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An Improved Greenspan Acoustic Viscometer

J. Wilhelm,[1,2] K. A. Gillis,[1] J. B. Mehl,[1] and M. R. Moldover[1,3]

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An improved Greenspan acoustic viscometer (double Helmholtz resonator) was used to measure the viscosity of gases at temperatures from 250 to 400 K and at pressures up to 3.4 MPa. The improvements include a vibration damping suspension and the relocation of the fill duct. The fill duct, which is needed to supply gas to the resonator, was connected to the center of the resonator to eliminate acoustic coupling between the resonator and the manifold. In anticipation of handling corrosive gases, all surfaces of the apparatus that are exposed to the test gas are made of metal. The viscometer was tested with argon, helium, xenon, nitrogen, and methane. Isothermal measurements were carried out at 298.15 and 348.15 K and at pressures up to 3.2 MPa. Without calibration, the results differed from published viscosity data by −0.8% to +0.3% (0.47% r.m.s.). These results are significantly better than previous results from Greenspan viscometers. The measurements also yielded the speed of sound, which differed from literature data by +0.16% to +0.20% (0.18% r.m.s.). Adding empirical effective-area and effective-volume corrections to the data analysis decreased the r.m.s. deviations to 0.12% for the viscosity and to 0.006% for the speed of sound. No unusual phenomena were encountered when the viscometer was tested with a helium-xenon mixture between 250 and 375 K.

KEY WORDS: acoustic resonator; argon; Greenspan viscometer; helium; helium-xenon mixture; methane; nitrogen; speed of sound; viscosity of gases; xenon.

1. INTRODUCTION

The frequencies and peak widths of gas-filled acoustic resonators are functions of the resonator shape and mode, and the thermophysical properties

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[1] Physical and Chemical Properties Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, U.S.A.

[2] Guest Scientist from Fachbereich Chemie, Universität Rostock, D-18051 Rostock, Germany.

[3] To whom correspondence should be addressed.

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of the gas. Resonators used for measuring the speed of sound, a quantity useful for determining the virial equation of state and the ideal-gas specific heat [1–4], are designed to have narrow resonance peaks so that the resonance frequencies can be determined precisely. In contrast, resonators used for measurements of viscosity or thermal conductivity are designed to have broader resonance peaks with widths strongly dependent on either the viscous diffusivity or the thermal diffusivity. Normally, measurements of peak widths are less precise than frequency measurements. The first attempt to measure viscosities using a double Helmholtz resonator was made in 1953 by Greenspan and Wimenitz [5], and it resulted in deviations from literature data of up to 38%. Today, an improved theory [6] and instrumentation yield significantly better results; in this work the measured viscosities deviate from literature data by less than 1%.

Here, we describe a Greenspan acoustic viscometer (double Helmholtz resonator) that was designed to work with the reactive gases used in semiconductor processing, i.e., all surfaces that are exposed to the test gas are made entirely of metal. Measurements on several gases were carried out to test the performance of the viscometer.

2. APPARATUS

2.1. Resonator

The resonator consists of two chambers connected with a duct (Fig. 1). An acoustic transducer (source) in one of the chambers generates an oscillating pressure that forces the gas in the resonator to oscillate through the duct between the chambers. A second transducer detects the pressure oscillations in the second chamber.

For design purposes, we used a simplified model [7] that is convenient for estimating the frequency response of the resonator. (A more accurate model [6] must be used to analyze the data.) In this simple model, the resonance frequency f_0 is given by

$$f_0^2 = \frac{c^2 r_\mathrm{d}^2}{2\pi L_\mathrm{d} V} \tag{1}$$

where c is the speed of sound, r_d is the duct radius, L_d is the duct length, and V is the chamber volume.

The acoustic velocity in the duct exceeds that in the chambers by a large factor that is on the order of (radius of the chamber/r_d)^2. The viscous dissipation rate per unit area is proportional to the square of the velocity.

