

# Compressibility Factors of Nitrogen-n-Butane Mixtures in the Gas Phase

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Nitrogen and n-butane are common components of industrial gaseous systems. The literature contains no experimental information on compressibility factors of nitrogen-n-butane mixtures in the gas phase.

The isotherms reported are at 310°, 340°, 370°, and 400° F. for a pressure range extending from 200 to 10,000 pounds per square inch absolute. Compressibility factors are tabulated for pressures at experimental values, and also at rounded values for mixtures of 0.100, 0.300, 0.500, 0.700, and 0.900 mole fraction of n-butane.

The results presented should prove useful in developing means for predicting the behavior of mixtures of hydrocarbons with nitrogen.

Nitrogen and n-butane are common and important components of industrial gaseous mixtures. The volumetric behavior of pure nitrogen has been of interest to numerous investigators (1-5, 11, 16, 17, 19, 20, 24, 25). Tabular and graphical presentations of the compressibility factors of nitrogen, prepared on the basis of the more precise data, are available (12, 23). Compressibility data on pure n-butane are likewise readily found (6, 10, 18, 22), and tabular and graphical summaries have also been prepared (23). Compressibility factors of some gaseous nitrogen-hydrocarbon mixtures are similarly found in the literature. Nitrogen-methane mixtures have been investigated by Keyes and Burks (15), and the results have been used as a test for the Beattie-Bridgeman equation of state applied to mixtures (7, 9, 14, 15). Nitrogen-ethane data have been presented by Reamer and others (21). Limited data, obtained in this laboratory, have been reported on gaseous nitrogen-propane mixtures (26). Nitrogen-ethylene mixtures have been studied by Hagenbach and Comings (13).

No previous investigation on the compressibility factors of gaseous nitrogen-n-butane mixtures were found. In the present work, the compressibility factor is defined in the usual manner as the dimensionless ratio of  $PV/RT$ .

## MATERIALS

Both the nitrogen and the n-butane used were acquired from the Mathieson Co., Joliet, Ill. The nitrogen was of the "pre-purified grade" and the n-butane was of the "instrument grade." According to the manufacturer's specifications, each gas was 99.9% pure.

Mass-spectroscopic analysis showed the nitrogen to conform to specifications. The purity of the n-butane was checked by means of a mass spectrometer, an infrared spectrophotometer, and a Hyd-Robot Podbielniak low temperature distillation column, and by examination of the slope of the curve of specific volume vs. pressure in the two-phase region. The analyses showed absence of detectable amounts of propane, isobutane, and pentanes. Slight traces of air were found present to an extent below quantitative but not qualitative detection. In view of the slight air impurities detected, a loading procedure was devised, so as to enable removal of the air impurity. With the procedure used to prepare the mixtures, the probable purity of the n-butane was not less than 99.9%.

## APPARATUS AND PROCEDURE

The design and arrangement of the apparatus used, with the proper adaptations for investigations of mixtures

of nitrogen-n-butane, were based on the detailed descriptions and specifications of Beattie (8) and of Keyes (14). Aside from the loading equipment, the apparatus consisted of an Amagat-type dead-weight pressure gage with its hydraulic oil system, a mercury "U-tube," a calibrated mercury injector or pump kept in a thermostat at 45° C., and an equilibrium bomb of accurately known volume provided with a Stellite-tipped metering valve. The equilibrium bomb was kept in a separate thermostat. All the units were connected by means of high pressure capillary tubing through a manifold with the necessary valves for convenient operation.

The principles involved in measurements of pressure, volume, and temperature of the gases were simple. The fluid mixture under investigation was introduced into the equilibrium bomb. The bomb was held at constant temperature and had connections leading to the mercury injector and to the pressure gage. By injection of the proper amount of mercury into the equilibrium bomb, the gas mixture was compressed until a pressure balance was detected. The temperature was measured by means of a platinum resistance thermometer calibrated by the National Bureau of Standards in conjunction with a Mueller bridge and a high sensitivity galvanometer. The temperature was controlled to -0.05° C. in the high temperature thermostat and to -0.01° C. in the low temperature thermostat. The pressure was measured by means of an Amagat dead-weight gage (26), with calibrated weights and mercury U-tube (8). The pressure could be measured with a precision within 0.03%. The volumes were obtained from the carefully determined volume of the equilibrium bomb and the volumes of mercury injected by the calibrated mercury pump following the method outlined by Beattie (8). "Blank run" corrections (8) for the apparent volume changes of the apparatus with pressure were experimentally determined at all temperatures investigated and were applied as well as all other corrections specified (8). The precision of the volume measurements ranged from within 0.05% at pressures below 1000 pounds per square inch absolute to 0.5% at pressures in the neighborhood of 10,000.

In addition to pressure, volume, and temperature, two additional variables had to be determined: mole fraction and total number of moles of gas in the mixtures, which were measured during the loading procedure. The mass of butane charged was measured gravimetrically. Briefly, the butane was distilled from its storage cylinder into evacuated weighing bombs immersed in liquid nitrogen. Traces of air or nitrogen contamination were removed by successive evacuations of the frozen hydrocarbon. Two distillations and two separate weighings were performed prior to introducing the butane into the equilibrium bomb. Independent weighings showed that the weights could be reproduced to  $\pm 0.1$  mg. On this basis, the differences should have been precise to  $\pm 0.2$  mg. for charges of butane ranging from 800 to 5000 mg. The molecular weight for butane used in the calculations was 58.121.

