for this work. Their masses, dimensions, and resonant frequencies are given in Table I. Their diameters are nearly the same and their lengths are significantly different.

*2.1.1.2. Crystal Holder.* The crystal is suspended in the crystal holder by a soft stainless-steel wire, held in tension by two tungsten springs (not shown). Machinable ceramic spacers electrically separate the brass electrodes from the stainless-steel crystal holder. The bolts which fasten two of the electrodes to the crystal holder are separated electrically from the holder by glass fabric tubes. The other details of the crystal holder are similar to Ref. 4.

### *2.1.2. Cell and Oven*

A schematic drawing of the cell is shown in Fig. 2. The transducer is contained in a cavity, 2.9 cm in diameter and 19.7 cm long, drilled in a 316 stainless-steel cylinder, 12.7 cm in diameter and 30.5 cm long. When assembled, the remaining cell volume contains less than 25 $cm^3$ of fluid. Two platinum resistance thermometers are placed in vertical holes, 1 cm in diameter and 20 cm long, drilled in opposite ends of the cell, and on opposite sides of the transducer cavity. Two 1.6-mm-o.d. electrical leads, which pass through the lid of the cell, are sealed by small cones of polypyromelitimide (PPMI) [6]. The lid is sealed with a silver-platted, solid copper O ring, compressed by 12 stainless-steel bolts. A 3.2-mm-o.d. stainless-steel tube is silver soldered into the lid to permit the test fluids to enter.

The oven was constructed from commercially available rectangular and cylindrical modules. Each module was vacuum molded from aluminum silicate fibers and has iron–chrome–aluminum alloy heater wires imbedded in one surface. There is a 1.3-cm air gap between the heater wires and the cell.

## 2.2. Procedures

### *2.2.1. Gas Purity*

The measurements were made on commercially available research-grade argon and methane. The 99.99% pure argon contained less than 10 ppm nitrogen, 1 ppm hydrogen, 5 ppm oxygen, 5 ppm hydrocarbons, and 3 ppm water. The methane was also specified 99.99% pure by the supplier, with nitrogen, carbon dioxide, and ethane the most likely impurities.

### *2.2.2. Temperature and Pressure*

The temperature was measured with a commercially available platinum resistance thermometer of secondary standard quality, calibrated at NBS, and placed in a vertical hole in the top of the cell. The long-term temperature drift of the cell thermometer was less than 0.2 K/day. For comparison, another platinum resistance thermometer was placed in a vertical hole in the bottom of the cell (see Fig. 2). The two thermometers usually differed by less than 0.2 K. The uncertainty in the cell temperature is estimated to be smaller than 0.2 K.

The temperature was controlled by a platinum wire sensor, connected to a proportional, off-line power controller. The sensor was placed in the air gap between the cylindrical heater and the cell. The long-term temperature drift of the cell temperature was less than 0.2 K/day.

The pressure was measured with a commercially available quartz transducer pressure gauge used at ambient temperature. Its calibration was checked by comparing it with an oil, deadweight piston gauge. The two gauges differed by less than 0.05 % at pressures larger than 1 MPa.

### *2.2.3. Density*

Fluid densities were obtained from temperature and pressure measurements and an equation of state [7]. The estimated errors in the densities are smaller than 0.2 %.

### *2.2.4. Viscosity*

An impedance analyzer was used for measuring the electrical conductance of the transducer as a function of frequency. Measurements of $\Delta f_{\mathrm{vac}}$, the bandwidth of the crystal resonance curve in vacuum, were carried out at the end of every run. Measurements on the transducer in vacuum were made at a voltage level of 0.01 V, to obtain conductances which were independent of voltage. The bandwidth in vacuum, $\Delta f_{\mathrm{vac}}$, was usually about 0.1 Hz. For this case, the frequency increment during a scan was 0.01 Hz. However, the value of $\Delta f_{\mathrm{vac}}$ could be read to about 0.002 Hz. For the transducer in vacuum, conductances at resonance were usually in the range 150–300 $\mu$S and could be read to 0.2 $\mu$S. Figure 3 shows our measured values of $Q_{\mathrm{air}}^{-1} = \Delta f/f$ for an unswept torsional crystal and a swept torsional crystal at temperatures to 600 K. $Q_{\mathrm{air}}^{-1}$ for the swept crystal is nearly independent of temperature in this range.

