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*Evaluation of Data Availability and Quality for Interaction
Second Virial Coefficients of Use to the Gas Industry*

*Bernard J. Van Wie, Mark A. Langenberg, Wayne C.-W. Chang, Kesavalu H. Kumar,
and Kenneth E. Starling*

National Bureau of Standards

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Foreword

The National Standard Reference Data System was established in 1963 for the purpose of promoting the critical evaluation and dissemination of numerical data of the physical sciences. The program is coordinated by the Office of Standard Reference Data of the National Bureau of Standards but involves the efforts of many groups in universities, government laboratories, and private industry. The primary aim of the program is to provide compilations of critically evaluated physical and chemical property data. These tables are published in the *Journal of Physical and Chemical Reference Data*, in the NSRDS-NBS series of the National Bureau of Standards, and through other appropriate channels.

A necessary preliminary step to the critical evaluation process is the retrieval from the world scientific literature of all papers reporting measurements on the property in question, followed by the extraction and organization of the numerical data contained in these papers. The present Technical Note summarizes the available data on interaction second virial coefficients for hydrocarbons and other compounds commonly present in natural and synthetic gas. We hope this summary will be useful to those engaged in correlation of such data and that it will stimulate new experimental measurements on systems for which the existing data are inadequate.

Further information on NSRDS and the publications and computerized databases which form the primary output of the program may be obtained by writing to the Office of Standard Reference Data, National Bureau of Standards, Gaithersburg, MD 20899.

DAVID R. LIDE, JR., *Director*
Office of Standard Reference Data

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Abstract

Binary interaction second virial coefficient information useful to the natural and syngas industries has been compiled and evaluated. An extensive literature search has been conducted to obtain publicly available information dating back to 1900. Assessments were made to determine data availability, quality and to summarize data needs. No evaluations were made for values resulting from thermodynamic correlations; however, references containing such information are listed. Each binary gas system has been individually evaluated and references containing the highest quality and most abundant measurements have been listed in a table of recommended values.

1. INTRODUCTION

As one of four participants in a Gas Research Institute funded project, it has been the task at the University of Oklahoma to assess the availability and quality of binary interaction second virial coefficient, B_{12} , data for the natural gas and syngas industries and to project needs for new data. This paper focuses on data availability and quality assessments. Suggestions for the procurement of new data are also made. Interaction second virial coefficients, B_{12} 's, can be used for describing real gas behavior of mixtures and for this reason are of importance to the natural gas and syngas industries. Not only are virial coefficients useful in describing PVT relationships through the virial form of the equation of state, but also in describing thermodynamic properties such as fugacities, internal energies, enthalpies, heat capacities, entropies, Joule-Thomson coefficients and gas phase excess properties. The capability for accurate prediction of such properties is important to the gas industries for correlation development and plant design.

Specific instances in which interaction second virial coefficients may be of greatest importance are in vapor-liquid equilibrium correlation development work where the component fugacities in the gas phase may be calculated using B_{12} data. These fugacities in turn are used to test activity coefficient correlations for the liquid phase, and in the development of mixture equations of state which have the theoretically correct composition dependence and are accurate in the lower density limit. Interaction second virial coefficients also are used to determine intermolecular potential function parameters; this application is of greater theoretical than industrial use. Little use has been made in the gas industry of interaction second virial coefficients at extremely low temperatures for two reasons: 1) the effect of higher virials becomes more important; and 2) there are fewer accurate interaction second virial coefficient data available due to the fact that the range of pressure over which one can secure data gets smaller due to condensation as temperature is decreased. Therefore, one must depend upon equations of state in order to obtain information about gas properties at temperatures where interaction second virial coefficients are less available.

In the past, industry was able to rely on lower quality data, because financial inequities in custody transfer and excess plant costs due to overdesign were not exorbitant.

However, in an age of diminishing natural resources and an ever tightening economy, industry is forced to be more conservative. Since accurate data are required for better thermodynamic correlations and improved process designs, a critical look at data availability, accuracy and needs is important.

It is also important to look at data for the syngas industries. Basically the design and operation of syngas plants and the storage and custody transfer of syngas requires the same kinds of data as are needed for the natural gas industries. However, many components are present in syngas which are either nonexistent or are only minor constituents in natural gas.

