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A Vibrating-Wire Viscometer for Dilute and Dense Gases[1]

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The vibrating-wire technique has been applied to design a viscometer for precise measurements on gases in the temperature range 25 to 250°C at pressures from 0.1 to 40 MPa employing two Chromel wires with different radii. The technique has been improved to avoid the influence of higher harmonic modes and the degeneracy of perpendicular modes, to eliminate electromagnetic noise from the signal, and to minimize the influence of the magnetic damping. The decrement and frequency of the oscillation have to be determined by extrapolation to zero displacement, and wires with a perfectly smooth surface are needed to meet the requirements of the measuring theory. The viscosity measurements are characterized by a precision of $\pm 0.05\%$ at ambient temperature. Considering the uncertainty of the reference data used for calibration, the total uncertainty amounts to $\pm 0.2\%$ within the calibrated range of the boundary-layer thickness.

KEY WORDS: gas viscosity; high pressures; high temperatures; vibrating-wire viscometer.

1. INTRODUCTION

The presented instrument works in a similar way as vibrating-wire viscometers for measurements on liquids [1]. But the differences between the real instrument and the ideal model become more important and lead to non-negligible errors, because wires applied for a gas-phase viscometer have to be considerably thinner than those for measurements on liquids. The paper describes how to deal with the magnetic damping, the nonlinear

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inertial effects, and the influence of the wire surface. Special care has also been taken to suppress higher harmonic modes of oscillation, to avoid the degeneracy of perpendicular modes, and to eliminate electromagnetic noise from the signal.

2. THEORY OF THE METHOD

The theory of a vibrating-wire viscometer of Retsina et al. [2] can be used to determine the viscosity η of a fluid from the frequency ω and the logarithmic decrement Δ of a damped harmonic oscillation. However, the validity of the theory is restricted by conditions of the fluid mechanics, which have to be fulfilled.

First, the fluid has to be regarded as an incompressible continuum. Preliminary calculations showed that the Mach number, i.e., the relation between the flow velocity and the speed of sound in the fluid, and the Knudsen number, the ratio between the mean free path and the boundary-layer thickness, are very small and that compressibility and slip effects are negligible in the whole working range of the presented instrument. Furthermore, applying the working equations of Chen et al. [3], it has been proven that there is no significant influence of the outer boundary effect.

The theory is based on the Navier–Stokes equations, which are unsolvable in their complete form [Eq. (1)]. Hence, Retsina et al. [2] assumed that the maximum wire displacement y_{max} related to the wire radius R and the Reynolds number Re are very small according to Eq. (2), so that the convection terms $\rho(\mathbf{v} \cdot \nabla \mathbf{v})$ in the Navier–Stokes equations can be omitted. Provided that R is much smaller than the wire length L, there is no significant flow along the wire axis and the stream problem becomes planar.

$$\rho\left(\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v}\right) = -\nabla p + \eta \Delta \mathbf{v} \tag{1}$$

$$\varepsilon = \frac{y_{\mathrm{max}}}{R} \ll 1, \qquad \mathrm{Re} = \varepsilon \Omega \ll 1 \tag{2}$$

Besides ω and Δ, the fluid density ρ, the wire density ρ_s, and the decrement *in vacuo* Δ_0 are needed to calculate η in an iterative way using the working equations [Eqs. (3)–(6)]. Here, K_1 and K_0 are modified Bessel functions.

$$\Omega = \frac{\rho \omega R^2}{\eta} \tag{3}$$

$$A = (i - \Delta)\left[1 + \frac{2K_1(\sqrt{(i-\Delta)\,\Omega})}{\sqrt{(i-\Delta)\,\Omega}\; K_0(\sqrt{(i-\Delta)\,\Omega})}\right] \tag{4}$$

$$k = -1 + 2\mathrm{Im}(A), \qquad k' = 2\mathrm{Re}(A) + 2\Delta \mathrm{Im}(A) \tag{5}$$

$$\Delta = \frac{(\rho/\rho_s)\, k' + 2\Delta_0}{2[1 + (\rho/\rho_s)\, k]} \tag{6}$$

3. APPARATUS

3.1. Vibrating-Wire Viscometer

Measurements of high precision require that Δ is much greater than Δ_0, but not too large, so that a sufficiently large number of oscillation cycles can be analyzed:

$$0.005 < \Delta < 0.08 \tag{7}$$

Therefore, two wires with different radii are used to cover wide ranges of density and viscosity with only one instrument (Fig. 1). One wire, with