The number of moles of nitrogen charged were measured volumetrically after the butane had been charged into the compressibility bomb. For this purpose, a loading bomb placed side by side with the mercury pump and kept at 45° C. in the low temperature thermostat was used. This bomb, however, did not form part of the system during the compressibility measurements and was used only during the loading procedure.

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TABLE I. COMPRESSIBILITY FACTORS FOR n-BUTANE-NITROGEN MIXTURES  
(Experimental data)

| Lb./Sq. Inch<br>Abs.              | z       |         |         |         | Lb./Sq. Inch<br>Abs.               | z       |         |         |         |
|-----------------------------------|---------|---------|---------|---------|------------------------------------|---------|---------|---------|---------|
|                                   | 310° F. | 340° F. | 370° F. | 400° F. |                                    | 310° F. | 340° F. | 370° F. | 400° F. |
| Mixture I, 9.468 mole % nitrogen  |         |         |         |         | Mixture III, 49.74 mole % nitrogen |         |         |         |         |
| 405.9                             | 0.7615  | ..      | ..      | ..      | 1805.                              | 0.6169  | 0.6603  | 0.7028  | 0.7428  |
| 432.2                             | ..      | 0.7819  | ..      | ..      | 1931.                              | 0.6262  | 0.6661  | 0.7054  | 0.7436  |
| 454.1                             | 0.7231  | ..      | ..      | ..      | 2216.                              | 0.6556  | 0.6881  | 0.7213  | 0.7542  |
| 461.9                             | ..      | ..      | 0.7998  | ..      | 2494.                              | 0.6887  | 0.7161  | 0.7448  | 0.7736  |
| 484.2                             | 0.6996  | 0.7509  | 0.7888  | ..      | 2782.                              | 0.7307  | 0.7522  | 0.7753  | 0.7996  |
| 533.5                             | 0.6586  | 0.7200  | 0.7636  | ..      | 3057.                              | 0.7716  | 0.7887  | 0.8077  | 0.8282  |
| 582.8                             | 0.6137  | 0.6882  | 0.7399  | ..      | 3362.                              | 0.8177  | 0.8313  | 0.8466  | 0.8633  |
| 632.2                             | 0.5653  | 0.6552  | 0.7147  | ..      | 3982.                              | 0.9146  | 0.9219  | 0.9307  | 0.9408  |
| 662.7                             | 0.5334  | 0.6348  | 0.6987  | ..      | 4592.                              | 1.011   | 1.012   | 1.015   | 1.020   |
| 711.7                             | 0.4812  | 0.5996  | 0.6731  | ..      | 5207.                              | 1.108   | 1.104   | 1.103   | 1.104   |
| 766.6                             | 0.4283  | 0.5618  | 0.6414  | ..      | 5823.                              | 1.204   | 1.197   | 1.191   | 1.184   |
| 794.7                             | ..      | 0.5423  | ..      | ..      | 6436.                              | 1.299   | 1.289   | 1.280   | 1.273   |
| 816.1                             | 0.3937  | 0.5285  | 0.6197  | ..      | 7047.                              | 1.393   | 1.378   | 1.367   | 1.356   |
| 865.6                             | 0.3716  | 0.4982  | 0.5954  | ..      | 7666.                              | 1.488   | 1.469   | 1.454   | 1.440   |
| 899.1                             | ..      | 0.4804  | ..      | ..      | 8282.                              | 1.581   | 1.559   | 1.540   | 1.522   |
| 915.2                             | 0.3601  | 0.4726  | 0.5727  | ..      | 8854.                              | 1.664   | 1.640   | 1.618   | 1.597   |
| 942.8                             | 0.3552  | 0.4612  | 0.5605  | ..      | 9182.                              | 1.716   | 1.688   | 1.664   | 1.641   |
| 998.3                             | ..      | 0.4431  | ..      | ..      | 9660.                              | 1.786   | 1.756   | 1.730   | 1.705   |
| 1017.                             | 0.3544  | ..      | 0.5330  | ..      | 9844.                              | 1.814   | 1.783   | 1.754   | 1.730   |
| 1078.                             | 0.3575  | 0.4272  | 0.5152  | ..      | 9997.                              | 1.836   | 1.804   | 1.776   | 1.750   |
| 1177.                             | ..      | 0.4225  | 0.4964  | ..      |                                    |         |         |         |         |
| 1252.                             | 0.3832  | ..      | ..      | ..      | Mixture IV, 70.16 mole % nitrogen  |         |         |         |         |
| 1282.                             | ..      | 0.4275  | 0.4880  | ..      | 488.1                              | 0.9215  | ..      | ..      | ..      |
| 1359.                             | 0.3978  | 0.4350  | 0.4873  | ..      | 514.4                              | ..      | 0.9322  | ..      | ..      |
| 1514.                             | 0.4263  | 0.4552  | 0.4952  | ..      | 534.2                              | 0.9154  | 0.9289  | 0.9402  | ..      |
| 1651.                             | 0.4524  | 0.4759  | 0.5098  | ..      | 583.6                              | 0.9087  | 0.9226  | 0.9350  | 0.9457  |
| 1805.                             | 0.4815  | 0.5010  | 0.5299  | ..      | 633.0                              | 0.9000  | 0.9166  | 0.9302  | 0.9423  |
| 1931.                             | 0.5056  | 0.5238  | 0.5483  | ..      | 663.0                              | 0.8956  | 0.9128  | 0.9270  | 0.9391  |
| 2216.                             | ..      | 0.5753  | 0.5931  | ..      | 712.5                              | 0.8879  | 0.9069  | 0.9222  | 0.9351  |
| 2494.                             | 0.6155  | 0.6261  | 0.6402  | ..      | 767.4                              | 0.8808  | 0.9008  | 0.9172  | 0.9310  |
