METHOD FOR DETERMINATION OF DENSITY OF CRYOGENIC LIQUIDS AND MIXTURES

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INTRODUCTION

Cryogenic liquids play an important role in many phases of industry and the space program. A knowledge of their physical and thermodynamic properties, e.g., density, heat capacity, surface tension, etc., is needed in solving many problems. It is essential to determine these properties accurately, preferably with relative ease.

This paper describes the development of a low-temperature experimental apparatus which is used for the determination of density of cryogenic liquids and mixtures. The density can be determined under atmospheric and subatmospheric pressure and in a temperature range of 77° to 273°K.

In selecting a suitable experimental method to obtain accurate density data, a review of the methods previously used for such measurements was made. After careful study of this review and error analyses of different techniques, it was concluded that the pycnometer method would yield the most accurate density results. This conclusion was based on the following considerations:

The main variables that affect the accuracy of the density values are temperature, pressure, mass, volume and composition. Because the equipment which is used to measure the temperature and pressure is essentially the same for all the methods, the effect of these two variables on the selection of the methods was eliminated. This led to the conclusion that the achievement of density data more accurate than those presently available depended on the improvement of the accuracy in measuring the mass, volume, and composition.

The pycnometer method as devised is simple. The density of a cryogenic liquid (pure or mixture) is determined by condensing the component gases from a high-pressure weighing bomb into a 30-cm³ low-temperature quartz pycnometer. The mass of each gas condensed is determined by direct differential weighing of the bomb with a 300-g analytical balance (0.1 mg sensitivity) using a set of class M standards calibrated by the National Bureau of Standards. The composition is accurately determined from the known masses of each component and the total mass of the mixture. Allowance is made for the amount of gases in the transfer line between the weighing bomb and the pycnometer. This transfer line is ⅛-in. copper tubing of a predetermined volume.

The volume of the pycnometer is measured at room temperature by differential weighings with mercury or distilled water. The volume at any other temperature is calculated by making use of the known thermal expansion data of fused quartz.
thermal expansion coefficient of the quartz used in this study is being determined as a function of temperature by the National Bureau of Standards.

The pycnometer is placed in a double dewar cryostat controlled to ±0.002°C. A detailed error analysis [1] shows that this apparatus is capable of density measurements to within an absolute accuracy of ±8 × 10⁻⁵ g/cm³ for pure methane. For an equimolal mixture of methane–propane, the calculated error is about twice that of pure methane.

**THE EXPERIMENTAL APPARATUS**

The experimental apparatus is presented schematically in Fig. 1. Not shown in the diagram is an insulation jacket which surrounds the analytical balance to reduce the thermal gradient in the balance case. The apparatus consists of the high-pressure weighing bomb, pycnometer, cryostat, and temperature measuring equipment.

**High-Pressure Weighing Bomb, B**

The weighing bomb with a specially designed light-weight valve (see Fig. 2) was constructed by welding together two 2-in. radius hemispheres hydroformed from flat