Measurements of $\Delta f_{\mathrm{fluid}}$, the resonance curve bandwidth for the transducer in a fluid, were made with a voltage drop of 0.1 V. $\Delta f_{\mathrm{fluid}}$ was usually between 1 and 15 Hz. The frequency increment during a scan ranged from

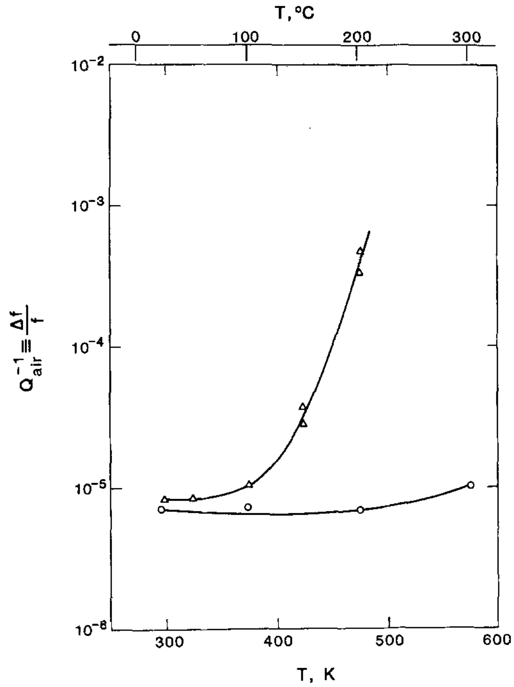

**Fig. 3.** $Q_{air}^{-1}$ of a torsional crystal as a function of temperature: (△) unswept crystal; (○) swept crystal.

0.05 Hz for the most dense gases to 0.02 Hz for low-density gases. Conductances at resonance were between 3 μS for the most dense gases and 25 μS for low-density gases. These conductances were read to the nearest 0.01 μS. Several hundred conductance–frequency pairs were measured during each scan of the resonance curve. The conductance of the cable between the analyzer and the transducer was nulled in the analyzer. The conductances at frequencies far from resonance (± 1 kHz) were measured after every scan and used to adjust the conductances measured at frequencies near resonance. All frequency scans were made at the slowest possible scan rates (about one step/s) to obtain conductances which were independent of the scan rate. Many comparisons were made between bandwidths measured with the impedance analyzer and bandwidths measured with a conductance bridge [4]. No significant differences were observed.

## 3. RESULTS AND DISCUSSION

Shear viscosity measurements for argon, at temperatures to 500 K, at pressures to 50 MPa, and at densities to 17 mol · $L^{-1}$, are presented in

**Table II.** Viscosities of Compressed Gaseous Argon

| $T$ (K) | $P$ (MPa) | $\rho$ (mol · L$^{-1}$) | $\eta$ ($\mu$Pa · s) | $100(\eta_{\text{meas}} - \eta_{\text{calc}})/\eta_{\text{calc}}$[a] | Crystal ID[b] |
|---|---|---|---|---|---|
| $T$ = ambient | | | | | |
| 292.90 | 50.362 | 17.675 | 49.3 | −0.4 | D |
| 295.90 | 41.619 | 15.694 | 44.1 | 0.2 | D |
| 293.50 | 39.124 | 15.200 | 42.9 | 0.5 | D |
| 295.00 | 35.040 | 13.899 | 40.0 | 0.8 | D |
| 293.50 | 27.906 | 11.724 | 35.7 | 1.4 | D |
| 295.90 | 27.878 | 11.581 | 35.6 | 1.1 | D |
| 295.90 | 20.890 | 8.933 | 31.3 | 1.3 | D |
| 293.50 | 20.739 | 8.977 | 31.1 | 1.1 | D |
| 293.80 | 13.892 | 6.037 | 27.3 | 0.8 | D |
| 295.90 | 13.882 | 5.973 | 27.0 | −0.6 | D |
| 293.50 | 9.539 | 4.111 | 25.2 | −0.2 | D |
| 296.00 | 9.444 | 4.024 | 25.3 | −0.1 | D |
| 297.40 | 6.989 | 2.934 | 24.0 | −1.9 | D |
| 293.50 | 6.925 | 2.953 | 24.3 | 0.1 | D |
| 296.00 | 6.911 | 2.917 | 24.5 | 0.6 | D |
| 293.80 | 5.170 | 2.183 | 23.9 | 0.6 | D |
| 293.50 | 3.485 | 1.460 | 23.1 | −0.6 | D |
| 296.00 | 3.311 | 1.373 | 23.5 | 0.9 | D |
| $T$ = 400 K | | | | | |
| 399.78 | 55.110 | 13.736 | 46.5 | 1.6 | D |
| 400.04 | 54.808 | 13.674 | 45.4 | −0.4 | D |
| 400.16 | 53.463 | 13.431 | 44.9 | −0.6 | 2 |
| 400.00 | 51.757 | 13.127 | 44.3 | −0.4 | D |
| 400.00 | 48.305 | 12.477 | 43.9 | 1.5 | D |
| 400.10 | 48.286 | 12.471 | 42.8 | −0.8 | 2 |
| 399.74 | 48.241 | 12.474 | 43.8 | 1.3 | D |
| 400.22 | 45.006 | 11.819 | 42.3 | 0.8 | 2 |
| 400.07 | 44.896 | 11.802 | 42.7 | 1.7 | 2 |
| 399.99 | 44.629 | 11.750 | 41.4 | −1.1 | D |
| 400.20 | 43.367 | 11.485 | 41.8 | 1.1 | 2 |
| 400.07 | 41.425 | 11.083 | 40.8 | 0.4 | 2 |
| 400.80 | 41.424 | 11.084 | 41.0 | 0.6 | D |
| 399.69 | 41.351 | 11.078 | 41.5 | 2.1 | D |
| 400.09 | 38.007 | 10.339 | 40.0 | 1.4 | 2 |