In many cases B_{12} data can be supplemented with B_{12} values calculated from existing PVT data. Non-virial equations of state (if available and accurate) can also be used to supply required properties information for the gas industries. However, where accurate PVT and B_{12} data are lacking (and at pressures which are low enough for a virial equation of state using only the second virial coefficient to be accurate) the measurement of new B_{12} data is of significant industrial importance.

It should also be noted that the significance of data quality is relative to the particular application for which the data are to be used. For instance, when gas properties are to be predicted for a gas of predominately one component, the contributions to bulk properties from unlike interactions with other components are minimal. Therefore, accurate B_{12} information is of little importance for bulk gas properties prediction for gases consisting of predominately one component. In such cases even low quality data or rough estimates of B_{12} 's with the use of moderately reliable correlations are probably adequate for properties predictions.

On the other hand accurate B_{12} information is critical in instances where one is interested in properties for gas processing such as in separations. Also accurate B_{12} information may be very important for custody transfer of gases containing large amounts of carbon dioxide or nitrogen and for the custody transfer of syngas. One must also be concerned with the pressure of the gas for which properties predictions are important. The contributions of unlike interactions are more significant at higher pressures and therefore higher quality data are more important at higher pressures. Also, one must be aware of the fact that the effect of higher virials becomes more important as pressure increases and therefore the existence of accurate B_{12} 's alone may not be sufficient. A specific case in which higher virials are important is in the design of compressors.

2. PROCEDURES FOR ASSESSMENT OF DATA AVAILABILITY, QUALITY AND NEEDS

2.1 Data Availability

A literature search was performed to locate and retrieve available interaction second virial coefficient information dating back to 1900 for all binaries of importance to the natural gas and syngas industries. Extensive bibliographical and data range information

were also available through a compilation by Dymond and Smith, *The Virial Coefficients of Pure Gases and Mixtures*,¹ and from supplementary material to a publication by Hayden and O'Connell appearing in *Ind. Eng. Chem. Proc. Des. Dev.*² An additional source by Warowny and Stecki³, *The Second Cross Virial Coefficients of Gaseous Mixtures*, is available and may be of use as a cross reference for investigators interested in doing their own assessments. However, this text contains little additional information for systems of interest in this paper. The compilation by Dymond and Smith contains information about uncertainties for some data sets which served as an aid in making evaluations of data reliability, and needs. However, none of the above references contain a critical comparison between the data from individual authors, nor do they discuss data needs. Hence, the present assessment is of great value.

As shown in Figure 1 the number of references for each binary system was placed in the grid on a triangular data availability chart so that a quick assessment of available data could be made. Blank grids in the data availability chart are representative of binary systems for which no data references were found. Where a blank grid appears for a binary system of high importance to the gas industry, a critical data need exists. In cases where a large number of references are indicated in a grid for a particular binary system, data needs are expected to be less critical, and the focus for that particular binary system can be shifted to determination of which of these references contain data of highest quality.

Binaries with the greatest abundance of references generally correspond to those binaries which are of greatest importance to the gas industry (ie.: binaries involving major constituents of pipeline quality gas and dehydrated sweet natural gas as it enters the processing plant). There is a large number of B_{12} data references involving hydrogen, in part due to the many industrial processes involving hydrogen. Helium binaries also have an abundance of references, which can be attributed in part to data needs for processes for recovery of helium from natural gas. Binaries of H_2S and H_2O , which also are of importance to the gas industry, have relatively fewer numbers of references, possibly because of difficulties in experimental measurements, since H_2S is poisonous and highly corrosive, and adsorption problems in H_2O systems cause difficulties in obtaining high accuracy virial coefficient (and/or PVT) data. Also relatively few references are available for a number of syngas binary systems.

2.2 Data Quality

In the present work, the rankings 'A', 'B', 'C' and 'X' in Table 1 were used in order to classify the available interaction second virial coefficient data or theoretical values of different investigators. An 'A' ranking corresponds to data which are accurate to within 5% or $5 \text{ cm}^3/\text{mol}$ and are of quality sufficient for process design correlation development. 'B' ranked data are those which have inaccuracies greater than 5% or $5 \text{ cm}^3/\text{mol}$, but less than 10% or $15 \text{ cm}^3/\text{mol}$. The 'B' ranked data are sufficient for design purposes. A 'C' ranking corresponds to data which are inaccurate by more than 10% or $15 \text{ cm}^3/\text{mol}$, and are less than adequate for most industrial uses. An 'X' ranking corresponds to B_{12} values which have been determined through the use of a correlation.

