

Figure 2. Gas phase region, 70 per cent methane, 30 per cent *n*-butane mixture

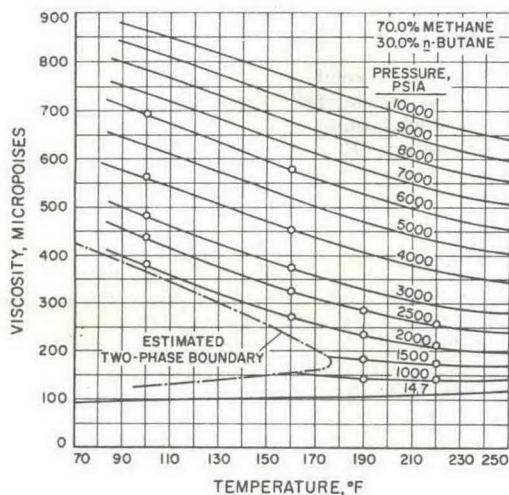


Figure 3. Viscosity vs. temperature, 70 per cent methane, 30 per cent *n*-butane mixture

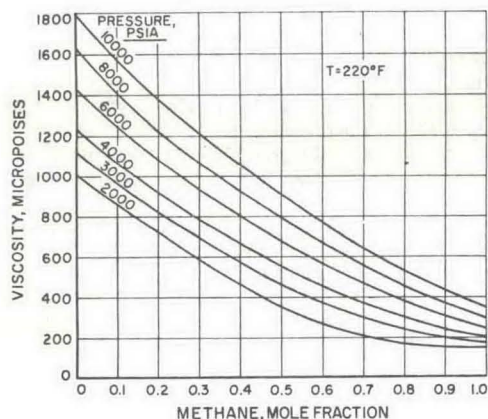


Figure 4. Viscosity vs. composition at 220° F.

This was proven by plotting residual thermal conductivity vs. density on log-log coordinates with a single curve representing all data. This type of plot was used by Brebach and Thodos (3) to correlate viscosity behavior. The correlation, plotted on linear coordinates, has been used successfully in previous investigations (7, 18) on pure components for checking the internal consistency of data, interpolation and limited extrapolation.

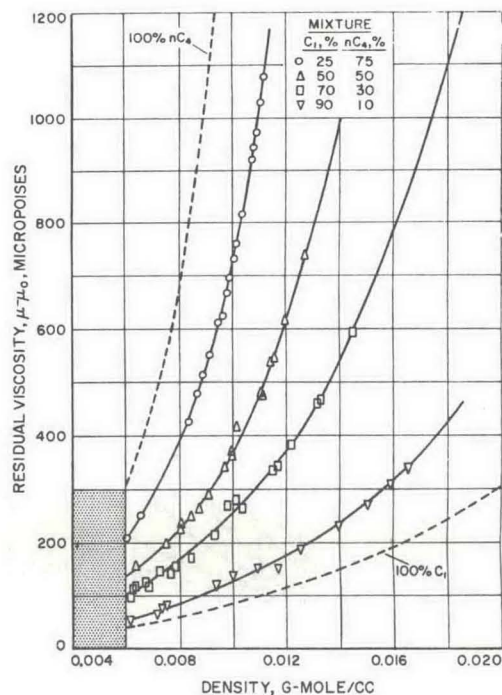


Figure 5. Residual viscosity plot of methane-*n*-butane mixtures, high density region

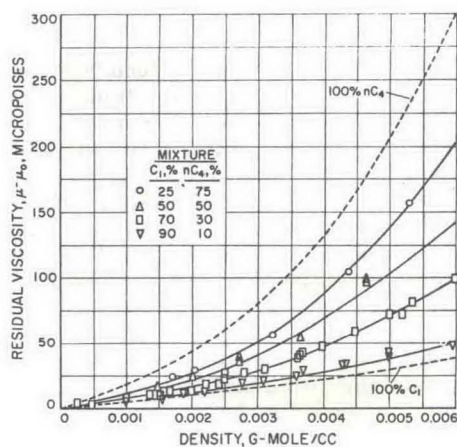


Figure 6. Residual viscosity plot of methane-*n*-butane mixtures, low density region

It was reasonable to expect that the data on a single homogeneous mixture could also be rectified by a residual plot. Exploratory tests with the data of Bicher and Katz (2) confirmed this.