| 2782.                             | ..      | 0.6798  | 0.6890  | ..      | 816.9                              | 0.8744  | 0.8955  | 0.9127  | 0.9274  |
| 3057.                             | 0.7242  | 0.7301  | 0.7379  | ..      | 866.4                              | 0.8681  | 0.8903  | 0.9085  | 0.9239  |
| 3362.                             | 0.7821  | 0.7857  | 0.7900  | ..      | 916.0                              | 0.8622  | 0.8854  | 0.9044  | 0.9205  |
| 3982.                             | 0.8968  | 0.8978  | 0.8957  | ..      | 943.6                              | 0.8589  | 0.8827  | 0.9022  | 0.9188  |
| 4592.                             | 1.011   | 1.007   | 1.000   | ..      | 1018.                              | 0.8506  | 0.8760  | 0.8966  | 0.9143  |
| 5207.                             | 1.123   | 1.114   | 1.105   | ..      | 1079.                              | 0.8446  | 0.8710  | 0.8924  | 0.9107  |
| 5823.                             | 1.234   | 1.223   | 1.208   | ..      | 1178.                              | 0.8355  | 0.8635  | 0.8863  | 0.9058  |
| 6436.                             | 1.342   | 1.327   | 1.309   | ..      | 1283.                              | 0.8274  | 0.8567  | 0.8807  | 0.9014  |
| 7047.                             | 1.449   | 1.427   | 1.409   | ..      | 1360.                              | 0.8222  | 0.8524  | 0.8772  | 0.8984  |
| 7666.                             | 1.556   | 1.531   | 1.509   | ..      | 1514.                              | 0.8145  | 0.8457  | 0.8718  | 0.8942  |
| 8282.                             | 1.660   | 1.633   | 1.604   | ..      | 1651.                              | 0.8103  | 0.8420  | 0.8688  | 0.8920  |
| 8854.                             | 1.758   | 1.727   | 1.695   | ..      | 1806.                              | 0.8086  | 0.8404  | 0.8674  | 0.8911  |
| 9182.                             | 1.813   | 1.780   | 1.749   | ..      | 1931.                              | 0.8094  | 0.8408  | 0.8678  | 0.8916  |
| 9660.                             | 1.892   | 1.857   | 1.822   | ..      | 2217.                              | 0.8173  | 0.8468  | 0.8730  | 0.8964  |
| 9844.                             | 1.922   | 1.889   | 1.850   | ..      | 2494.                              | 0.8318  | 0.8591  | 0.8834  | 0.9058  |
| Mixture II, 29.57 mole % nitrogen |         |         |         |         | 2782.                              | 0.8520  | 0.8763  | 0.8989  | 0.9194  |
| 422.2                             | 0.8597  | ..      | ..      | ..      | 3057.                              | 0.8756  | 0.8967  | 0.9171  | 0.9359  |
| 429.6                             | 0.8566  | ..      | ..      | ..      | 3362.                              | 0.9044  | 0.9224  | 0.9405  | 0.9574  |
| 445.5                             | ..      | 0.8736  | ..      | ..      | 3982.                              | 0.9697  | 0.9831  | 0.9960  | 1.009   |
| 466.9                             | ..      | ..      | 0.8869  | ..      | 4592.                              | 1.040   | 1.048   | 1.056   | 1.065   |
| 484.3                             | 0.8375  | 0.8622  | 0.8827  | ..      | 5207.                              | 1.115   | 1.119   | 1.122   | 1.127   |
| 533.6                             | 0.8201  | 0.8480  | 0.8705  | 0.8981  | 5823.                              | 1.190   | 1.190   | 1.190   | 1.191   |
| 583.0                             | 0.8028  | 0.8339  | 0.8589  | 0.8791  | 6436.                              | 1.267   | 1.262   | 1.259   | 1.256   |
| 632.4                             | 0.7856  | 0.8197  | 0.8470  | 0.8694  | 7047.                              | 1.343   | 1.335   | 1.328   | 1.322   |
| 662.5                             | 0.7752  | 0.8111  | 0.8400  | 0.8635  | 7666.                              | 1.421   | 1.409   | 1.398   | 1.389   |
| 712.0                             | 0.7584  | 0.7975  | 0.8284  | 0.8541  | 8282.                              | 1.497   | 1.483   | 1.469   | 1.456   |
| 766.9                             | 0.7405  | 0.7828  | 0.8162  | 0.8440  | 8854.                              | 1.567   | 1.549   | 1.532   | 1.517   |
| 816.4                             | 0.7240  | 0.7698  | 0.8082  | 0.8351  | 9182.                              | 1.608   | 1.589   | 1.571   | 1.554   |
| 866.0                             | 0.7091  | 0.7561  | 0.7954  | 0.8260  | 9660.                              | 1.667   | 1.646   | 1.625   | 1.607   |
| 915.5                             | 0.6940  | 0.7452  | 0.7852  | 0.8178  | 9844.                              | 1.689   | 1.668   | 1.645   | 1.627   |
| 943.2                             | 0.6852  | 0.7387  | 0.7798  | 0.8134  | 9997.                              | 1.707   | 1.686   | 1.663   | 1.643   |
| 1017.                             | 0.6668  | 0.7223  | 0.7656  | 0.8009  |                                    |         |         |         |         |
| 1078.                             | 0.6529  | 0.7095  | 0.7555  | 0.7930  | Mixture IV, 70.16 mole % nitrogen  |         |         |         |         |
| 1178.                             | 0.6340  | 0.6928  | 0.7401  | 0.7799  | 488.1                              | 0.9738  | ..      | ..      | ..      |
| 1282.                             | 0.6203  | 0.6783  | 0.7266  | 0.7682  | 514.4                              | ..      | 0.9784  | ..      | ..      |
| 1360.                             | 0.6137  | 0.6701  | 0.7188  | 0.7606  | 534.2                              | 0.9717  | 0.9779  | 0.9832  | ..      |
| 1514.                             | 0.6079  | 0.6601  | 0.7079  | 0.7499  | 583.5                              | 0.9699  | 0.9760  | 0.9824  | 0.9867  |
| 1651.                             | 0.6096  | 0.6575  | 0.7030  | 0.7443  | 632.9                              | 0.9677  | 0.9748  | 0.9810  | 0.9860  |