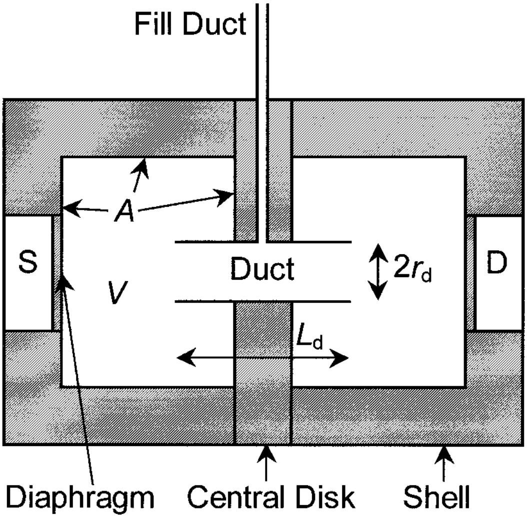

Fig. 1. Schematic cross-section of the Greenspan viscometer. The viscometer had an outer diameter of 6.2 cm and a length of 7.2 cm. The source and detector transducers are denoted by "S" and "D," respectively.

Thus, the main contribution to the dissipation of acoustic energy comes from viscous friction in the duct, within a viscous penetration length δ_v from the duct wall. The viscous penetration length is defined by

$$\delta_v = [\eta/(\pi f \rho)]^{1/2} \tag{2}$$

where η is the viscosity and ρ is the mass density. A second contribution to the dissipation comes from heat exchange between the gas and the walls of the chambers. When the gas is compressed and expanded in each chamber, the temperature of the gas also oscillates. The volume in which heat is exchanged between the gas and the chamber walls is $A\delta_t$, where A is the chamber surface area and δ_t is the thermal penetration length, defined by

$$\delta_t = [\lambda/(\pi f c_p \rho)]^{1/2} \tag{3}$$

Here, λ is the thermal conductivity and c_p is the isobaric heat capacity per unit mass. The surface area of the duct is much smaller than A, so thermal dissipation at the duct wall is neglected in this simple model. The diverging and converging flows at the ends of the duct make a third contribution to the dissipation. This contribution is measured by the orifice resistance parameter $\varepsilon_{\text{orif}}$, which is calculated numerically from the dimensions of the resonator [8, 9]. Certain gases (e.g., CH_4, CO_2) have symmetries such that many intermolecular collisions are required for their internal degrees of freedom to adjust to the temperature change associated with the acoustic oscillation. In such gases, the acoustic dissipation throughout the gas volume is characterized by the product $c_{\text{relax}}\tau_{\text{relax}}(\rho)$, where c_{relax} is the heat capacity of the slowly relaxing degrees of freedom and $\tau_{\text{relax}}(\rho)$ is the relaxation time that is assumed to be proportional to ρ^{-1}.

The quality factor Q is defined as the ratio of the resonance frequency f_0 to $2g$, the width of the resonance peak at $1/\sqrt{2}$ times its maximum height. The reciprocal quality factor Q^{-1} is the sum of the four contributions to the dissipation.

$$\frac{1}{Q}=\frac{\delta_{\text{v}}}{r_{\text{d}}}+2\varepsilon_{\text{orif}}\frac{\delta_{\text{v}}}{L_{\text{d}}}+(\gamma-1)\frac{\delta_{\text{t}}A}{\pi V}+(\gamma-1)\frac{c_{\text{relax}}}{c_p}\frac{2\pi f_0\tau_{\text{relax}}}{1+(2\pi f_0\tau_{\text{relax}})^2} \tag{4}$$

According to Eq. (4), the resonator properties r_{d}, L_{d}, V, A, and $\varepsilon_{\text{orif}}$ and the gas properties ρ, λ, c_p, γ, c_{relax}, and τ_{relax} are needed to determine η from measurements of the Q. (Here, $\gamma = c_p/c_V$ is the adiabatic exponent.) In order to minimize the propagation of the uncertainties of λ, c_p, γ, c_{relax}, and τ_{relax} into the uncertainty of the measured viscosity, it is desirable to make the viscous term in Eq. (4) much bigger than the thermal term. This requires a large chamber volume V and a small duct radius r_{d}. Furthermore, $L_{\text{d}}/r_{\text{d}}$ should be large to reduce the importance of the end effect parameter $\varepsilon_{\text{orif}}$. However, if these were the only considerations, one would design a resonator that is large, is difficult to handle, and has low resonance frequencies and small Q's. Such a resonator would provide weak, broad resonance signals with a low signal-to-noise ratio. The noise background is mainly generated by external vibrations and external electromagnetic fields.