[a] $\eta_{\text{calc}}$ is from Ref. 1.

[b] D and 2 are crystal D and crystal 2, respectively, in Table I.

Table II. *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ (mol · L$^{-1}$) | $\eta$ ($\mu$Pa · s) | $100(\eta_{\text{meas}} - \eta_{\text{calc}})/\eta_{\text{calc}}$[a] | Crystal ID[b] |
|---|---|---|---|---|---|
| $T = 400$ K | | | | | |
| 399.99 | 37.982 | 10.336 | 38.9 | −1.4 | D |
| 400.14 | 37.961 | 10.327 | 39.2 | −0.4 | 2 |
| 400.17 | 34.607 | 9.563 | 39.0 | 2.0 | 2 |
| 399.67 | 34.518 | 9.555 | 38.9 | 1.8 | D |
| 400.14 | 34.479 | 9.534 | 38.3 | 0.5 | 2 |
| 399.97 | 34.436 | 9.528 | 38.3 | 0.4 | D |
| 400.07 | 31.057 | 8.719 | 37.5 | 1.3 | 2 |
| 400.11 | 31.022 | 8.710 | 36.9 | −0.3 | 2 |
| 399.76 | 27.602 | 7.866 | 35.7 | −0.2 | D |
| 400.20 | 27.600 | 7.856 | 35.8 | −0.1 | 2 |
| 399.65 | 27.549 | 7.855 | 36.4 | 1.7 | D |
| 399.65 | 25.854 | 7.420 | 35.9 | 1.9 | 2 |
| 400.09 | 24.227 | 6.985 | 35.0 | 0.8 | 2 |
| 400.13 | 24.154 | 6.965 | 34.7 | 0.1 | 2 |
| 399.65 | 22.520 | 6.512 | 34.7 | 1.6 | 2 |
| 400.13 | 20.716 | 6.041 | 33.5 | −0.5 | 2 |
| 400.21 | 20.684 | 6.031 | 33.9 | 0.9 | 2 |
| 399.91 | 20.681 | 6.035 | 33.3 | −0.9 | D |
| 399.66 | 20.621 | 6.023 | 34.0 | 1.3 | D |
| 399.58 | 18.965 | 5.568 | 33.4 | 1.1 | 2 |
| 400.13 | 17.287 | 5.090 | 33.3 | 2.0 | 2 |
| 400.14 | 17.259 | 5.082 | 33.0 | 1.2 | 2 |
| 399.55 | 15.529 | 4.599 | 32.4 | 1.0 | 2 |
| 399.93 | 13.811 | 4.101 | 31.8 | 0.3 | D |
| 400.10 | 13.755 | 4.083 | 32.0 | 0.9 | 2 |
| 399.71 | 13.745 | 4.085 | 31.8 | 0.5 | D |
| 399.54 | 12.056 | 3.596 | 31.6 | 1.3 | 2 |
| 400.13 | 10.375 | 3.097 | 31.3 | 1.4 | 2 |
| 400.09 | 10.331 | 3.085 | 30.8 | −0.1 | 2 |
| 399.94 | 6.912 | 2.073 | 29.9 | −0.5 | D |
| 400.23 | 6.882 | 2.063 | 29.9 | −0.5 | 2 |
| 399.71 | 6.806 | 2.043 | 20.2 | 0.7 | D |
| 399.95 | 3.429 | 1.031 | 29.7 | 1.0 | D |
| 400.16 | 3.425 | 1.029 | 29.4 | 0.0 | 2 |
| 400.29 | 3.423 | 1.028 | 29.5 | 0.3 | 2 |
| 399.97 | 3.295 | 0.990 | 29.7 | 0.9 | D |
| 400.17 | 1.371 | 0.412 | 28.8 | −1.2 | 2 |