**TABLE I. COMPRESSIBILITY FACTORS FOR n-BUTANE-NITROGEN MIXTURES (Contd.)**

| Lb./Sq. Inch<br>Abs. | z       |         |         |         |
|----------------------|---------|---------|---------|---------|
|                      | 310° F. | 340° F. | 370° F. | 400° F. |
| 663.0                | 0.9664  | 0.9739  | 0.9802  | 0.9854  |
| 712.5                | 0.9636  | 0.9726  | 0.9792  | 0.9847  |
| 767.4                | 0.9630  | 0.9714  | 0.9784  | 0.9843  |
| 816.9                | 0.9613  | 0.9703  | 0.9777  | 0.9840  |
| 866.5                | 0.9598  | 0.9692  | 0.9771  | 0.9836  |
| 916.0                | 0.9585  | 0.9684  | 0.9763  | 0.9833  |
| 943.6                | 0.9583  | 0.9679  | 0.9762  | 0.9831  |
| 1018.                | 0.9565  | 0.9667  | 0.9754  | 0.9828  |
| 1079.                | 0.9560  | 0.9660  | 0.9752  | 0.9829  |
| 1178.                | 0.9543  | 0.9651  | 0.9749  | 0.9830  |
| 1283.                | 0.9531  | 0.9645  | 0.9748  | 0.9834  |
| 1360.                | 0.9522  | 0.9644  | 0.9750  | 0.9839  |
| 1514.                | 0.9517  | 0.9648  | 0.9760  | 0.9855  |
| 1651.                | 0.9521  | 0.9658  | 0.9776  | 0.9874  |
| 1806.                | 0.9546  | 0.9678  | 0.9800  | 0.9903  |
| 1931.                | 0.9558  | 0.9700  | 0.9827  | 0.9932  |
| 2217.                | 0.9624  | 0.9770  | 0.9901  | 1.001   |
| 2494.                | 0.9721  | 0.9864  | 0.9995  | 1.010   |
| 2782.                | 0.9849  | 0.9987  | 1.011   | 1.022   |
| 3057.                | 0.9990  | 1.011   | 1.024   | 1.034   |
| 3362.                | 1.017   | 1.029   | 1.041   | 1.049   |
| 3982.                | 1.059   | 1.069   | 1.079   | 1.085   |
| 4592.                | 1.107   | 1.114   | 1.121   | 1.125   |
| 5207.                | 1.159   | 1.163   | 1.167   | 1.169   |
| 5823.                | 1.213   | 1.215   | 1.216   | 1.216   |
| 6436.                | 1.270   | 1.268   | 1.267   | 1.264   |
| 7047.                | 1.327   | 1.322   | 1.319   | 1.313   |
| 7666.                | 1.386   | 1.379   | 1.372   | 1.363   |
| 8282.                | 1.445   | 1.436   | 1.426   | 1.414   |
| 8854.                | 1.499   | 1.486   | 1.474   | 1.460   |
| 9182.                | 1.531   | 1.518   | 1.505   | 1.490   |
| 9660.                | 1.577   | 1.562   | 1.547   | 1.530   |
| 9843.                | 1.595   | 1.579   | 1.563   | 1.546   |
| 9997.                | 1.610   | 1.593   | 1.577   | 1.559   |