Table 1
Ranking Classification for Interaction Second Virial Coefficients

Rank	Uncertainty in B_{12} Data [E]
A	$[E] < 5\%$ or $5 \text{ cm}^3/\text{mol}$
B	5% or $5 \text{ cm}^3/\text{mol} < [E] < 10\%$ or $15 \text{ cm}^3/\text{mol}$
C	10% or $15 \text{ cm}^3/\text{mol} < [E]$
X	Correlated B_{12} Values

Several methods were employed to aid in establishing the category, 'A', 'B' or 'C', into which binary system data should be placed. An initial assessment of data quality was often possible through authors' statements concerning the accuracy of their data. Care must be taken not to rely entirely on an author's statements, however. Authors may have either over estimated or under estimated the quality of their data. Uncertainties in authors' statements were often resolved through the use of other accuracy assessment techniques. These techniques included an evaluation of the accuracy or precision expected from the experimental procedures for data measurement used by the various authors. The compilation of Dymond and Smith¹ contained some information regarding the assessment of experimental techniques. This proved helpful in some instances. A more reliable method for the determination of data quality was the comparison of plotted B_{12} data versus inverse temperature for the various sources reporting data for a given binary system. Such plots produced a nearly straight line for most of the high quality interaction second virial coefficient data considered in this work. Lower quality data were often revealed by scatter in the data or lack of consistency with expected trends of B_{12} versus $\frac{1}{T}$ (and/or the temperature derivative, $dB_{12}/d(\frac{1}{T})$). Also, where two or more sources report data which are in agreement (within 5% or $5 \text{ cm}^3/\text{mol}$, whichever is the greater) and showed little scatter, these data sets were often regarded as top quality and assigned an 'A' ranking. Where agreement of the data from one source was within 10% but not within 5% (or less than $15 \text{ cm}^3/\text{mol}$ but greater than $5 \text{ cm}^3/\text{mol}$) of that of a second source of data which are known to be accurate, the first data set was assigned a 'B' ranking. Data sets disagreeing with high quality data by more than 10% (or more than $15 \text{ cm}^3/\text{mol}$) or showing a large amount of scatter and/or inconsistencies in trends were assigned a 'C' ranking.

Measurements are scarce for some binaries and for some temperature ranges, making the task of data assessment from plotted data more difficult. In some instances assessments could be inferred from a particular author's reputation. The data of reliable authors were often given a high ranking even when other data were not available for comparison. On the other hand, several authors' data were determined to be consistently of lower than 'A' quality (through comparison plots and the amount of scatter in their data). Thus when data for these authors were encountered in other binary systems they were not normally weighted as heavily in comparisons for data assessment as the measurements made by authors determined to be more reliable.

In other instances where data are scarce or where there is significant disagreement between the data from different sources, assessments were aided through a graphical comparison of plots of these data with quality data for similar binary systems. For instance, one would expect similar trends for B_{12} versus $\frac{1}{T}$ (and for the temperature derivative,

$dB_{12}/d(\frac{1}{T})$) for all high quality data for binaries of paraffin hydrocarbons with other paraffin hydrocarbons. Whether data for a particular binary either agreed or disagreed with these general trends was helpful in determining data quality.

Statements with regard to the evaluation of data should be regarded with some caution. For instance, when the data from two sources receive an 'A' or 'B' ranking because the data show excellent agreement, it is still possible (although unlikely) that both have reported data which are substantially in error. Also, the fact that a particular author's data are found to be either unreliable or reliable for one or more binaries, does not mean that the same is necessarily true for all binary systems measured by that particular author. In addition, new evidence may arise which would alter data rankings. Therefore, in any case where extremely accurate data assessments are imperative, it is recommended that the original papers be consulted and independent assessments be performed. Nevertheless this work will serve as a valuable resource in making initial quality assessments of such data.