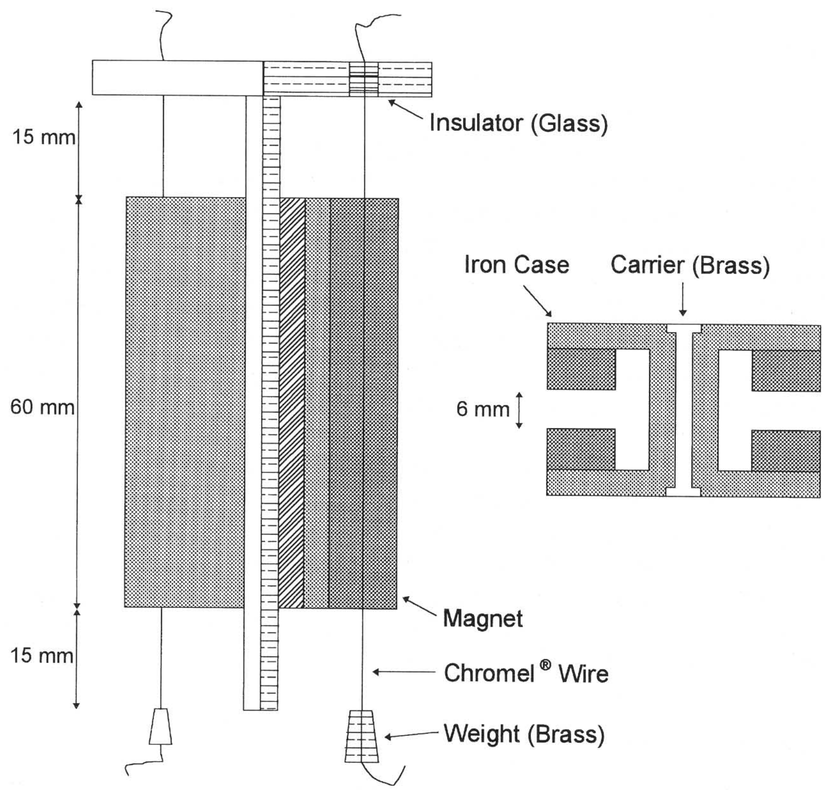

Fig. 1. Diagram of the viscometer cell.

a diameter of 25 μm, is assigned for measurements on gases at pressures of a few bars, whereas the second one, with a diameter of 50 μm, is suited for supercritical pressures. Simultaneous measurements with both wires in the intermediate pressure range allow verification of the operation of the instrument. Even the determination of η and ρ of gases, for which the equation of state is unknown, is possible in this way with a precision of about $\pm 1\%$.

The wires are clamped tightly between glass blocks at the upper ends and between the halves of brass stubbed cones at the lower ends. The gaps between the glass blocks and the brass halves must be perpendicular to the oscillation plane to avoid relative motion of the wires that would cause the coupling of two perpendicular modes of oscillation. Care has been taken not to add torsion to the wires due to the lower electrical connection. The masses m of the weights at the lower ends are 1.1 and 4.5 g, respectively, so that the angular frequeny of the oscillation *in vacuo* is 1.8 kHz for both wires:

$$\omega_0 \approx \frac{\pi}{L} \sqrt{\frac{mg}{\rho_s \pi R^2}} \tag{8}$$

For each wire the magnetic field with a vertical length L_{magn} of 60 mm is formed by four permanent magnets of $30 \times 10 \times 6$ mm made of Sm_2Co_{17} (IBS Magnet, Germany). The magnets are separated by 6 mm, so that the magnetic flux density B is 0.5 T. The optimum ratio between L and L_{magn} turned out to be 1.5 with regard to the suppression of the third harmonic mode of oscillation. If the arrangement is symmetric, the even harmonics are expected neither to be initiated nor to be observed. This is shown in Fig. 2 and has been proven experimentally by generating steady-state oscillations and measuring their amplitude as a function of the source frequency (Fig. 3).