Mixture V, 90.64 mole % nitrogen

|       |       |       |       |       |
|-------|-------|-------|-------|-------|
| 663.9 | 1.008 | ..    | ..    | ..    |
| 713.6 | 1.010 | 1.012 | 1.013 | ..    |
| 768.4 | 1.011 | 1.013 | 1.015 | 1.017 |
| 817.9 | 1.012 | 1.014 | 1.016 | 1.018 |
| 867.4 | 1.012 | 1.015 | 1.017 | 1.019 |
| 916.9 | 1.013 | 1.016 | 1.018 | 1.020 |
| 944.4 | 1.014 | 1.016 | 1.019 | 1.021 |
| 1019. | 1.015 | 1.018 | 1.020 | 1.023 |
| 1080. | 1.016 | 1.019 | 1.022 | 1.024 |
| 1179. | 1.018 | 1.022 | 1.025 | 1.027 |
| 1256. | 1.021 | 1.024 | ..    | 1.030 |
| 1360. | 1.023 | 1.026 | 1.030 | 1.032 |
| 1515. | 1.027 | 1.031 | 1.034 | 1.037 |
| 1652. | 1.030 | 1.035 | 1.038 | 1.041 |
| 1806. | 1.036 | 1.040 | 1.043 | 1.046 |
| 1932. | 1.040 | 1.044 | 1.047 | 1.051 |
| 2217. | 1.049 | 1.054 | 1.057 | 1.061 |
| 2495. | 1.060 | 1.064 | 1.068 | 1.071 |
| 2782. | 1.072 | 1.076 | 1.080 | 1.082 |
| 3057. | 1.084 | 1.088 | 1.091 | 1.094 |
| 3362. | 1.098 | 1.101 | 1.104 | 1.107 |
| 3982. | 1.129 | 1.132 | 1.134 | 1.135 |
| 4592. | 1.162 | 1.164 | 1.164 | 1.165 |
| 5207. | 1.198 | 1.198 | 1.197 | 1.197 |
| 5823. | 1.235 | 1.233 | 1.232 | 1.230 |
| 6436. | 1.274 | 1.271 | 1.267 | 1.264 |
| 7047. | 1.313 | 1.316 | 1.321 | 1.298 |
| 7666. | 1.354 | 1.356 | 1.340 | 1.333 |
| 8282. | 1.395 | 1.386 | 1.378 | 1.370 |
| 8854. | 1.432 | 1.421 | 1.411 | 1.401 |
| 9182. | 1.456 | 1.444 | 1.433 | 1.423 |
| 9660. | 1.488 | 1.475 | 1.463 | 1.452 |
| 9844. | 1.501 | 1.487 | 1.474 | 1.463 |
| 9997. | 1.512 | 1.498 | 1.484 | 1.472 |

