



# Measurements of the viscosities of Kr and Xe by the two-capillary viscometry



H. Lin<sup>a</sup>, J. Che<sup>b</sup>, J.T. Zhang<sup>a,\*</sup>, X.J. Feng<sup>a</sup>

<sup>a</sup> National Institute of Metrology, Beijing 100029, China

<sup>b</sup> College of Information, Shanxi Agriculture University, Taiyu 030801, China

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## ABSTRACT

Coiled two-capillary viscometers can measure gas viscosities with high accuracy over a wide temperature range. The authors built a two-capillary viscometer and used it to measure the zero-density viscosity of Kr and Xe at temperatures from 243.15 K to 393.15 K with relative standard uncertainties of 0.10% and 0.11%, respectively. Additional measurements on Ar were used to understand and avoid the error due to inadequate preheating or precooling of the gas entering the viscometer. The error, which occurred at volume flow rates above  $4 \text{ m}^3 \text{ s}^{-1}$ , was approximately proportional to the temperature difference between the reference capillary and the probe capillary.

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## 1. Introduction

Capillary viscometers are simple to construct and applicable for a wide temperature range. The devices can be operated in either the absolute or the ratio mode. The devices operate according to the principle of Hagen – Poiseuille flow. Berg [1,2] studied the corrections for viscosity measurements with capillary viscometers of compressible dilute gases. Previous studies [2–7] have verified the high performance of single and two-capillary viscometers in wide temperature ranges. Berg stated that the length of a coiled capillary, which is required for the single – capillary viscometer operating in the absolute mode, is surprisingly difficult to measure accurately. To avoid the difficulty, we used the suggestion in literature [8] of using a single-capillary viscometer to measure the viscosity ratios of gases by the aid of flow meters of low nonlinearity and high reproducibility. For such an operation, the flow meter's uncertainty for absolute measurements is less important. An alternative method, two-capillary viscometry, was conceived according to the principle of coiled single-capillary viscometry [4,5] to exploit the advanced *ab initio* calculations of the viscosities of helium [9] and the accurate viscosity ratios of the gases against helium (He) at 298.15 K [10]. Two-capillary viscometers measure the viscosity of a

gas by comparing it with the known viscosity of He at the same isobar and isotherm. The uncertainty of such a ratio measurement is dependent on the reproducibility of the two-capillary viscometer. Literature [9] reports the recent achievement of improved *ab initio* calculations for thermophysical properties of He, such that the viscosities of He are calculated from 1 K to 10 000 K with the unprecedented relative standard uncertainty from 0.002% above 50 K and 0.05% at lower temperatures. The new calculations agree with the previous calculations [11,12] within 0.01% above 20 K and 0.1% at 1 K. In addition, Berg and Moldover [10] reviewed measurements for the zero-density viscosities contributed by 18 instruments at 298.15 K for eleven gases: He, Ne, Ar, Kr, Xe, H<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub> and SF<sub>6</sub>. The viscosity ratios between the gases and He are determined with the relative standard uncertainties of  $(2.7\text{--}3.6) \times 10^{-4}$ .

By referring to literature [4,5], we installed a two-capillary viscometer for measurements of the viscosities of gases in a wide temperature range. After we investigated some systematic errors with the two-capillary viscometer [6,7] we reached the relative standard uncertainty of 0.062% for measurements of the viscosities of dilute Ar in a wide temperature range. The measurements agree well with the data given by May et al. [4,5], Vogel [13,14], Wilhem and Vogel [15], and the *ab initio* calculations [16,17] in the overlapping temperature ranges. The fractional differences are within 0.08%.

Accurate measurements for the zero-density viscosities of Xe and Kr are rarer than those for Ar. Among those rare researches,

\* Corresponding author.

E-mail address: [zhangjint@nim.ac.cn](mailto:zhangjint@nim.ac.cn) (J.T. Zhang).

many had the instruments calibrated by the values obtained from Kestin's group, usually the 1959 paper by Kestin and Leidenfrost [18], where the adjustment of the instrument's calibration constant was based on Bearden's measurement of dry air made in 1939 [19]. Thus, accurate absolute measurements of gas viscosity are even more rare because, before 2006, the calibrations of viscometers, including those with oscillating disks and vibrating wires as well as capillaries, often traced to measurements by Kestin and Leidenfrost [18], which in turn traced to Bearden's 1939 measurement of dry air [19].

Bich et al. [20] published in 1990 a theory – based correlation of gas viscosities. They calculated the reduced collision integral from the interatomic potentials that were adjusted to experimental data, including viscosity. Although the correlation derived indirectly from viscosity measurements, it had the advantage of covering temperatures from the normal boiling point to 5000 K. Vogel [13] published the viscosities of neon, argon krypton and xenon in temperatures from 293 K to 668 K measured with the oscillating-disk viscometer. Wilhelm and Vogel [15] published the measurements for argon, krypton and propane at 298 K and 348 K by a vibrating – wire viscometer, whose calibration relied on data from Kestin and Leidenfrost [18]. May et al. [9] reported the measurements for Xe in temperatures from 202.88 K to 298.15 K by the two-capillary viscometer. They obtained the viscosity of He at 298.15 K by averaging the *ab initio* value with the value obtained from a primary single-capillary viscometer with a known radius and length. Thus, their values are independent from those of existing measurements.