Table II. *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ (mol $\cdot$ L$^{-1}$) | $\eta$ ($\mu$Pa $\cdot$ s) | $100(\eta_{\mathrm{meas}} - \eta_{\mathrm{calc}})/\eta_{\mathrm{calc}}$ [a] | Crystal ID[b] |
|---|---|---|---|---|---|
| | | $T = 500$ K | | | |
| 499.75 | 57.420 | 11.389 | 48.2 | 3.6 | 2 |
| 499.24 | 55.768 | 11.150 | 46.9 | 1.7 | 2 |
| 499.78 | 52.421 | 10.618 | 46.8 | 3.5 | 2 |
| 499.67 | 49.986 | 10.230 | 45.9 | 3.0 | 2 |
| 499.71 | 44.804 | 9.366 | 44.3 | 2.3 | 2 |
| 499.67 | 43.119 | 9.077 | 43.9 | 2.5 | 2 |
| 499.63 | 37.925 | 8.150 | 42.6 | 2.5 | 2 |
| 499.04 | 36.211 | 7.834 | 42.1 | 2.3 | 2 |
| 499.57 | 34.474 | 7.509 | 40.9 | 0.3 | 2 |
| 499.58 | 31.030 | 6.846 | 40.7 | 2.1 | 2 |
| 499.70 | 27.591 | 6.162 | 39.1 | −0.1 | 2 |
| 499.58 | 24.180 | 5.465 | 39.2 | 2.2 | 2 |
| 499.76 | 20.696 | 4.729 | 37.8 | 0.5 | 2 |
| 499.51 | 17.226 | 3.981 | 37.7 | 2.0 | 2 |
| 500.50 | 11.040 | 2.582 | 36.3 | 1.4 | 2 |
| 500.29 | 10.975 | 2.576 | 36.3 | 1.3 | 2 |
| 499.51 | 10.350 | 2.437 | 36.0 | 0.8 | 2 |
| 500.05 | 6.871 | 1.630 | 35.8 | 1.9 | 2 |
| 499.97 | 3.456 | 0.826 | 34.9 | 0.7 | 2 |
| 499.52 | 3.433 | 0.821 | 34.8 | 0.5 | 2 |
| 499.93 | 1.350 | 0.324 | 34.4 | 0.0 | 2 |

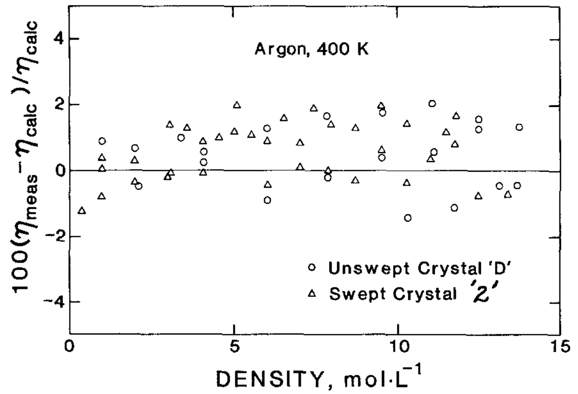

Fig. 4. Differences between measured and calculated viscosities [1] for argon at 400 K: (○) unswept crystal, D; (△) swept crystal, 2.