**TABLE II. COMPRESSIBILITY FACTORS FOR NITROGEN-BUTANE MIXTURES (Smoothed data)**

| Pressure,<br>Lb./Sq. Inch | Mole Fraction Butane |       |       |       |       |
|---------------------------|----------------------|-------|-------|-------|-------|
|                           | 0.100                | 0.300 | 0.500 | 0.700 | 0.900 |
| At 310° F.                |                      |       |       |       |       |
| 200                       | 1.002                | 0.989 | 0.969 | 0.937 | 0.895 |
| 400                       | 1.004                | 0.978 | 0.938 | 0.868 | 0.766 |
| 600                       | 1.007                | 0.970 | 0.908 | 0.796 | 0.600 |
| 800                       | 1.010                | 0.962 | 0.882 | 0.736 | 0.404 |
| 1000                      | 1.014                | 0.958 | 0.858 | 0.674 | 0.356 |
| 1250                      | 1.019                | 0.952 | 0.833 | 0.626 | 0.376 |
| 1500                      | 1.025                | 0.951 | 0.815 | 0.609 | 0.424 |
| 1750                      | 1.032                | 0.952 | 0.810 | 0.614 | 0.474 |
| 2000                      | 1.040                | 0.956 | 0.811 | 0.635 | 0.522 |
| 2250                      | 1.048                | 0.963 | 0.819 | 0.661 | 0.570 |
| 2500                      | 1.058                | 0.972 | 0.834 | 0.692 | 0.618 |
| 2750                      | 1.068                | 0.983 | 0.853 | 0.726 | 0.665 |
| 3000                      | 1.079                | 0.994 | 0.875 | 0.765 | 0.711 |
| 3500                      | 1.102                | 1.025 | 0.925 | 0.843 | 0.806 |
| 4000                      | 1.127                | 1.059 | 0.982 | 0.920 | 0.899 |
| 4500                      | 1.153                | 1.097 | 1.040 | 0.996 | 0.992 |
| 5000                      | 1.181                | 1.139 | 1.096 | 1.074 | 1.083 |
| 5500                      | 1.211                | 1.183 | 1.159 | 1.153 | 1.173 |
| 6000                      | 1.244                | 1.229 | 1.222 | 1.231 | 1.262 |
| 6500                      | 1.276                | 1.277 | 1.285 | 1.308 | 1.350 |
| 7000                      | 1.309                | 1.325 | 1.348 | 1.383 | 1.437 |
| 7500                      | 1.342                | 1.372 | 1.411 | 1.460 | 1.522 |
| 8000                      | 1.376                | 1.421 | 1.473 | 1.535 | 1.609 |
| 8500                      | 1.409                | 1.469 | 1.534 | 1.610 | 1.696 |
| 9000                      | 1.442                | 1.516 | 1.595 | 1.684 | 1.781 |
| 9500                      | 1.474                | 1.564 | 1.657 | 1.757 | 1.865 |
| 10000                     | 1.508                | 1.610 | 1.718 | 1.827 | 1.948 |
| At 340° F.                |                      |       |       |       |       |
| 200                       | 1.003                | 0.991 | 0.974 | 0.945 | 0.908 |
| 400                       | 1.005                | 0.982 | 0.947 | 0.888 | 0.804 |
| 600                       | 1.008                | 0.975 | 0.922 | 0.829 | 0.678 |
| 800                       | 1.012                | 0.970 | 0.899 | 0.775 | 0.541 |
| 1000                      | 1.016                | 0.966 | 0.880 | 0.728 | 0.437 |
| 1250                      | 1.021                | 0.964 | 0.860 | 0.686 | 0.428 |
| 1500                      | 1.028                | 0.963 | 0.848 | 0.663 | 0.457 |
| 1750                      | 1.035                | 0.966 | 0.842 | 0.660 | 0.496 |
| 2000                      | 1.043                | 0.970 | 0.844 | 0.672 | 0.539 |
| 2250                      | 1.051                | 0.976 | 0.850 | 0.693 | 0.583 |
| 2500                      | 1.061                | 0.985 | 0.861 | 0.719 | 0.629 |
| 2750                      | 1.071                | 0.996 | 0.875 | 0.749 | 0.674 |
| 3000                      | 1.082                | 1.007 | 0.894 | 0.782 | 0.720 |
| 3500                      | 1.104                | 1.035 | 0.939 | 0.853 | 0.810 |
| 4000                      | 1.128                | 1.068 | 0.991 | 0.926 | 0.900 |
| 4500                      | 1.154                | 1.103 | 1.045 | 0.998 | 0.989 |
| 5000                      | 1.180                | 1.143 | 1.098 | 1.073 | 1.076 |
| 5500                      | 1.209                | 1.184 | 1.158 | 1.148 | 1.161 |
| 6000                      | 1.240                | 1.228 | 1.217 | 1.222 | 1.247 |
| 6500                      | 1.271                | 1.273 | 1.277 | 1.296 | 1.332 |
| 7000                      | 1.302                | 1.319 | 1.337 | 1.369 | 1.416 |
| 7500                      | 1.334                | 1.365 | 1.396 | 1.441 | 1.500 |
| 8000                      | 1.366                | 1.410 | 1.456 | 1.513 | 1.583 |
| 8500                      | 1.399                | 1.456 | 1.516 | 1.584 | 1.666 |
| 9000                      | 1.430                | 1.502 | 1.574 | 1.657 | 1.749 |
| 9500                      | 1.462                | 1.546 | 1.632 | 1.727 | 1.830 |
| 10000                     | 1.494                | 1.591 | 1.689 | 1.794 | 1.909 |
| At 370° F.                |                      |       |       |       |       |
| 200                       | 1.003                | 0.993 | 0.978 | 0.953 | 0.919 |
| 400                       | 1.006                | 0.986 | 0.955 | 0.904 | 0.828 |
| 600                       | 1.009                | 0.981 | 0.934 | 0.855 | 0.730 |
| 800                       | 1.013                | 0.978 | 0.914 | 0.808 | 0.628 |
| 1000                      | 1.018                | 0.975 | 0.899 | 0.770 | 0.541 |
| 1250                      | 1.024                | 0.974 | 0.884 | 0.732 | 0.482 |
| 1500                      | 1.030                | 0.974 | 0.874 | 0.710 | 0.496 |
| 1750                      | 1.037                | 0.977 | 0.868 | 0.704 | 0.524 |
| 2000                      | 1.045                | 0.982 | 0.871 | 0.710 | 0.561 |
| 2250                      | 1.054                | 0.989 | 0.876 | 0.725 | 0.600 |
| 2500                      | 1.064                | 0.997 | 0.886 | 0.747 | 0.643 |
| 2750                      | 1.074                | 1.007 | 0.898 | 0.773 | 0.686 |
| 3000                      | 1.085                | 1.018 | 0.914 | 0.802 | 0.729 |

**TABLE II. COMPRESSIBILITY FACTORS FOR NITROGEN-BUTANE MIXTURES (Contd.)**

(Smoothed data)

