

SEP 2 1966

8862

DOLA JP64 0134

## Viscosity of Methane-*n*-Butane Mixtures

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Data are presented for methane-*n*-butane mixtures containing nominally 25, 50, 70, and 90 mole per cent methane. Each mixture is shown to have a viscosity field similar to that of a pure component and the data for each can be correlated by use of a residual plot. Use of molar densities in the residual plot is shown to simplify the presentation of the composition dependence of viscosity.

PREVIOUSLY REPORTED investigations have defined the viscosity behavior of ethane (11), propane (18), and *n*-butane (7) for significant ranges of pressure and temperature. Detailed studies of the viscosity behavior in the critical region for ethane, propane, and *n*-butane have been reported (17).

In this investigation, the objective was to obtain sufficient data on the viscosity of mixtures of methane and *n*-butane to permit study of the effects of composition on viscosity behavior. These data have been used in a definitive test of the applicability of the residual viscosity concept in correlating mixture viscosity data. The residual concept was used also to extrapolate the data on individual mixtures.

A search of the literature revealed only one previous investigation of the viscosity of a binary hydrocarbon system for an extensive pressure-temperature field. Bicher and Katz (2) presented values of viscosity for the pure components and mixtures in the system methane-propane. Pressures were between atmospheric and 5000 p.s.i.a. Temperatures ranged from 77° F. to 437° F. The accuracy claimed was  $\pm 3\%$ , average. These data were used in preliminary trials of several concepts and for comparison of behavior trends. No data were found on methane-*n*-butane mixtures.

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### APPARATUS AND MATERIALS

The viscometer employed in this investigation operated on the same principle as the instrument employed previously in pure component investigations (9, 18). As a normal operating procedure, check runs on nitrogen at several conditions were carried out before and after each experimental measurement of hydrocarbon systems to insure that the instrument behaved properly.

Auxiliary vessels and manifolding have been added to permit preparation and mixing of samples of specified composition. The sampling system has been modified to permit obtaining representative samples for analysis. The result of this change in installation is to completely isolate the test sample within the thermostat, thus eliminating the possibility of condensation or diffusion due to temperature gradients.

A schematic diagram of the complete system is shown in Figure 1. Samples were prepared by displacing a certain volume of each component from vessel *B* to vessel *C* at constant pressure and temperature. The volume of each component determines the mass and thus the composition of the sample. To mix the sample, the system exterior to vessel *C* is evacuated. The sample confined in vessel *C* is then flashed to fill the viscometer and vessel *B*. Mixing of

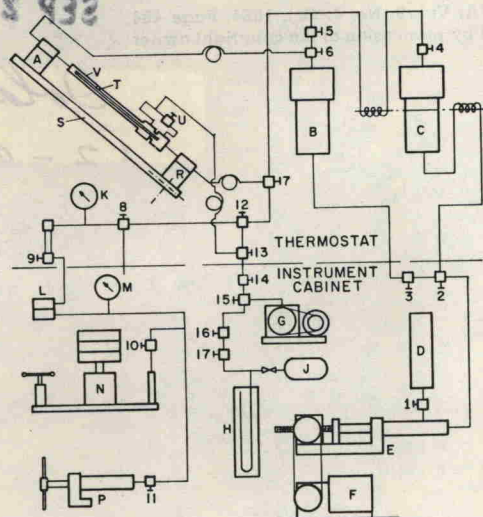


Figure 1. Schematic diagram, viscometer and auxiliary equipment

#### Vessels and equipment

- A. Viscometer instrument
- B. Stationary vessel
- C. Mixing vessel
- D. Mercury reservoir
- E. Mercury injection pump
- F. Electric motor for mercury pump
- G. Vacuum pump
- H. Manometer
- J. Sample bomb for mass spectrometer
- K. Estimating pressure gage
- L. Diaphragm separator
- M. Heise gage (0–10,000 p.s.i.g.)
- N. Deadweight gage (50–5,000 p.s.i.g.)
- P. High-pressure pump (5,000–10,000 p.s.i.g.)

#### Valves

- 1. Mercury reservoir control
- 2. Mixing vessel mercury control
- 3. Stationary vessel mercury control
- 4. Mixing vessel test fluid control
- 5. Stationary vessel test fluid control
- 6. Viscometer test fluid control
- 7. Viscometer test fluid control
- 8. Charging inlet control
- 9. Deadweight separator control
- 10. Deadweight gage control
- 11. Pressure pump control
- 12. Vacuum-sample system control from the manifold
- 13. Vacuum-sample system control from the viscometer
- 14. Vent valve
- 15. Vacuum pump control
- 16 and 17. Mass spectrometer sample high-pressure confinement cell

the sample is accomplished by displacing the sample back and forth between the two vessels on a path through the viscometer while maintaining a pressure that will keep the sample in a single phase. Movement of the sample is accomplished by displacement of mercury between the two vessels.