2.3 Data Needs

As shown in Figure 2 a triangular chart consisting of grids for each binary system was also used to summarize data needs. Where data are accurate and adequately cover the full range of data requirements for current or anticipated future gas industry needs, the letter 'S' was entered into the appropriate grid for the corresponding binary system indicating that data for this system are sufficient. In cases where existing data are insufficient for current or projected future needs, the procurement of new data is recommended. Data procurement is recommended at one of three priority levels. Priority level '1' is used for cases where new data are imperative (i.e.: in cases where procurement of such data is of highest priority to the gas industry); Priority level '2' is used for cases where new data are needed (i.e., instances where at least 'B' quality data are in existence but 'A' quality data are desired, or when it is desired to expand the range over which data are available, or when existing data are inadequate to meet anticipated high priority future needs); Priority level '3' is used for cases where new data are desired (i.e.: in cases where data ranges need to be expanded for lower priority systems or in cases where additional data are desired in order to clarify uncertainties in data rankings for binaries of moderate priority).

As evidenced in Figure 2 there is a direct relationship between the abundance of references and data needs. Where data are most abundant, such as for CH_4 binaries with C_2H_6 , C_3H_8 , $n\text{-C}_4\text{H}_{10}$, $n\text{-C}_5\text{H}_{12}$, N_2 and He , existing data are sufficient. In other cases, where the number of references are fewer for binary systems of importance to the gas industry, there is usually a corresponding data need. Where at least a few references are available for a particular binary of importance, the need for procurement of new data usually has been set as priority level '3' rather than priority level '2'. The lower priority levels are often an indication that new data are desired principally to verify data quality or to expand data ranges to other temperature regions.

The only imperative data need for new interaction second virial coefficients is for the $\text{CH}_4\text{-CO}$ binary, which is a high priority binary for the syngas industry. Another

major need is for the $\text{CH}_4\text{-CO}_2$ binary at priority level '2'; these new data are necessary to verify the quality of existing data. Since relatively few data are available for the $\text{CH}_4\text{-CO}_2$ binary (which is of importance to both the natural gas and syngas industries), further data procurement is recommended for this binary. Much of the needed B_{12} information for the $\text{CH}_4\text{-CO}_2$ binary may be derivable from available PVT data, however, and therefore procurement of new data is only recommended at priority level '2', rather than at priority level '1'. Other needs at priority levels '2' and '3' exist for natural gas binaries and for many of the syngas binaries of greater industrial importance. In cases where data are nonexistent for binaries of only very low industrial importance, no needs are specified.

3. SUMMARY OF DATA RANGES AND DATA QUALITY

Table 2 lists for each binary system the quality ranking and temperature ranges for interaction second virial coefficient data or theoretical values for the corresponding sources. A superscript '†' was used to indicate B_{12} data which were derived from information reported by another source. A superscript '‡' was used to indicate data sets for which data quality rankings are less certain or where data quality varies within a given set.

Table 3 lists the recommended B_{12} data over different temperature ranges for each binary system. The authors who measured the data and data rank are also listed. Where two or more authors have each reported quality information for the same system and over a similar temperature range one of two approaches was taken in recommending data: 1) both are listed when two or more authors have reported the same amount of data; 2) only one author is listed where one author has clearly measured more data points and/or data over a broader temperature range than other authors. Where only 'B' quality data exist over a certain temperature range for a certain binary system the higher quality and more abundant data are listed. However, it is not recommended that the 'B' data be used when extremely reliable data are needed. Systems for which only 'C' quality data are available do not appear in Table 3.

4. EVALUATIONS FOR INDIVIDUAL BINARY SYSTEMS DATA

Evaluations in this section are arranged by binary system following the same order as that used to organize the availability chart in Figure 1. For instance, methane binary evaluations appear first beginning with the $\text{CH}_4\text{-C}_2\text{H}_6$ system followed by the $\text{CH}_4\text{-C}_3\text{H}_8$ system and proceeding vertically down the availability chart. After the discussion of methane binaries, ethane binaries are discussed, followed by propane binaries and continuing by proceeding horizontally across the top of the availability chart with each successive group of binary systems.