In this paper, we report the measurements for the zero-density viscosities of Xe and Kr with a two-capillary viscometer. The temperatures ranged from 243.15 K to 393.15 K for Kr and from 298.15 K

$$(\dot{n})_T = \left( \frac{\pi r_d^4}{16L} \right)_T \frac{(p_1^2 - p_2^2)}{\eta_{0,T}^{\text{gas}} RT} C^{\text{gas}}(T, p_1, p_2), \quad (1)$$

where  $L$  is the length of the capillary;  $p_1$  and  $p_2$  label respectively the pressure at the entrance and the exit of the capillary;  $R$  is the universal gas constant;  $r_d$  stands for the inner radius of the capillary;  $T$  denotes the gas temperature;  $\eta_{0,T}^{\text{gas}}$  is the zero-density viscosity of the gas in which the subscript 0 denotes zero pressure. The coefficient  $C^{\text{gas}}(T, p_1, p_2)$  given in Ref. [2] contains the corrections: 1) the density dependence of viscosity and deviations of the gas compressibility from the ideal gas law; 2) the slip effect of the gas at the capillary's walls; 3) the entrance effect of viscous gas flow; 4) the gas expansion along the capillary; 5) the temperature nonuniformity. In addition,  $C^{\text{gas}}(T, p_1, p_2)$  also contains the function accounting for the centrifugal effect with the coiled capillaries. We calculated  $C^{\text{gas}}(T, p_1, p_2)$  according to the functions given in Ref. [2]. The correction coefficients in the hydrodynamic model quoted those used by May et al. [4,5]:  $K_{\text{slip}} = 1.00$  and  $K_{\text{ent}} = 0$ . The entrance coefficient  $K_{\text{ent}}$  was set to zero because, at the location of the tap for  $p_1$ , the diameter of the capillary matched the diameter of the preceding tubing. The values of  $K_{\text{exp}}$  and  $K_{\text{therm}}$  cited those defined by Berg [2]. The second viscosity virial coefficient of Xe was determined by the method indicated in Ref. [5].

May et al. [4] devised the two-capillary viscometer that comprises two coiled capillaries in series, in which the upstream capillary is maintained at the reference temperature,  $T_{\text{ref}} = 298.15$  K and the downstream capillary constituting the probe unit is at the variable temperature  $T$ . Equating the molar flow rates in both capillaries yields

$$\frac{\eta_{0,T}^{\text{He}}}{\eta_{0,298.15}^{\text{He}}} = \frac{(p_3^2 - p_4^2)^{\text{He}}}{(p_1^2 - p_2^2)^{\text{He}}} \frac{C^{\text{He}}(T, p_3, p_4)}{C^{\text{He}}(298.15, p_1, p_2)} \left( \frac{\pi r_d^4}{16L} \right)_T \bigg/ \left( \frac{\pi r_d^4}{16L} \right)_{298.15} \frac{298.15}{T}. \quad (2)$$

to 393.15 K for Xe, respectively. We compared the present measurements with the published results, and investigated the uncertainty contribution arising from the extrapolation of zero volume flow rates for the measurements of Kr and Xe.

## 2. Principle

The principles for single- and two-capillary viscometry have been detailed in literature [2,4]. In this section, we simply review the key principles for two-capillary viscometry. Capillary viscometers for gases use the Hagen–Poiseuille equation modified for compressible fluids. For compressible gases in coiled capillary flow, the hydrodynamic model is approximated through the corrections discussed in Ref. [2]. Then, the molar flow rate  $\dot{n}$  is:

In the same way, substituting a sample gas for the helium yields the same form of viscosity ratio. The unknown viscosity of the gas at  $T$  is then obtained by comparing two ratios, that is

$$\eta_{0,T}^{\text{gas}} = \eta_{0,298}^{\text{He}} \left( \frac{\eta_{0,T}^{\text{He}}}{\eta_{0,298}^{\text{He}}} \right)_{\text{ab initio}} \left( \frac{\eta_{0,298}^{\text{gas}}}{\eta_{0,298}^{\text{He}}} \right) R_{T,298}^{\text{gas,He}}, \quad (3)$$

where  $(\eta_{0,T}^{\text{He}}/\eta_{0,298}^{\text{He}})_{\text{ab initio}}$  accounts for the ratio of the zero-density viscosities of helium at the temperature of  $T$  and 298.15 K obtained from *ab initio* calculations, and  $\eta_{0,298}^{\text{gas}}/\eta_{0,298}^{\text{He}}$  accounts for the ratio of the zero-density viscosities of the sample gas to helium at 298.15 K. All the correction terms are coupled into the temperature dependent ratio  $R_{T,298.15}^{\text{gas,He}}$  that is calculated by:

$$R_{T,298.15}^{\text{gas,He}} = \frac{(p_3^2 - p_4^2)^{\text{gas}}}{(p_1^2 - p_2^2)^{\text{gas}}} \frac{(p_1^2 - p_2^2)^{\text{He}}}{(p_3^2 - p_4^2)^{\text{He}}} \frac{C^{\text{gas}}(T, p_3, p_4)}{C^{\text{He}}(T, p_3, p_4)} \frac{C^{\text{He}}(298.15K, p_1, p_2)}{C^{\text{gas}}(298.15K, p_1, p_2)}, \quad (4)$$

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