Figure 1 is a sketch of the resonator used in this work. Its dimensions are $r_{\text{d}} = 0.23$ cm, $L_{\text{d}} = 3.1$ cm, $V = 29\ \text{cm}^3$, $A = 55\ \text{cm}^2$, and $\varepsilon_{\text{orif}} = 0.972$. The resonance frequencies ranged from 150 to 1100 Hz for the gases and temperatures reported here. For measurements with noble gases, Q^{-1} consists of approximately 80% viscous dissipation in the duct, 10% thermal dissipation, and 10% viscous disspation near the ends of the ducts. For polyatomic gases, the thermal dissipation is smaller, due to their smaller values of γ. The dissipation associated with c_{relax} does not exist for noble gases and is very small for most measurements on polyatomic gases.

All parts of the resonator and of the manifold that are exposed to the test gas are made of stainless steel. For the duct, a stainless-steel tube with an electropolished inner surface was used. To make the ends of the duct sharply right-angled, the tube was cut with an electron-discharge machine.

The piezoelectric-stack transducers (Kinetic Ceramics Inc.)[4] that are used as source and detector are separated from the test gas by 100 μm thick stainless-steel diaphragms. The main body of the resonator consists of two shells and one central disk. These parts were assembled using an indium wire gasket. The diaphragms are soldered into the shells, and the duct is soldered into the central disk. However, the indium as well as the lead-tin solder might be unsuitable for some corrosive gases, and they have uneven surfaces, which may be responsible for the systematic errors described in Section 3.1. In a future version of the resonator, the diaphragms will be machined into the shells, the duct will be welded to the central disk, and gold wire will be used instead of indium.

2.2. Fill Duct

A fill duct is used to move the test gas into and out of the resonator. The original version of the resonator had a fill duct leading into one of the chambers. With this design, the pressure oscillations in the chamber propagated into the fill duct. The theory of the Greenspan viscometer [6] includes a correction term for acoustic losses in the fill duct. However, for the calculation of this correction, the acoustic boundary condition at the end of the fill duct must be known. Figure 2 shows the results of viscosity measurements on argon, using a resonator that had a fill duct with a length of 30.5 cm and an inner diameter of 400 μm. Despite the high acoustic impedance of this narrow duct, the correction had a significant influence at high pressures, where the resonator had high Q's. Although the fill duct was open to a large manifold, the correction for an open (absorbing) termination did not lead to an agreement with the reference viscosity data [10]. Neither did the correction for a closed (reflecting) end. This means that sound was partially reflected at the junction of the fill duct and the manifold. Unfortunately, the manifold was too complex to calculate its acoustic impedance.

We attempted to make the fill duct termination either strongly absorbing or strongly reflecting. Good absorption was achieved with tissue paper or with foam rubber, but this conflicted with the requirement of an all-metal surface. Some valves and some porous steel plugs were tested as reflecting terminations. We were not successful in building or purchasing a valve that was compact, closed tightly, had all-metal surfaces, and could be

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[4] In order to describe materials and experimental procedures adequately, it is occasionally necessary to identify commercial products by manufacturer's name or label. In no instance does such identification imply endorsement by the National Institute of Standards and Technology, nor does it imply that the particular product or equipment is necessarily the best available for the purpose.

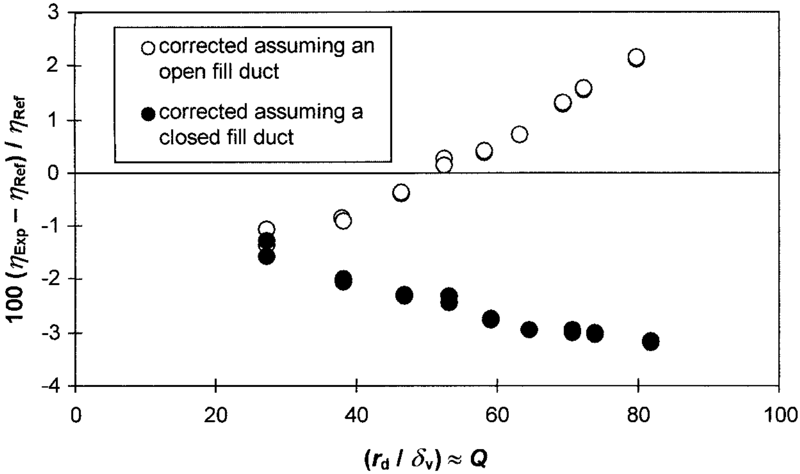

Fig. 2. Deviations of viscosities measured with the old Greenspan viscometer (fill duct leading into a chamber) on argon ($T = 298.15$ K, $p = 0.2$ to 1.8 MPa), assuming an open or a closed fill duct, from reference data [10].

remotely operated. The results with the steel plugs were also unsatisfactory. Although they had a typical pore diameter of 0.5 μm, their acoustic impedance still could not be considered infinite.