The experimental equipment is intended for measurements at pressures up to 40 MPa and at temperatures up to 250°C. A pressure vessel from High Pressure Equipment Co. (USA) and valves with graphite packing (Autoclave Engineers, USA) are used. The viscometer cell is attached to the top closure of the vessel. The pressure is determined using four pressure transmitters (Paroscientific Inc., USA) characterized by an uncertainty of 0.01% of reading and 0.01% of full scale.

3.2. Electronic System

Initiation and detection of the wire motion are controlled by a PC equipped with a 12-bit multifunction I/O board (National Instruments

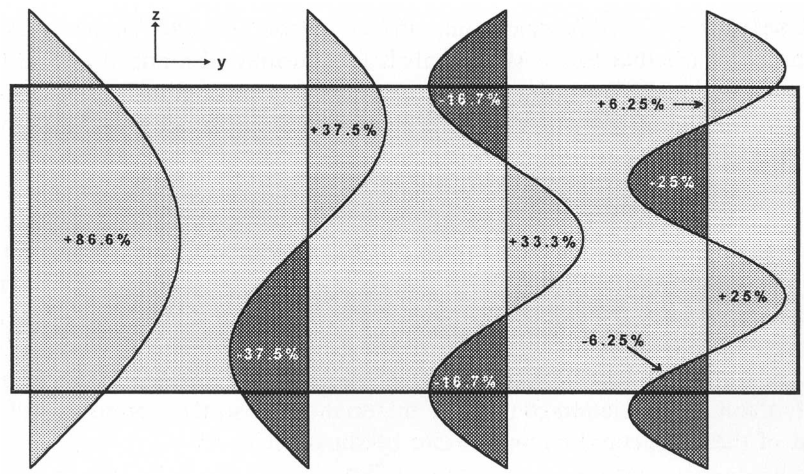

Fig. 2. First to fourth harmonic modes of the wire oscillation for $L = 1.5L_{magn}$.

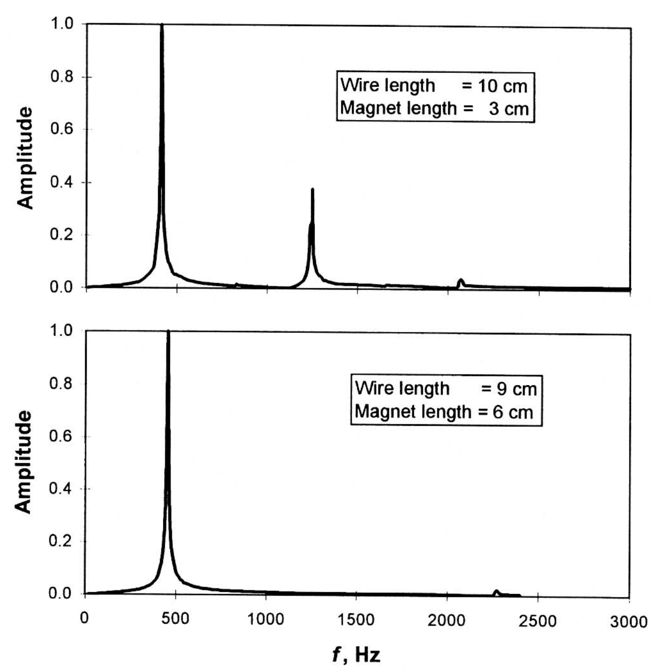

Fig. 3. Resonance behavior of vibrating wires.

AT-MIO-16E-10, USA) and compatible software (NI LabWindows/CVI). Since there are different DMA channels for output and input, it is possible to start the initiation pulse and data acquisition simultaneously with one trigger command. Although internal gains between 0.5 and 100 are available, an additional amplifier is necessary.

An important feature of the amplifier circuit is its electrical resistance R_Ω. According to the Lenzian rule a force F acts on a conductor in a magnetic field opposite to its velocity v. F is proportional to the current I. Thus, a magnetic damping constant D_{magn} [Eq. (9)] and a magnetic decrement Δ_{magn} [Eq. (10)] exist, corresponding to the parameters D_0 and Δ_0 expected *in vacuo*. As Δ_{magn} is independent of fluid presence, it appears as a constant. Consequently, the apparent vacuum decrement represents not only the internal friction of the wire material, but also the sum of this effect and of the magnetic damping. It can be shown that

$$D_{\mathrm{magn}} = -\frac{F}{vL_{\mathrm{magn}}} = \frac{B^2 L_{\mathrm{magn}}}{R_\Omega} \tag{9}$$

$$\Delta_{\mathrm{magn}} = \frac{D_{\mathrm{magn}}}{2\pi\rho_s R^2 \omega_0} = \frac{B^2 L_{\mathrm{magn}}}{2\pi\rho_s R^2 \omega_0 R_\Omega} \tag{10}$$

The existence of Δ_{magn} has been proven by measurements of the decrement in air under variation of the magnetic field using a tungsten wire of 10-μm diameter with $\omega = 2060$ Hz, $L_{\mathrm{magn}} = 52$ mm, and $R_\Omega = 460\Omega$ (Fig. 4).