Residual plots were employed successfully by Eakin and Ellington to ascertain and generalize molecular weight dependence for pure light hydrocarbons (8, 10). Therefore, it was logical that efforts be made to employ the concept to show composition dependence within a given system. On log-log plots, involving density, the available binary system data exhibited considerable disordering and intersection of constant composition lines. This might be expected from studies such as that of Hirschfelder and coworkers (13), to determine the conditions necessary for two-component gaseous systems to exhibit viscosity maxima with composition. Molar bases are conventionally employed to determine molecular effects and so plots of residual viscosity vs. density in gram mole/cc. were prepared (Figure 5 and 6); linear coordinates were employed to avoid emphasizing the scatter of data at low densities. The pure methane data employed were those of Comings and Mayland (5) and the pure *n*-butane data, those of Dolan and coworkers (7).

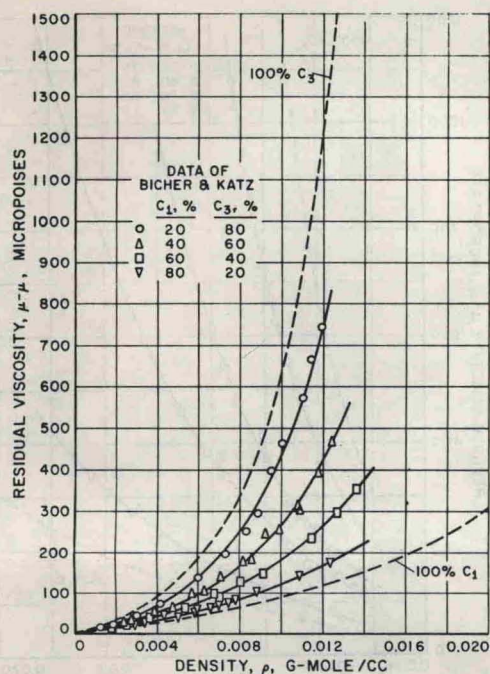


Figure 7. Residual viscosity plot of methane-propane mixtures

Several observations can be made on the basis of Figures 5 and 6. The effects of change of composition in the methane-*n*-butane system show regular trends between the limits of the two pure components. A plot of the methane-propane mixture data of Bicher and Katz (2), utilizing the propane data of Starling, Eakin, and Ellington (18) and the methane data of Comings and Mayland (5), Figure 7, yielded similar results. Possibilities for development of empirical mixing rules are immediately evident; one would be a fit to the curves obtained by cuts at constant density. It is felt that the residual plot using molar densities can be employed to estimate the viscosity of other methane-*n*-butane mixtures with reasonable accuracy and augment the other correlations available. It should be mentioned that a proven method for estimating the atmospheric pressure viscosity values exists (4). This method employs a mixing rule developed by Herning and Zipperer (12) and atmospheric pressure information on the pure components. This mixing rule has the form

$$\mu_m = \frac{\sum x_i \mu_i (M_i)^{1/2}}{\sum x_i (M_i)^{1/2}}$$

The pure component viscosities μ_i can be determined by the Sutherland Equation

$$\mu = \frac{BT^{3/2}}{T+S}$$

with appropriate constants, B and S . For the purpose of this investigation the Sutherland constants for methane, ethane and propane were those of Trautz and Sorg (19). The constants for *n*-butane were determined from the data of Dolan and coworkers (7). The Herning and Zipperer equation had previously been tested on light natural gases (4). The authors tested the mixing rule on the methane-

propane data of Trautz and Sorg; it reproduced the data with an average deviation of $\pm 0.5\%$.

SUMMARY

The data produced by this investigation serve to define the effect of composition on viscosity in the methane-*n*-butane system. The region covered is from 100 to 10,000 p.s.i.a. for temperatures from 100° to 460° F. It has been demonstrated that the concept of residual viscosity correlates the behavior of mixtures as well as it does data on pure components.

ACKNOWLEDGMENT

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NOMENCLATURE

- B = Sutherland constant
- M_i = Molecular weight of the i^{th} component
- S = Sutherland constant
- T = Temperature, °R.
- μ = viscosity, micropoise
- μ_m = viscosity of the mixture, micropoise

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