The best and easiest solution was to attach the fill duct to a small hole (160 μm in diameter) in the center of the main duct. At that location, the pressure oscillation has a node, so that there is no force driving sound into the fill duct. Thus, no correction is needed. The results of the measurements with this new resonator design are described in Section 3.

2.3. Pressurizing System and Thermostat

The isothermal measuring series with the Greenspan viscometer are completely automated. Starting at the maximum pressure, measurements are taken, and then the pressure is reduced in successive steps. Since acoustic measurements require very good temperature stability, the waiting period between measurements is at least 1 h to ensure equillibrium.

The manifold and all wetted parts of the valves are made of stainless steel. Bellows sealed valves (Nupro SS-4BW) with pneumatic actuation (Nupro 3C) are used. The compressed air needed to operate these valves is controlled with solenoid valves.

The resonator is suspended with three strings from the top of a pressure vessel. Additionally, three flexible copper braids lead from the vessel to the resonator to ensure good heat exchange. Because of the relatively low resonance frequencies, extra precautions were taken to isolate the resonator from vibrations, particularly from those of the stirred bath. Therefore, the fill duct and the electrical cables for the transducers and for the thermometer are very thin and flexible.

The pressure vessel is filled with a reference gas, usually argon. A pressure controller (Ruska 7000) adjusts the reference gas pressure to the pressure of the test gas, so that the diaphragms are protected from bursting. A differential pressure sensor separates the reference gas from the test gas. The estimated uncertainty of the pressure measurement is less than 0.2 kPa.

The apparatus is thermostatted in a silicone oil bath that is heated by means of a resistance heater and a 1 kW power supply. The power supply is controlled by a PID controller (Cryocon 34) which ensures a root-mean-square (r.m.s.) stability of the bath temperature of 3 mK. A platinum-resistance thermometer (Rosemount 162D), calibrated on ITS-90, is used to measure the temperature of the resonator. It is connected to a $7\frac{1}{2}$-digit multimeter (HP 3458A). The estimated uncertainty of the temperature measurement is 10 mK. Thus, the contribution of the uncertainties of pressure and temperature to the uncertainty of the measured viscosity is negligible.

The apparatus works at pressures up to 3.4 MPa and temperatures up to 400 K. These limits are set by the pressure controller and the piezoelectric transducers.

2.4. Measurement of the Frequency Response

To drive the acoustic oscillations, the source transducer is excited with voltages between 5 $\mathrm{V_{pp}}$ and 170 $\mathrm{V_{pp}}$ (volts peak-to-peak), generated with a synthesizer (HP 3325A) and a power amplifier. A high drive voltage can be applied at low pressures, but it must be reduced at high pressures in order to avoid turbulence in the gas flow. We found it convenient to apply a drive voltage that is proportional to $p^{-1.5}$ throughout a series of measurements. That way, the Reynolds number *Re* is kept nearly constant. The Reynolds number is defined here by

$$Re = \frac{u\rho r_\mathrm{d}}{\eta} = \frac{U_\mathrm{S} x_\mathrm{S} A_\mathrm{S}}{2V} \frac{\gamma_0 p Q r_\mathrm{d}}{\pi f_0 \eta L_\mathrm{d}} \tag{5}$$

where u is the maximum flow velocity in the duct, U_S is the drive voltage in $\mathrm{V_{pp}}$, $x_\mathrm{S} = 1.5\,\mu\mathrm{m}\cdot\mathrm{V}^{-1}$ is the elongation of the source transducer, $A_\mathrm{S} = 0.8\,\mathrm{cm}^2$ is the cross-section area of the source transducer, and γ_0 is the adiabatic exponent of the gas in the zero-pressure limit. A steady flow is laminar at *Re* up to 2300, but for oscillating flows, this limit can be much higher, depending on the ratio of the penetration length δ_v to the duct radius r_d [11]. To verify that turbulence effects are not significant, the Q of the resonator was measured as a function of the drive voltage. For all gases examined with the Greenspan viscometer, Q was independent of the amplitude for $Re < 50{,}000$.