**Table III.** Viscosities of Compressed Gaseous Methane

| $T$ (K) | $P$ (MPa) | $\rho$ (mol $\cdot$ L$^{-1}$) | $\eta$ ($\mu$Pa $\cdot$ s) | $100(\eta_{\text{meas}} - \eta_{\text{calc}})/\eta_{\text{calc}}$ [a] | Crystal ID[b] |
|---|---|---|---|---|---|
| $T$ = ambient | | | | | |
| 294.50 | 54.817 | 17.834 | 37.4 | 3.8 | 2 |
| 297.40 | 54.717 | 17.674 | 36.2 | 1.8 | D |
| 297.40 | 48.478 | 16.870 | 34.0 | 2.1 | D |
| 294.50 | 41.935 | 16.034 | 31.5 | 1.8 | 2 |
| 295.20 | 41.930 | 15.989 | 31.3 | 1.6 | D |
| 297.40 | 41.587 | 15.808 | 30.7 | 0.7 | D |
| 295.20 | 34.703 | 14.614 | 27.8 | 0.8 | D |
| 294.70 | 34.285 | 14.553 | 27.0 | −1.7 | D |
| 294.50 | 28.137 | 13.004 | 24.4 | 0.6 | 2 |
| 295.20 | 28.117 | 12.951 | 24.2 | −0.0 | D |
| 294.70 | 27.719 | 12.867 | 23.6 | −2.0 | D |
| 295.20 | 20.841 | 10.372 | 10.1 | 0.5 | D |
| 294.70 | 20.797 | 10.386 | 20.0 | −0.1 | D |
| 294.70 | 13.937 | 7.001 | 15.9 | 0.3 | D |
| 295.20 | 13.891 | 6.953 | 16.0 | 0.6 | D |
| 298.00 | 6.934 | 3.149 | 12.9 | 0.4 | 2 |
| 295.20 | 6.920 | 3.188 | 12.5 | −1.9 | D |
| 294.70 | 6.867 | 3.169 | 12.7 | −0.3 | D |
| 295.20 | 6.843 | 3.148 | 12.8 | 0.9 | D |
| 298.00 | 3.460 | 1.484 | 12.0 | 1.2 | 2 |
| 295.20 | 3.384 | 1.466 | 11.6 | −1.7 | D |
| 294.50 | 3.350 | 1.455 | 11.6 | −1.1 | 2 |
| $T$ = 400 K | | | | | |
| 399.60 | 56.553 | 13.647 | 30.0 | 4.7 | D |
| 400.24 | 53.761 | 13.236 | 29.5 | 5.8 | 2 |
| 399.64 | 48.275 | 12.424 | 27.2 | 2.8 | D |
| 399.61 | 41.300 | 11.217 | 24.9 | 1.6 | D |
| 399.58 | 34.468 | 9.836 | 22.8 | 1.1 | D |
| 401.61 | 33.912 | 9.652 | 22.2 | −0.8 | 2 |
| 399.56 | 27.611 | 8.221 | 20.8 | 1.5 | D |
| 400.25 | 24.229 | 7.319 | 20.1 | 2.5 | 2 |
| 401.61 | 20.665 | 6.299 | 18.4 | −1.1 | 2 |
| 399.57 | 20.359 | 6.255 | 18.7 | 1.0 | D |
| 400.24 | 17.344 | 5.357 | 18.1 | 2.6 | 2 |
| 399.59 | 13.859 | 4.309 | 16.8 | 0.1 | D |
| 400.25 | 10.464 | 3.244 | 16.2 | 1.2 | 2 |
| 399.59 | 6.693 | 2.065 | 15.2 | −0.4 | D |
| 399.62 | 3.463 | 1.058 | 14.6 | −0.9 | D |

[a] $\eta_{\text{calc}}$ is from Ref. 2.

[b] D and 2 are crystal D and crystal 2, respectively, in Table I.

**Table III.** *(Continued)*

| $T$(K) | $P$(MPa) | $\rho$ (mol · L$^{-1}$) | $\eta$ ($\mu$Pa · s) | $100(\eta_{\text{meas}} - \eta_{\text{calc}})/\eta_{\text{calc}}$ [a] | Crystal ID[b] |
|---|---|---|---|---|---|
| | | $T = 500$ K | | | |
| 500.44 | 57.653 | 11.094 | 27.2 | 1.1 | 2 |
| 499.50 | 56.187 | 10.923 | 27.0 | 1.2 | 2 |
| 500.39 | 51.602 | 10.281 | 26.0 | 1.1 | 2 |
| 499.50 | 48.172 | 9.805 | 25.3 | 0.7 | 2 |
| 500.39 | 44.746 | 9.267 | 24.6 | 0.9 | 2 |
| 499.50 | 41.193 | 8.716 | 24.0 | 1.2 | 2 |
| 500.25 | 37.880 | 8.145 | 23.2 | 0.5 | 2 |
| 499.53 | 34.285 | 7.521 | 22.4 | 0.1 | 2 |
| 500.25 | 31.069 | 6.904 | 21.7 | −0.3 | 2 |
| 499.53 | 27.245 | 6.177 | 20.8 | −1.1 | 2 |
| 500.14 | 24.163 | 5.541 | 20.3 | −0.8 | 2 |
| 499.56 | 20.606 | 4.795 | 19.7 | −0.7 | 2 |
| 500.14 | 17.231 | 4.047 | 19.0 | −1.1 | 2 |
| 499.56 | 13.667 | 3.245 | 18.4 | −1.4 | 2 |
| 500.05 | 10.283 | 2.455 | 17.6 | −3.0 | 2 |
| 499.58 | 6.851 | 1.645 | 17.2 | −2.7 | 2 |
| 499.44 | 6.735 | 1.618 | 17.2 | −2.6 | 2 |