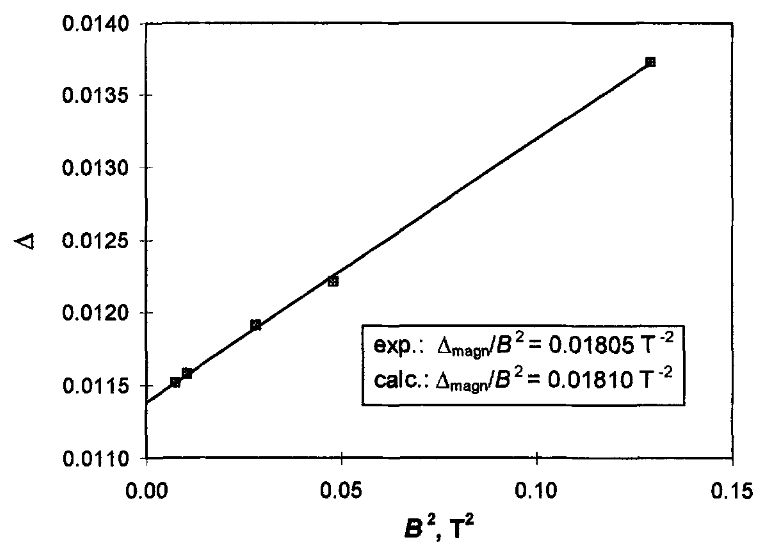

Fig. 4. Effect of the magnetic damping for measurements in atmospheric air.

In the applied amplifier circuits R_Ω had to be increased to 100 kΩ for the 25-μm wire and to 25 kΩ for the 50-μm wire to prevent the magnetic part of the damping from becoming too high. For both wires the calculated Δ_{magn} is 1.0×10^{-5}, whereas the measured Δ_0 is 2.5×10^{-5}, i.e., the internal friction of Chromel contributes an amount of 1.5×10^{-5}.

4. OPERATING PROCEDURES

4.1. Initiation of Oscillation, Data Acquisition, and Analysis

The oscillation is initiated by applying a sinusoidal voltage pulse with a frequency close to the resonance frequency of the wire. This is advantageous compared with the application of two consecutive opposite DC pulses [1], because the stimulation of the fifth harmonic mode of oscillation is avoided. Since a rectangular AC voltage can be expressed as the sum of uneven sine modes, the fifth mode is to be expected to occur after initiation with a rectangular pulse, which has been observed.

A single data acquisition with 2000 readings at a rate of 20 kHz takes just 100 ms. To reach a high accuracy it proved to be useful to carry out 100 acquisitions and to average the measured oscillation curves, so that asynchronous noise is minimized. The signal-to-noise ratio is improved in this way by a factor of 10. The delays between the initiations must be long enough for a complete decay. Thus, this technique cannot be recommended for vacuum measurements.

The parameters ω and Δ are determined by means of a nonlinear fit on the basis of the Newton–Raphson procedure using Eq. (11), where U is the measured voltage.

$$U = U_{max} \sin(\omega t + \varphi)\, e^{-\Delta \omega t} \tag{11}$$

A typical oscillation and its deviation from the fitted curve are presented in Fig. 5. The first cycle of the voltage curve, which originates from the initiation pulse, is excluded from the analysis. The diagrams are always shown on the PC screen during the measurement and fitting procedure so that disturbances can be recognized immediately.

4.2. Extrapolation to Zero Displacement

As stated in Section 2, a low dimensionless amplitude ε is an essential condition for the validity of the measuring theory. This is difficult to fulfill in practice, even if the wire is relatively thick. However, if a very thin wire, as is employed in a gas viscometer, is displaced by only 1% of its radius, the signal voltage will almost disappear behind the electromagnetic noise.