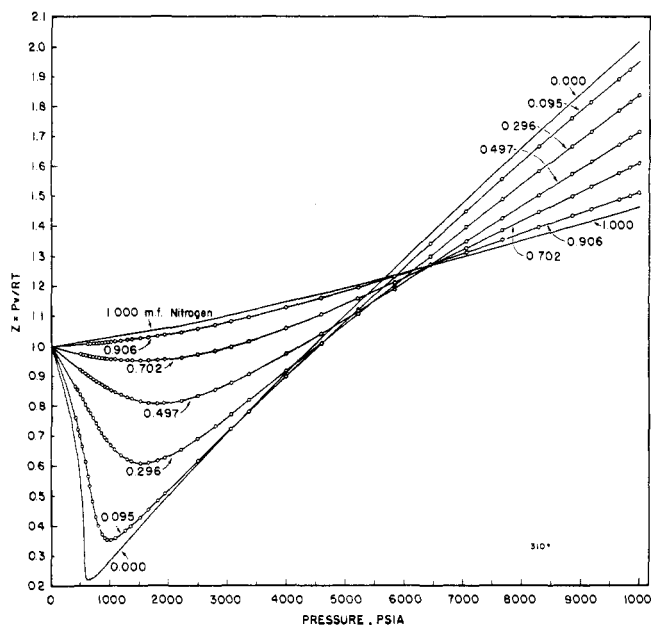
| Pressure,<br>Lb./Sq. Inch | Mole Fraction Butane |       |       |       |       |
|---------------------------|----------------------|-------|-------|-------|-------|
|                           | 0.100                | 0.300 | 0.500 | 0.700 | 0.900 |
| 3500                      | 1.106                | 1.046 | 0.954 | 0.865 | 0.815 |
| 4000                      | 1.129                | 1.076 | 1.000 | 0.934 | 0.902 |
| 4500                      | 1.154                | 1.109 | 1.051 | 1.002 | 0.986 |
| 5000                      | 1.180                | 1.146 | 1.101 | 1.072 | 1.069 |
| 5500                      | 1.207                | 1.186 | 1.158 | 1.144 | 1.152 |
| 6000                      | 1.236                | 1.228 | 1.215 | 1.215 | 1.236 |
| 6500                      | 1.265                | 1.270 | 1.272 | 1.286 | 1.319 |
| 7000                      | 1.295                | 1.314 | 1.328 | 1.356 | 1.400 |
| 7500                      | 1.326                | 1.357 | 1.385 | 1.425 | 1.481 |
| 8000                      | 1.357                | 1.400 | 1.441 | 1.494 | 1.561 |
| 8500                      | 1.388                | 1.443 | 1.498 | 1.563 | 1.640 |
| 9000                      | 1.419                | 1.487 | 1.555 | 1.632 | 1.721 |
| 9500                      | 1.450                | 1.529 | 1.610 | 1.700 | 1.800 |
| 10000                     | 1.481                | 1.572 | 1.665 | 1.766 | 1.877 |

At 400° F.

|       |       |       |       |       |       |
|-------|-------|-------|-------|-------|-------|
| 200   | 1.004 | 0.995 | 0.982 | 0.960 | 0.929 |
| 400   | 1.007 | 0.990 | 0.962 | 0.919 | 0.858 |
| 600   | 1.011 | 0.986 | 0.945 | 0.878 | 0.773 |
| 800   | 1.015 | 0.985 | 0.929 | 0.839 | 0.700 |
| 1000  | 1.020 | 0.984 | 0.916 | 0.806 | 0.630 |
| 1250  | 1.026 | 0.983 | 0.904 | 0.772 | 0.569 |
| 1500  | 1.032 | 0.985 | 0.896 | 0.752 | 0.538 |
| 1750  | 1.040 | 0.988 | 0.892 | 0.744 | 0.555 |
| 2000  | 1.048 | 0.993 | 0.893 | 0.747 | 0.583 |
| 2250  | 1.058 | 1.000 | 0.898 | 0.757 | 0.617 |
| 2500  | 1.067 | 1.009 | 0.908 | 0.774 | 0.655 |
| 2750  | 1.077 | 1.018 | 0.919 | 0.796 | 0.696 |
| 3000  | 1.087 | 1.029 | 0.933 | 0.821 | 0.740 |
| 3500  | 1.108 | 1.055 | 0.968 | 0.879 | 0.822 |
| 4000  | 1.130 | 1.084 | 1.009 | 0.943 | 0.905 |
| 4500  | 1.154 | 1.116 | 1.056 | 1.007 | 0.983 |
| 5000  | 1.179 | 1.149 | 1.103 | 1.071 | 1.064 |
| 5500  | 1.205 | 1.187 | 1.158 | 1.142 | 1.144 |
| 6000  | 1.232 | 1.227 | 1.213 | 1.210 | 1.226 |
| 6500  | 1.260 | 1.267 | 1.268 | 1.278 | 1.307 |
| 7000  | 1.289 | 1.308 | 1.322 | 1.345 | 1.387 |
| 7500  | 1.318 | 1.348 | 1.375 | 1.412 | 1.465 |
| 8000  | 1.348 | 1.389 | 1.429 | 1.480 | 1.541 |
| 8500  | 1.378 | 1.430 | 1.483 | 1.545 | 1.619 |
| 9000  | 1.408 | 1.471 | 1.537 | 1.610 | 1.695 |
| 9500  | 1.437 | 1.511 | 1.591 | 1.676 | 1.774 |
| 10000 | 1.467 | 1.552 | 1.643 | 1.742 | 1.848 |

In charging the nitrogen, the loading bomb was evacuated, filled with mercury from the mercury pump, and then filled with nitrogen as mercury was withdrawn from the loading bomb into the mercury pump by withdrawing the steel piston of the pump a predetermined number of turns. The volume of nitrogen in the loading bomb was determined over a range of pressures. The approximate quantity of nitrogen was then introduced into the compressibility bomb, which was held at liquid air temperatures to minimize danger of butane diffusion, by allowing nitrogen to flow into the equilibrium bomb sufficiently to cause a precalculated pressure drop. The precise quantity of nitrogen injected was determined by injecting mercury from the mercury pump into the loading bomb and recording the pump readings over the same pressure range as before injection.

The results, consisting of three series of mercury pump readings over a pressure range from 200 to 700 pounds per square inch absolute, were plotted on large graph paper. The number of moles of nitrogen charged were calculated from the volume of mercury injected into the loading bomb necessary to equalize the pressures before and after the injection. This volume was read from the



**Figure 1. Compressibility factors vs. pressure for nitrogen-n-butane mixtures at 310° F. at constant compositions**

graphs usually at 250 pounds per square inch absolute. The compressibility factor of nitrogen at 45° C. and 250 pounds per square inch (23) was used for this calculation. The line connecting the loading bomb to the metering valve of the equilibrium bomb formed part of the volume of the loading bomb. By this method all line corrections and volume corrections due to deformation of the system with pressure are included in the measurements and ultimately cancel out. The precision of this measurement was within 0.03% with respect to pressure and within  $\pm 0.02\%$  with respect to volume measurement. The molecular weight for nitrogen used in the calculations was 28.016.