The signal voltage on the detector side is between $10\,\mu V_{pp}$ and $40\,\mu V_{pp}$. Thus, care had to be taken to shield the detector signal from the electromagnetic fields of the surrounding power lines and the drive signal. A lock-in amplifier (Stanford Research Systems 850) is used to measure the detected signal with a reasonable signal-to-noise ratio. The signal voltage is divided by the drive voltage, which is measured with a second lock-in amplifier.

To measure the frequency response of the resonator, the drive frequency is scanned in the range $f_0 \pm 2g$ in steps of $g/5$. First, the range is scanned in ascending order and then, to verify the reproducibility, in descending order. After each frequency change, the lock-in amplifier needs a settling time that depends on the filter settings. With a time constant set to 0.3 s and a filter slope of 24 dB/oct, a delay of 4.5 s is required. Then the signal is measured and averaged for 10 s. A complete resonance scan takes about 10 min.

For simplicity, we have discussed the Greenspan viscometer in terms of two fitting parameters: f_0 and g. In practice, we used the detailed model [6] to analyze the measured frequency response of the resonator. This model also has two parameters that characterize the resonance, but it accounts for the departures of the frequency response from a simple Lorentzian function. Additional fitting parameters account for phase shifts in the transducers and for the background signal that is mainly caused by the electromagnetic field of the drive current. Typically, the model fits the complex amplitude as a function of frequency with fractional root mean square deviations between 10^{-4} and 10^{-3}.

3. MEASUREMENTS

In principle, the Greenspan viscometer is an absolute instrument, i.e., no calibration is necessary if the dimensions of the resonator are known with high accuracy. When the present instrument was used without calibration, the viscosity results for five gases at pressures up to 3.2 MPa deviated from the literature data by 0.47 % r.m.s. These results are significantly better than previous results from Greenspan viscometers [7], even though the present data span wider ranges of gases and measurement conditions. In order to attain better accuracy, we calibrated the present viscometer.

3.1. Calibration

Five gases with well-known viscosities were studied. The gases are listed in Table I, together with their purities, the pressure ranges and the temperatures at which measurements were made, and the literature references for all required properties. For helium, the best thermophysical data are the results of *ab initio* calculations [13], but we had to use experimental literature data for the relatively small density dependences of η [14] and λ [15]. The minimum pressure for the helium isotherms was 0.8 MPa; at lower pressures, the Q was less than 20 and the signal-to-background ratio became too small. The measurements on xenon did not extend above 1.1 MPa because the reference data for the viscosity of xenon [14] were stated to be unreliable at higher pressures. For the measurements on methane, a correction for relaxation effects was necessary. At the lowest pressures, this correction changed the results for the viscosity of methane by 0.2%. The heat capacity c_{relax} of the relaxing vibrational degrees of freedom was obtained by subtracting the translational and rotational parts from c_p. The values for τ_{relax} were taken from Ref. 1. For the measurements on nitrogen, the correction for relaxation effects turned out to be negligible.

As shown in Fig. 3, the viscosities that were measured with the uncalibrated Greenspan viscometer deviate from the reference values within the range −0.8% to +0.3%. Their r.m.s. deviation is 0.47%. We found that these differences could be reduced significantly by introducing a

Table I. Gases Used for Calibration

Gas	Purity stated by supplier	p (MPa)	T (K)	Reference η	Reference λ	Reference ρ, c, c_p, γ
Ar	99.9999% (Matheson)	0.2 to 3.2	298.15 348.15	[10]	[12]	[2]
He	99.9999% (Matheson)	0.8 to 3.2	298.15 348.15	[13] (η_0), [14] ($\Delta\eta$)	[13] (λ_0), [15] ($\Delta\lambda$)	[13]
CH_4	99.999% (Messer)	0.2 to 3.2	298.15 348.15	[16]	[17]	[1]
N_2	99.9999% (Messer)	0.2 to 3.2	298.15	[14]	[18]	[4]
Xe	99.995% (Matheson)	0.2 to 1.1	298.15	[14]	[15]	[3]