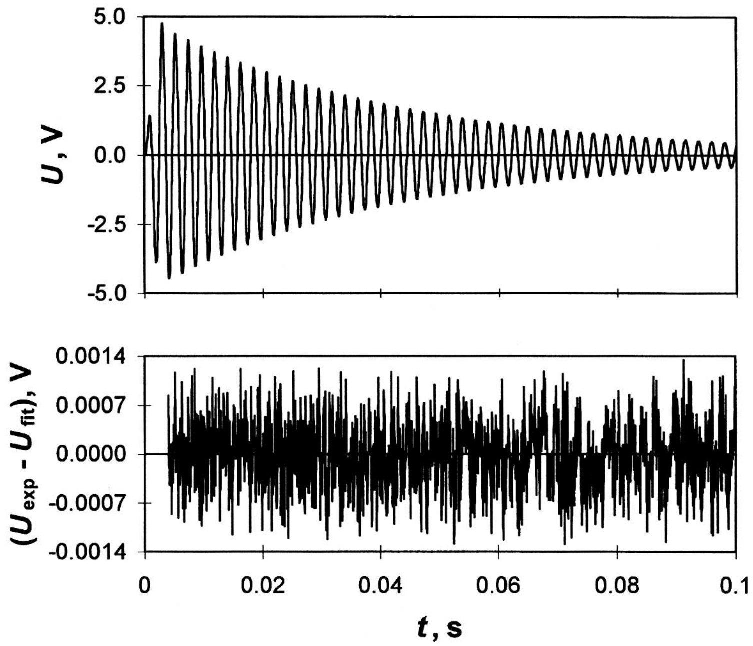

Fig. 5. Oscillation curve and deviation plot.

Since appropriately undisturbed measurements require greater displacements, it turned out to be necessary to determine ω and Δ as functions of ε^2 and to extrapolate to vanishing displacement.

The quadratic relationship can be explained with regard to the behavior of the dimensionless stream function Φ. Retsina et al. [2] expanded Φ as a Taylor series [Eq. (12)] and proved that, due to symmetry, the linear term $\varepsilon\Phi^{[1]}$ makes no contribution to the motion.

$$\Phi = \Phi^{[0]} + \varepsilon\Phi^{[1]} + \varepsilon^2\Phi^{[2]} + \cdots \tag{12}$$

As a consequence of the density dependence of the Reynolds number Re, the slope of the extrapolation line is almost negligible at low densities but increases considerably at higher densities (Fig. 6). The decrements determined in this way have an uncertainty of less than $\pm 0.05\%$. The uncertainty of the frequencies is even smaller: $\pm 0.005\%$.

One viscosity determination including data acquisition and analysis of oscillations, which are initiated with different voltages, takes about 15 min. During this time the pressure, temperature, and gain of the amplifier are kept constant. A time-saving procedure is to measure a curve of 5000 data

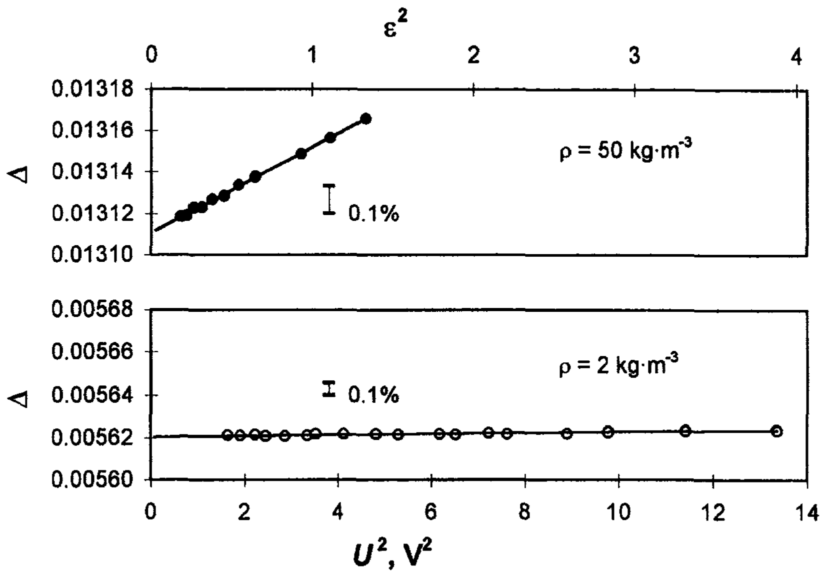

Fig. 6. Extrapolation to zero displacement for decrements measured on argon.

points and to analyze equally long segments of it. Both procedures should provide the same results; otherwise, there is an effect of mode coupling, and the wire suspension is consequently to be checked.