Considering the precision of each variable, it may be said with considerable confidence that the over-all precision of the compressibility factors determined is within 0.5% at the lower pressures and within 1.0% at the highest pressures. The estimated precision is consistent with the maximum deviations observed on large scale cross plots of the experimental results.

## EXPERIMENTAL DATA

Tabulations of the experimental values obtained in this investigation are shown in Table I where the averages of the two experimental volume values obtained at each pressure are used.

For convenience, the experimental compressibility factors have been smoothed and are summarized at even values of pressure and of mole fraction of n-butane. The smoothed data are presented in Table II. The smoothing was done by means of large scale graphs of the types illustrated in Figures 1 and 2. The compressibility factors of the pure components were obtained from published tabulations (23).

The smoothness of the large scale plots indicates the data to be self-consistent. Consistency with the published values of the compressibility factors of the pure components (23) is also apparent. The root-mean-square deviation between the experimental points and the smooth curves was less than 0.5%.

## ACKNOWLEDGMENT

The purity of the gases used and some experimental calibrations of equipment were checked using some equip-

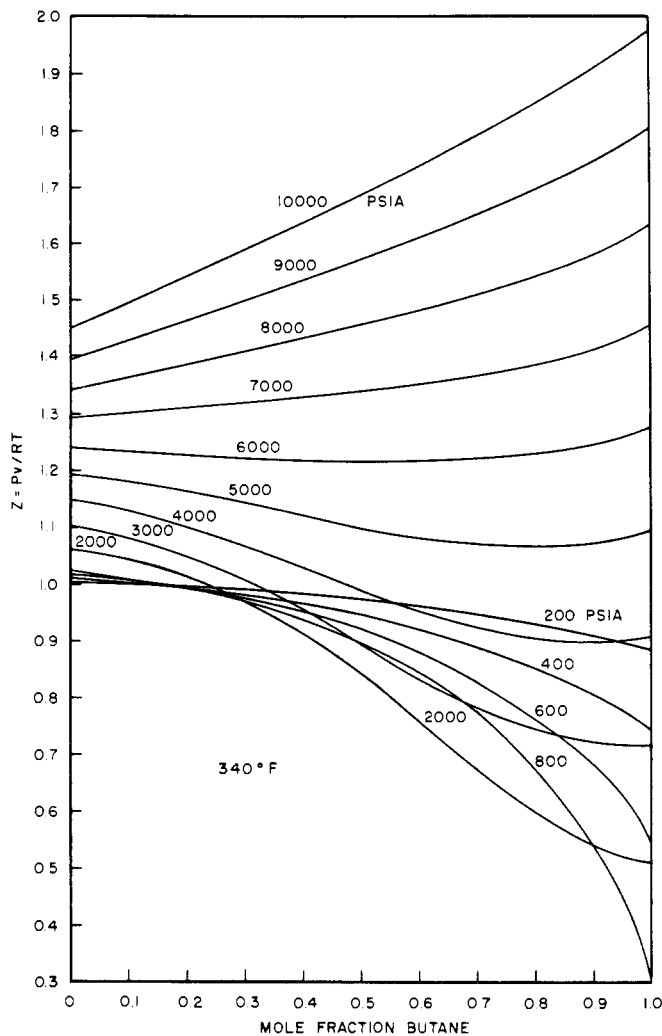


Figure 2. Compressibility factors vs. mole fraction of n-butane at constant pressures

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## Diffusion Coefficients in Hydrocarbon Systems Methane-n-Butane-Methane in Liquid Phase

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There is insufficient information about the diffusion coefficients of the lighter hydrocarbons in the liquid phase for predicting the nonequilibrium behavior of petroleum during production and refining operations.

By following methods established earlier, the Fick diffusion coefficient of methane in the liquid phase of the methane-n-butane system was determined at pressures up to nearly 2000 pounds/square inch in the temperature interval between 10° and 220° F. The results are presented as a function of the state of the phase.

These measurements indicate that the composition and temperature of the liquid phase have a significant influence on the Fick diffusion coefficient. The magnitude of variations in the coefficient within the range of temperatures and compositions investigated makes it necessary to take both of these factors into account in treating many physical situations of industrial interest.

#### INTRODUCTION

A knowledge of the molecular transport characteristics of the lighter hydrocarbons is of value in many operations associated with the production and refining of petroleum. Little experimental work is available in this field except the early measurements of Pomeroy (9) and of Lacey and coworkers (1,4,5,7). Kirkwood (6) has assembled the basic relations of transport and these have been applied to a number of situations of engineering interest (8). Drickamer made a number of investigations of transport in liquid and gas phases at elevated pressures, of which his studies of diffusion through an interface (18, 19) are given as examples. There has been increasing interest in resistances at interfaces (2) but, in the case of hydrocarbon systems involving transfer from a liquid to a gas phase (16), it appears that this resistance is not large. Such behavior is