Several sources have reported correlated interaction second virial coefficient values for several binary systems. For instance, Hayden and O'Connell² have reported correlated values for many binaries. However, these values usually showed significant deviations from experimental values. Correlated values more consistent with experimental data have been reported for several binary systems by Guggenheim and McGlashan;⁴ Huff and Reed;⁵

Vilcu et al.;⁶ Vilcu and Gainer;⁷ and Katayama et al.⁸ These correlated values were often not discussed in individual binary system evaluations since the plotting of these values would detract from the evaluations of the actual data. However, property ranges are given for the correlated values in Table 3. Data rank for the correlated values is indicated by an 'X' to distinguish them from actual experimental data and from values derived from experimental data.

4.1 Methane Binaries

4.1.1 CH₄-C₂H₆

Figure 3 shows a plot of all available B_{12} data for the CH₄-C₂H₆ binary in the temperature range of 215 to 373 K (-73 to 212° F; $\frac{1}{T}=0.0047$ to 0.0027). As exhibited in this plot, the data of Dantzler et al.,⁹ the data of Michels and Nederbragt¹⁰ the data of Wormald et al.,¹¹ the datum of Katayama et al.,⁸ the values reported by Gunn¹² and the datum reported by Mason and Eakin¹³ show little scatter and consistent general trends of B_{12} versus inverse absolute temperature. Because these data agree to within better than 5%, they are all regarded as being of 'A' quality. The 'A' ranking for these sources is supported by uncertainties reported in Dymond and Smith;¹ $\pm 2 \text{ cm}^3/\text{mol}$ for Dantzler et al., less than 5% for Wormald et al. and $\pm 5 \text{ cm}^3/\text{mol}$ for Mason and Eakin. It should be mentioned that the values of Gunn were actually derived from the PVT data of Michels. The reason Gunn's derived values do not coincide precisely with the data of Michels is that Gunn reported as his best values the average interaction second virial coefficients calculated for methane-ethane mixtures of varying compositions. It is the present authors' opinion that the interaction second virial coefficients derived from PVT data for a 50:50 mixture are more accurate and that Gunn should have taken weighted averages.

A smooth curve constructed from the data of Hoover et al.¹⁴ is in considerable disagreement with the general trends for B_{12} versus $\frac{1}{T}$ and the temperature derivative, $dB_{12}/d(\frac{1}{T})$ for the other six investigators. Therefore the data of Hoover et al. are given an overall ranking of 'C'. However, it should be noted that the 273 K datum of Hoover et al. is in good agreement with others' data. Also it is possible that the curvature expected in the B_{12} versus $\frac{1}{T}$ curve at lower temperatures may incorporate the lower temperature datum of Hoover et al. at 215 K.

4.1.2 CH₄-C₃H₈

Of the four sources Dantzler et al.⁹ (Dantzler et al. also reported values which were interpolated and extrapolated from the data of Gunn); Gunn¹² (Gunn's data were derived from the PVT data of Sage and Lacey¹⁵); Mason and Eakin;¹³ and Wormald et al.¹¹ who have reported B_{12} information for the CH₄-C₃H₈ binary system, the data for the 245 to 511 K (-18.7 to 460° F; $\frac{1}{T}=0.0041$ to 0.002) temperature range generally agree within 2% as exhibited in Figure 4. Exceptions to this good agreement occurs at 373 K (211.7° F; $\frac{1}{T}=0.0027$) where the datum of Dantzler et al. disagrees with the values of Gunn by 6

cm^3/mol , and at 298 K (77° F; $\frac{1}{T}=0.0033$) where the datum of Dantzler et al. disagrees with a curve constructed from the data of Wormald et al. by about 6%. (Note that an extrapolated value from the data of Gunn is in better agreement with the datum of Dantzler et al. at 298 K, but the agreement of extrapolated values must be regarded with caution because of uncertainties which are inherent in extrapolation techniques.)

Gunn's data at (about) 373 K and Wormald's datum at (about) 298 K are more accurate than Dantzler's datum at 373 K and at 298 K respectively because Dantzler's datum deviates from the smooth curve which would result from considering the data of Wormald et al. and Gunn collectively. The smooth curve constructed from the combined data of Gunn and Wormald et al. exhibits the same trends for B_{12} versus $\frac{1}{T}$ (and for the temperature derivative, $dB_{12}/d(\frac{1}{T})$) as other binaries of methane with paraffin hydrocarbons. The temperature derivative, $dB_{12}/d(\frac{1}{T})$, of Dantzler's data disagrees with that of Gunn's and Wormald's data. However, since Dantzler's data cross the curve which could be constructed from Gunn's and Wormald's combined data sets, there is good agreement (within 2%) for Dantzler's interior data points.