4.3. Calibration

Since the wire quantities R and ρ_s are not sufficiently precise as given by the supplier (Goodfellow Ltd., UK), calibration is required. A closer view of the algorithm for the viscosity calculation makes clear that only the term $\rho_s R^2$ is of importance. Thus, the tabulated value of ρ_s can be used and only R has to be calibrated. Measurements on argon, nitrogen, and helium have been carried out at room temperature with reference to reliable viscosity values provided by Kestin and Leidenfrost [4].

If the outlined theoretical model of the experiment is consistent with the experimental performance, then a single measurement on a single gas with known properties at one thermodynamic state suffices to determine R. The same value of R should be obtained for any gas under any conditions. The results of such measurements on argon for different wire materials are presented in Fig. 7. It shows a plot of the apparent wire radius, reduced by an arbitrary nominal radius, as a function of the boundary-layer thickness δ:

$$\delta = (\eta / \rho \omega_0)^{1/2} \tag{13}$$

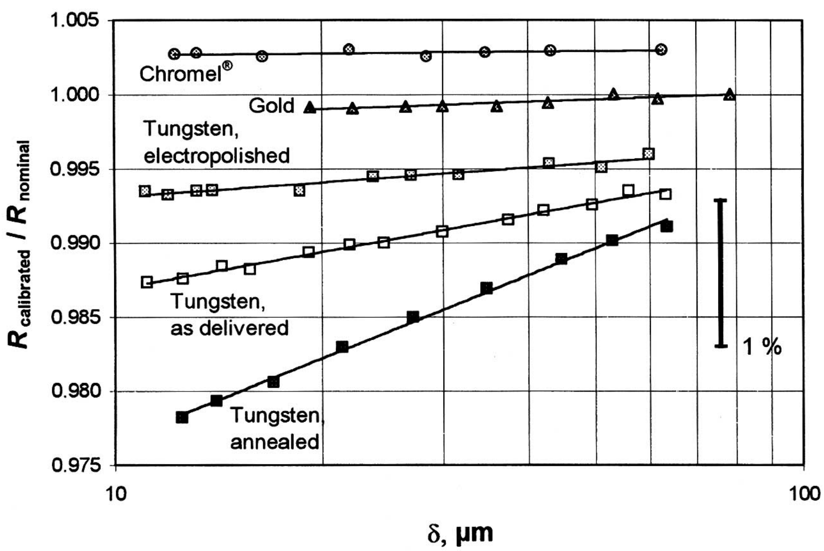

Fig. 7. Wire radius, calibrated with argon, as a function of the boundary-layer thickness.

It is clear that the effective radius depends on δ and on the wire material as well as its surface treatment. The reason for this behavior is that real wires differ from a perfectly circular cross section assumed in the theory. An identical functional dependence of R on the boundary-layer thickness has resulted from measurements on nitrogen and helium.

It emerges from examining the wires by means of an electron microscope that the rough surface of tungsten becomes distinctly rougher when annealed in air, whereas electrolytic polishing in diluted sodium hydroxide leads to a smoother surface. Relatively good results could be obtained with a gold wire, but it is difficult to use in practice due to its low tensile strength. Vacuum-annealed Chromel thermocouple wire has been identified to have the smoothest surface of all tested materials, as illustrated by the very flat curve shown in Fig. 7. Despite its relatively low density of $8500\ \mathrm{kg \cdot m^{-3}}$ and its high thermal expansion coefficient of $17.2 \times 10^{-6}\ \mathrm{K^{-1}}$, Chromel has been employed as the wire material for this instrument.

5. CONCLUSIONS

A vibrating-wire viscometer has been constructed to determine the viscosity of gases at temperatures up to 250°C and pressures up to 40 MPa. The calibrated instrument can be operated with an accuracy better than $\pm 0.2\%$. It has been proven that the measured oscillation quantities have to be extrapolated to zero displacement and that wires with a perfectly smooth surface are to be used to meet the theoretical requirements.

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