correction factor $(1+\alpha)$ into Eq. (4) that accounts for the uncertainty of the orifice resistance coefficient $\varepsilon_{\text{orif}}$. This is plausible because the ends of the duct might not have perfect right angles, as was assumed for the calculation of $\varepsilon_{\text{orif}}$ [8, 9]. However, adjusting $\varepsilon_{\text{orif}}$ corrects all the experimental viscosities by the same factor, and it does not affect the obvious Q-dependence of the deviations for argon and xenon. (See Fig. 3.) Because these gases have large values of γ, we considered thermal effects to explain the trends. We considered the possibility that the inner surfaces of the chambers had an effective area $(1+\beta)\,A$ that exceeded the area calculated from dimensional measurements. Irregularities, especially at the soldered seals and at metal o-ring seals might account for the factor $(1+\beta)$. Upon introducing the parameters α and β, the expression for Q^{-1} becomes

$$Q_{\text{corr}}^{-1}=\frac{\delta_{\text{v}}}{r_{\text{d}}}+2\varepsilon_{\text{orif}}(1+\alpha)\frac{\delta_{\text{v}}}{L_{\text{d}}}+(\gamma-1)\frac{(1+\beta)\,A\delta_{\text{t}}}{\pi V} \tag{6}$$

Relaxation effects are omitted here. The trends of the argon and xenon data imply that β increases as δ_{t} decreases with increasing pressure. Thus, we tested the assumption that $\beta(\delta_{\text{t}})=d_{\text{x}}/\delta_{\text{t}}$, where d_{x} is an empirical parameter with the dimensions of a length. We recognize that a correction term with such a simple form could not represent extremely precise data over a

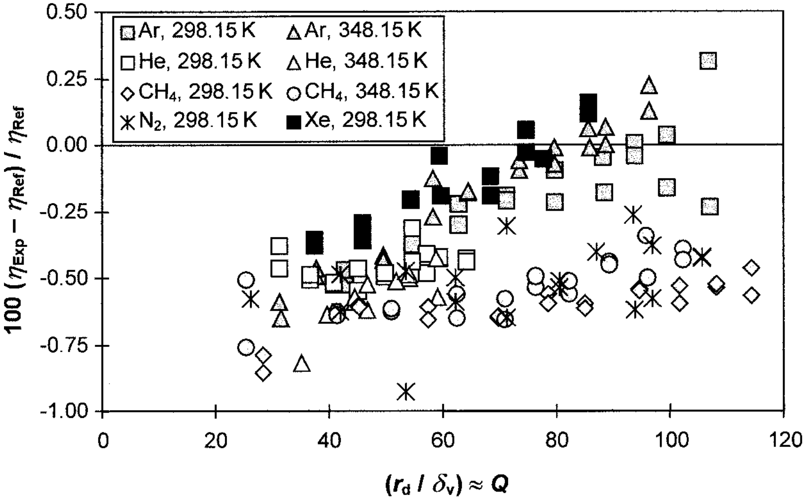

Fig. 3. Deviations of viscosities measured with the uncalibrated Greenspan viscometer from reference data for argon [10], helium [13, 14], methane [16], nitrogen [14], and xenon [14].

very wide range of δ_t. With this last assumption, we defined a viscosity correction function

$$\eta_{\text{corr}} = \eta \left[1 - \frac{4\varepsilon_{\text{orif}}\,\alpha}{L_d/r_d + 2\varepsilon_{\text{orif}}} - 2\,\frac{\gamma - 1}{\delta_v}\,\frac{A d_x}{\pi V(1/r_d + 2\varepsilon_{\text{orif}}/L_d)} \right] \tag{7}$$

A linear fit to all the deviations in Fig. 3 as functions of $(\gamma-1)/\delta_v$ returned the values $\alpha = -0.0343$ and $d_x = 1.19\,\mu\text{m}$. According to Eq. (27) of Ref. 9, this value of α would result if the right angles at the ends of the duct were rounded with a radius of only $1.09 \times 10^{-3}\ r_d = 2.5\,\mu\text{m}$. Such a small radius is consistent with the way in which the duct was manufactured. The value $d_x = 1.19\,\mu\text{m}$ implies that for the δ_t-range of the measurements reported here, the effective area is 1 to 5% larger than the geometric area. With these values of α and d_x, Eq. (7) can be used to correct all future measurements with this viscometer. The corrected viscosity data (Fig. 4) have an r.m.s deviation from literature data of 0.12%.