On the basis of these considerations, the $\text{CH}_4\text{-C}_3\text{H}_8$ data of Wormald et al., the values of Gunn and the datum of Mason and Eakin are of 'A' quality, while the data of Dantzler et al. are of 'B' quality. The values which were interpolated and extrapolated from the data of Gunn are given an 'X' ranking since these have not been experimentally determined.

4.1.3 $\text{CH}_4\text{-n-C}_4\text{H}_{10}$

Of five sources reporting data for the $\text{CH}_4\text{-n-C}_4\text{H}_{10}$ binary (see Figure 5) over the temperature range of 277 to 583 K (38.9 to 595° F; $\frac{1}{T}=0.0036$ to 0.0017), the high temperature data of Gunn¹² (the high temperature data of Gunn designated by a triangle in Figure 5 were derived from the PVT data of Tang,¹⁶); the datum of Mason and Eakin,¹³ and the data of Wormald et al.¹¹ are considered more reliable than the data reported by Beattie and Stockmayer,¹⁷ and Dantzler et al.⁹ The reliability of Gunn's data, the datum of Mason and Eakin and the data of Wormald is based on the following: 1) the internal consistency of these data (with the exception of the 327 K ($\frac{1}{T}=0.0031$) and 314 K ($\frac{1}{T}=0.0032$) data of Wormald et al.); 2) the reliability of the data from these authors for other similar alkane binaries (see Table 2); 3) the agreement of these data with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative ($dB_{12}/d(\frac{1}{T})$)) for the reliable data of other similar binary alkane systems as shown in Figure 6, namely $\text{CH}_4\text{-C}_2\text{H}_6$, $\text{CH}_4\text{-C}_3\text{H}_8$, $\text{CH}_4\text{-i-C}_4\text{H}_{10}$, $\text{CH}_4\text{-n-C}_5\text{H}_{12}$ and $\text{CH}_4\text{-neo-C}_5\text{H}_{12}$; and 4) Gunn and Wormald et al. are in agreement at (about) 374 K ($\frac{1}{T}=0.027$), and Mason and Eakin, and Wormald are in agreement at (about) 289 K ($\frac{1}{T}=0.0035$).

It should be mentioned that Wormald et al. who derived their data from excess enthalpies, report uncertainties as large as 23%. This may explain the 314 and 327 K data points which deviate considerably from a smooth curve constructed from other reliable data. The remaining data of Wormald et al., however, are considered reliable for the reasons stated above.

On the other hand, the data of Dantzler et al., disagree with a curve constructed from the combined data of Gunn, of Wormald et al. and of Mason and Eakin by as much as 10%. Also the temperature derivative $dB_{12}/d(\frac{1}{T})$ for a curve constructed from the data of Dantzler et al. disagrees with a similar curve constructed from considering collectively the data of Gunn, of Wormald et al. and of Mason and Eakin. Further evidence for the lower quality ranking comes from an observation in Figure 6 where it is shown that a curve constructed from the the data of Dantzler et al. is approaching one that could be constructed from quality methane–neopentane binary data as temperature is decreased (as one moves toward larger values of $\frac{1}{T}$). For all other systems in the present investigation the general trends for quality data show a divergence as temperature is lowered for similar binary systems. It should be mentioned that the higher temperature data of Dantzler et al. appear to be in better agreement with other methane–n-butane binary data and are, therefore, regarded as being of better quality. All the data of Dantzler et al. appear to be in good agreement with interpolated and extrapolated values from Gunn's data. However, since Dantzler et al. performed the interpolations and extrapolations, these values may be biased toward the data of Dantzler et al. Therefore, such values are not regarded highly in the evaluation process.

