

## Viscosity of Nitrogen, Helium, Hydrogen, and Argon from -100 to 25°C up to 150-250 atm\*

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The range of a previously developed capillary-flow viscometer has been extended to  $-100^{\circ}\text{C}$ , and the techniques of obtaining and determining the attainment of steady state have been improved. The viscosities of nitrogen, helium, hydrogen, and argon have been measured by an absolute method from  $-100$  or  $-90^{\circ}$  to  $25^{\circ}\text{C}$  and up to 150-175 atm (250 atm for  $\text{N}_2$  at  $25^{\circ}\text{C}$ ). The accuracy is estimated to be 0.1%-0.2%, but is somewhat worse for Ar at  $-100^{\circ}\text{C}$ . A number of empirical and theoretical analyses of the viscosity-density dependence have been made; the evidence is inconclusive as to the presence of a logarithmic term in the density expansion. Correlations of the second and the third virial coefficients of viscosity have been made.

### INTRODUCTION

We report here a series of absolute measurements of the viscosity of four gases as a function of temperature and density. The purpose of this work is to obtain experimental data on this transport property in regions where such information is unavailable or insufficient, to investigate the temperature dependence of the virial coefficients for the viscosity, and to provide, if possible, a test of proposed theories on the density expansion.

We have in previous publications<sup>1,2</sup> described our viscometer and given results for a number of gases at temperatures from  $-50$  to  $150^{\circ}\text{C}$ . The present work extends the temperature range to  $-90^{\circ}\text{C}$  for  $\text{N}_2$  and He, and to  $-100^{\circ}\text{C}$  for  $\text{H}_2$  and Ar, and repeats the previous measurements at  $-50$  and  $25^{\circ}\text{C}$  for more closely spaced values of the density. As before, the upper pressure limits are in the range 150-175 atm, except for  $\text{N}_2$  at  $25^{\circ}\text{C}$ , which we studied to about 250 atm. The precision of the data is better than 0.1%, and the results should be accurate to 0.1%-0.2%, with the exception of Ar at  $-100^{\circ}\text{C}$  (see discussion below).

At the time of publication of Ref. 1, there were hardly any other published measurements of the viscosity of gases below  $0^{\circ}\text{C}$  and above 1 atm, and none of comparable accuracy. This situation has improved in recent years, especially with Kao and Kobayashi's<sup>3</sup> work on  $\text{N}_2$  and He from  $-90$  to  $50^{\circ}\text{C}$  and Diller's<sup>4</sup> on

p- $\text{H}_2$  below  $100^{\circ}\text{K}$ , as well as some extensive measurements of much lesser accuracy mentioned below. However, none of these papers contains enough low-density data to give good values of the second virial coefficient of viscosity, which is of theoretical interest.<sup>5-9</sup> For this reason the present measurements have been extended to lower pressures than our previous work.<sup>1,2</sup>

### APPARATUS AND PROCEDURES

The apparatus and technique of measurement have been described in Ref. 1, and only some changes need be noted here; for a more extensive discussion, see Ref. 10. The apparatus is a constant-volume capillary-flow viscometer, in which an injector at  $25^{\circ}\text{C}$  drives gas at a known flow rate through a capillary at the experimental temperature, the resulting pressure difference being determined with a differential manometer.

The only major change is the addition of a fused-quartz precision pressure gauge (Texas Instruments Inc., Houston, Texas) in parallel with the differential manometer. This instrument registers a pressure difference by deflection of a quartz Bourdon tube. We had hoped to use it for absolute measurements, but found that the variation of the index of refraction of a gas with pressure affects the optical path in the instrument, so that its calibration varies with pressure. We therefore continued to use the mercury manometer, but made use of the pressure gauge to monitor the approach to steady state.

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<sup>1</sup> G. P. Flynn, R. V. Hanks, N. A. Lemaire, and J. Ross, *J. Chem. Phys.* **38**, 154 (1963). For corrections to the data given in Refs. 1-2, see comments below, especially Ref. 13, or Ref. 10 for more details.

<sup>2</sup> A. K. Barua, M. Afzal, G. P. Flynn, and J. Ross, *J. Chem. Phys.* **41**, 374 (1964). In Table V of their paper, *b* for  $\text{H}_2$  at  $75^{\circ}\text{C}$  should read 74.6 rather than 7.5.

<sup>3</sup> J. T. F. Kao and R. Kobayashi, *J. Chem. Phys.* **47**, 2836 (1967).

<sup>4</sup> D. E. Diller, *J. Chem. Phys.* **42**, 2089 (1965).

<sup>5</sup> D. E. Stogryn and J. O. Hirschfelder, *J. Chem. Phys.* **31**, 1545 (1959); **33**, 942 (1960).

<sup>6</sup> R. F. Snider and C. F. Curtiss, *Phys. Fluids* **1**, 122 (1958); C. F. Curtiss, M. B. McElroy, and D. K. Hoffman, *Intern. J. Eng. Sci.* **3**, 269 (1965).

<sup>7</sup> D. K. Hoffman and C. F. Curtiss, *Phys. Fluids* **8**, 890 (1965).

<sup>8</sup> S. K. Kim and J. Ross, *J. Chem. Phys.* **42**, 263 (1965); S. K. Kim, G. P. Flynn, and J. Ross, *ibid.* **43**, 4166 (1965).

<sup>9</sup> H. J. M. Hanley, R. D. McCarty, and J. V. Sengers, *J. Chem. Phys.* **50**, 857 (1969).

<sup>10</sup> J. A. Gracki, Ph.D. thesis, Brown University, 1969.