The second property deduced from the measurements is the speed of sound c. This enables us to check either the purity of the test gas or the composition of gas mixtures. For gases with an unknown equation of state, the density virial coefficients can be calculated from c. The speeds of sound for argon, helium, xenon, nitrogen, and methane that were measured with the uncalibrated resonator range from 0.16 to 0.20% (0.18% r.m.s.) higher

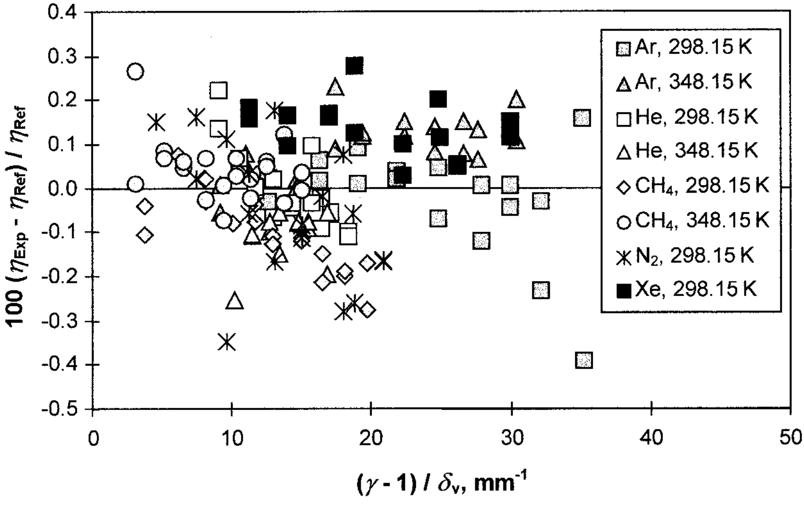

Fig. 4. Deviations of viscosities measured with the calibrated Greenspan viscometer from reference data for argon [10], helium [13, 14], methane [16], nitrogen [14], and xenon [14].

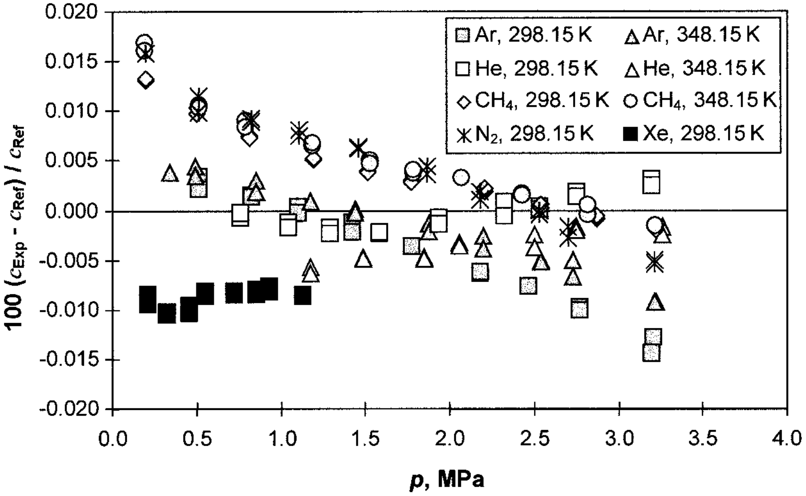

Fig. 5. Deviations of speeds of sound measured with the calibrated Greenspan viscometer from reference data for argon [2], helium [13], xenon [3], nitrogen [4], and methane [1].

than the literature data. Probably, the effective volumes of the chambers are smaller than those calculated from the dimensions because the volume occupied by the solder used to seal the diaphragms and the duct was not taken into account. After adjusting the value of V, all deviations of c from the literature data are within 0.02% (Fig. 5) and the r.m.s. deviation is 0.006%. This is a remarkable result for a low-Q resonator that was not primarily designed to measure the speed of sound.

3.2. Measurements on a Helium-Xenon Mixture

Helium-xenon mixtures are used as working fluids for thermoacoustic refrigerators because their Prandtl numbers Pr are very low. The Prandtl number is defined by

$$Pr = \eta c_p / \lambda \tag{8}$$

The NIST Fluid Science Group is measuring the thermophysical properties of these mixtures under the conditions of their potential applications. The results for the virial equation of state, determined with a Burnett apparatus and with a speed-of-sound system, have already been published [3]. As a part of this project, the viscosity of a mixture (supplier: Matheson) was measured with the Greenspan viscometer. The mole fraction of helium, x_{He}, was 0.47304. It was known with this high precision because we used