Table II. Viscosity measurements for methane in the same temperature, pressure, and density ranges are presented in Table III. All of the measurements at ambient temperature and some of the measurements at 400 K (labeled D) were made with the unswept crystal. The rest of the measurements at 400 K (labeled 2) and all measurements at 500 K were made with the swept crystal. The measurements, made with two crystals of significantly different lengths, are in good agreement. Tables II and III and Fig. 4 give comparisons between the data and accurate, wide-range correlating equations [1,2], developed previously at NBS. Most of the differences between measured and calculated viscosities are smaller than 2%. The differences, for argon at 400 K, are shown in Fig. 4.

## 4. ERRORS

Our error estimates are summarized in Table IV. (See also Ref. 4.) There is no published analysis of the errors in the derivation of Eq. (1), which is based on the simplest approximation to the theory of the instrument. The largest error in the crystal parameters is due mainly to the quartz cylinder being slightly out of round. The repeatability of $\Delta f_{\text{fluid}}$, and

**Table IV.** Error Estimates

| | |
|---|---|
| Crystal diameter | 0.2% |
| Fluid density | 0.2% |
| $\Delta f_{\text{fluid}}$ | 0.5% |
| $\Delta f_{\text{vac}}$ | 5% |
| Viscosity | 2% |

therefore the viscosity, ranges from 0.2 to 0.5%, depending on the density. The value of $\Delta f_{\text{vac}}$ is about 0.1 Hz at all temperatures and is very repeatable (0.005 Hz). The error in $\Delta f_{\text{fac}}$ is usually a small part of the error in $(\Delta f_{\text{fluid}} - \Delta f_{\text{vac}})$, which gives the viscosity. The error estimate for the viscosity is based on the preceding factors and on comparisons of our data for argon and methane with accurate, wide-range correlating equations. The differences are usually smaller than 2%.

Errors in the viscosities can also be caused by standing waves [3, 8, 9]. Anomalously large values for $\Delta f_{\text{fluid}}$, and hence the apparent viscosity, can occur when $\lambda_s/2 = W_s/f \cong d$, where $\lambda_s$ is the wavelength of a longitudinal wave in the fluid, $W_s$ is the speed of sound, and $d$ is a characteristic distance, probably a diameter, inside the crystal holder. For example, with crystal 2 in our transducer, we obtained standing waves for $\lambda_s/2 = 0.37$ cm and $W_s = 340\ \text{m} \cdot \text{s}^{-1}$, which occurred for argon at ambient temperature and at about $5\ \text{mol} \cdot \text{L}^{-1}$. Although we have been unable to eliminate standing waves, we have usually been able to avoid them by changing the length or the diameter of the crystal. Changing the length changes $f$ and therefore, $\lambda_s$. Changing the diameter changes the distance $d$.

## 5. SUMMARY

A new torsional piezoelectric crystal viscometer for use on compressed gases and liquids at temperatures to 600 K and at pressures to 70 MPa has been developed. The viscometer makes use of swept (electrolyzed) quartz crystals for good performance at high temperatures. The viscosity measurements have been automated by using an impedance analyzer with a microcomputer; however, complete runs cannot be made unattended. The apparatus has been tested on compressed gaseous argon and on methane at temperatures to 500 K and at pressures to 50 MPa with good results. The experimental error appears to be smaller than 2% in most cases. The apparatus is currently being used on carbon dioxide, ethane, and their mixtures. Further comparisons are being made between these pure fluid data and wide-range correlating equations.

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