In the preparation of the methane-*n*-butane mixtures fluids of research grade were used without further purification. The *n*-butane was Phillips Petroleum Co. Research Grade, certified 99.9 mole % pure. The methane was obtained from the Southern California Gas Co. A typical analysis by mass spectrometer yielded 99.6% methane, 0.1% nitrogen, the remainder being ethane, propane, *n*-butane, and carbon dioxide. Analysis of completed mixtures showed that the ethane, propane, nitrogen and carbon dioxide were present in quantities not detectable by the mass spectrometer.

## EXPERIMENTAL DATA

Data were obtained on four binary mixtures of the system methane-*n*-butane. This system was chosen due to the availability of density data (15, 16) and the possibility of more extensive study of the regions near the phase boundaries within the temperature limits of the present thermostat bath. The pressure and temperature ranges for the data are summarized in Table I. The calculated viscosity values and information pertinent to the calculation and correlation of data have been tabulated and deposited with the A.D.I.

Mixtures of desired compositions were prepared by volumetric displacement technique as described in the previous section. However, the mass spectrometer analysis of a series of samples showed a scatter in composition of  $\pm 3\%$  with respect to the desired composition.

The approximate equation used to calculate the viscosity value from the experimental measurements (6) was used to estimate the uncertainty of the calculated value due to the uncertainty in the experimental measurements. In the liquid phase, i.e., temperatures below the critical and pressures well removed from the bubble point pressure, the uncertainty of the calculated value is  $\pm 1.5\%$ . The major contribution to this uncertainty is the composition effect.

In the gas phase the uncertainty of a viscosity value is  $\pm 0.5\%$ , with the equipment dimensions contributing all of the error. In regions of high gradients of density with pressure and temperature, the uncertainty is also  $\pm 0.5\%$ . In this last case the uncertainty is due to uncertainties in pressure and composition.

The plots of viscosity vs. pressure and viscosity vs. temperature, Figures 2 and 3, illustrate the general behavior of the mixtures. The estimated phase boundaries were obtained by extrapolating from higher or lower pressures to the dew- and bubble-point pressures. All data are shown by open symbols. Where isotherms or isobars of viscosity extend beyond the range of the data, construction from interpolated values was necessary. The interpolated values were obtained by use of the residual viscosity-density plots discussed below, in conjunction with the viscosity-pressure and viscosity-temperature plots. The effect of composition on viscosity is illustrated in Figure 4.

## DATA TREATMENT

The concept of a transport property "residual" has its origin in the studies by Predvoditelev (14) of invariant quantities in heat transfer and liquid viscosity theory. Abas-Zade (1) applied the work of Predvoditelev to correlation of thermal conductivity data on liquids and vapors. The result of this application was that the residual thermal conductivity, defined as the thermal conductivity at the same temperature and a low pressure (atmospheric), at a given density was invariant with respect to temperature.

Table I. Temperatures and Pressure Ranges for Methane-*n*-Butane Data<sup>a</sup>

| Temp.<br>° F.            | Pressure,<br>P.S.I.A. | Temp.<br>° F.            | Pressure,<br>P.S.I.A. |
|--------------------------|-----------------------|--------------------------|-----------------------|
| 25 Mole Per Cent Methane |                       | 50 Mole Per Cent Methane |                       |
| 100                      | 1000 to 2500          | 100                      | 2000 to 4000          |
| 160                      | 1000 to 2500          | 160                      | 2000 to 3500          |
| 220                      | 1250 to 2500          | 220                      | 2000 to 3000          |
| 340                      | 700 to 2000           | 280                      | 600 to 3000           |
| 70 Mole Per Cent Methane |                       | 90 Mole Per Cent Methane |                       |
| 100                      | 1900 to 6000          | 70                       | 2000 to 3000          |
| 160                      | 1750 to 6000          | 100                      | 600 to 8000           |
| 169                      | 800 to 1825           | 160                      | 600 to 3000           |
| 171                      | 530 to 2000           | 220                      | 800 to 5000           |
| 190                      | 540 to 2500           |                          |                       |
| 220                      | 200 to 2500           |                          |                       |

<sup>a</sup>Composition uncertainty,  $\pm 3\%$