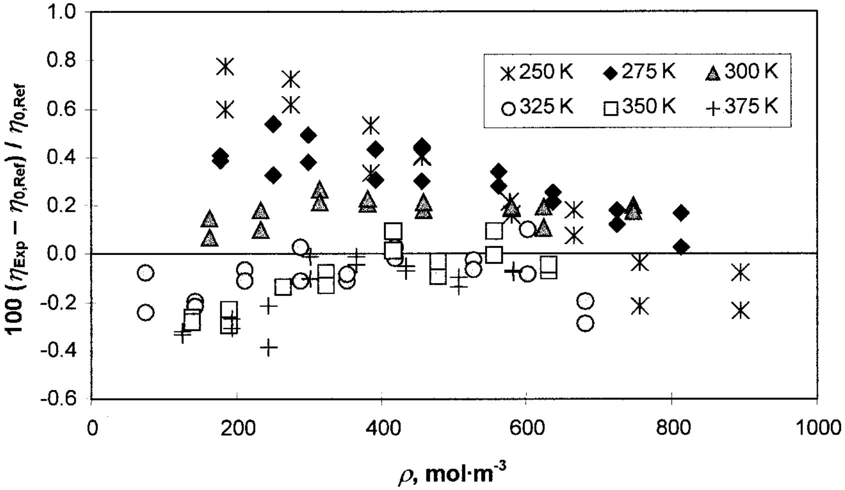

Fig. 6. Deviations of preliminary viscosities, measured with the calibrated Greenspan viscometer on a helium-xenon mixture ($x_{\mathrm{He}} = 0.47304$, $p = 0.2$ to 1.8 MPa, $T = 250$ to 375 K) and analyzed using Prandtl numbers of Kestin et al. [19], from the zero density values $\eta_0(T)$ of Kestin et al.

one of the samples from the Burnett/speed-of-sound work [3]. The data span the ranges 250 to 375 K and 0.2 to 1.8 MPa. Since the Prandtl number of this mixture is expected to be between 0.20 and 0.25 (instead of approximately 0.7 as for the measurements on pure gases described in Section 3.1), thermal dissipation plays a bigger role here (16% of Q^{-1} instead of 10%). Therefore, it is very important for the analysis of these measurements to know either the thermal conductivity or the Prandtl number with high precision. We are currently developing a resonator for measuring Prandtl numbers that will provide reliable data for Pr of helium-xenon mixtures. To obtain preliminary results for the viscosity, the measurements with the Greenspan viscometer were analyzed using zero-density values $Pr_0(T)$ calculated with a correlation of Kestin et al. [19]. Figure 6 shows the deviations of these preliminary viscosity data from the zero-density viscosities $\eta_0(T)$ of Kestin et al. [19]. Due to the uncertainty of the correlation, an agreement within 1% is reasonable. The trends of the deviation plots in Fig. 6 may represent either the density dependences of the viscosity or the Prandtl number or a combination of the two.

4. SUMMARY AND CONCLUSIONS

A double Helmholtz resonator (Greenspan viscometer) has been designed to be suitable for measurements on process gases used by the semiconductor industry. The most important requirement was to make all wetted surfaces metallic. Acoustic coupling between the resonator and the manifold was avoided by placing the fill duct in the center of the main duct where the Helmholtz mode has a node.

Measurements using argon, helium, xenon, nitrogen, and methane were carried out. The r.m.s. deviations of the viscosities measured in absolute mode from literature data were 0.47%, and they were reduced to 0.12% by introducing a calibration function. The speed-of-sound results obtained with the calibrated resonator deviated from literature data by less than ±0.02%, the r.m.s. deviation was 0.006%.

As part of a project to determine the thermophysical properties of working fluids for thermoacoustic refrigerators, the viscosity of a helium-xenon mixture was measured at temperatures from 250 to 375 K and pressures up to 1.8 MPa. The results agree with a correlation from the literature within ±1%.

In future work with Greenspan viscometers, we recommend that the ends of the duct be deliberately chamfered at an angle of 45° or rounded with a well-defined radius. Either change would permit the use of the theoretical value for $\varepsilon_{\mathrm{orif}}$; thereby eliminating the calibration parameter α. In order to eliminate the calibration parameter β, we recommend that the resonator be designed in such a way that the width of any crevice in its internal surface is much less than δ_{t}.

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