The data of Beattie and Stockmayer not only exhibit a dissimilar trend from a smooth curve constructed from other data, but also exhibit inconsistencies with the general trends of the data for other methane paraffin binaries shown in Figure 6. For instance, Figure 6 reveals that general trends for methane binary data show a decrease in values for binary interaction second virial coefficients as one looks at data for methane binaries with paraffin hydrocarbons of increasing chain length. One would therefore expect decreasing values for B_{12} 's for methane–paraffin hydrocarbon binaries in the following order: $\text{CH}_4\text{-C}_2\text{H}_6$, $\text{CH}_4\text{-C}_3\text{H}_8$, $\text{CH}_4\text{-i-C}_4\text{H}_{10}$, $\text{CH}_4\text{-n-C}_4\text{H}_{10}$, $\text{CH}_4\text{-neo-C}_5\text{H}_{12}$ and $\text{CH}_4\text{-n-C}_5\text{H}_{12}$. Contrary to the general trend expected, the data of Beattie and Stockmayer for the $\text{CH}_4\text{-n-C}_4\text{H}_{10}$ binary have lower values than quality data at corresponding temperatures for methane binaries with neopentane and n-pentane as shown in Figures 7.

Based on the above considerations the data of Gunn, the datum of Mason and Eakin and the data of Wormald et al. are ranked as 'A' quality; the data of Beattie and Stockmayer which deviate from Gunn's data by as much as $9.6 \text{ cm}^3/\text{mol}$ and the data of Dantzler et al. are ranked as 'B' quality; and the values extrapolated and interpolated from the data of Gunn by Dantzler et al. are given an 'X' ranking. It should be mentioned that although the overall ranking of the data of Beattie, and of Dantzler et al. is 'B' the higher temperature data of Dantzler et al. and the higher temperature data of Beattie et al. are in agreement with the data of Gunn within 5% as shown in Figure 5 and therefore these particular data points are of 'A' quality.

4.1.4 $\text{CH}_4\text{-i-C}_4\text{H}_{10}$

The interaction second virial coefficient data of Olds et al.¹⁸ and data of Gunn¹² for the $\text{CH}_4\text{-i-C}_4\text{H}_{10}$ binary agree within 5% over the 344 to 511 K (159.5 to 460.1° F; $\frac{1}{T}=0.0029$ to 0.0020) temperature range as exhibited in Figure 8. The data of Olds et al.¹⁸ and the data of Gunn¹² for the $\text{CH}_4\text{-i-C}_4\text{H}_{10}$ are considered to be of high quality based on: 1) the internal consistency of these data; 2) the agreement of these data with the

general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative $dB_{12}/d(\frac{1}{T})$) for methane binaries with paraffin hydrocarbons as shown in Figure 9; and 3) the fact that B_{12} data derived from PVT data from the laboratory where Olds measured his data (see Sage and Lacey (1959)¹⁹) have been shown to be of good quality in other instances. While the B_{12} 's of Gunn and of Olds et al. agree with the general trends exhibited for methane binaries with other paraffin hydrocarbons, as shown in Figure 9, the B_{12} datum at 288 K (58.7° F; $\frac{1}{T}=0.0035$) of Mason and Eakin¹³ deviates significantly from what one would expect for the CH₄-i-C₄H₁₀ B_{12} value at this temperature.

On the basis of the above considerations Gunn's data and the data of Olds et al. are ranked as 'A' quality, and the datum of Mason and Eakin as 'C' quality. It should also be mentioned that where CH₄-i-C₄H₁₀ binary data are needed, these data can be closely approximated by CH₄-n-C₄H₁₀ binary data as shown in Figure 9.

4.1.5 CH₄-n-C₅H₁₂

Comparisons are made in Figure 10 for the CH₄-n-C₅H₁₂ data reported by several sources (continuous lines join the data points of individual sources) with the high quality data for other methane-paraffin hydrocarbon binary systems. General trends for all CH₄-paraffin hydrocarbon system data show a convergence at higher temperatures (lower values of $\frac{1}{T}$) for reliable data. Deviations from this general trend for sources reporting data for the CH₄-n-C₅H₁₂ binary reveal data sets which are of lower quality. A close comparison of Figure 10 with the CH₄-n-C₅H₁₂ data plotted in Figure 11, reveals that the data which deviate more significantly from expected convergence trends are those of Massoudi and King²⁰ and those of Pecsok and Windsor.²¹ Data showing less significant deviations are those of Wormald et al.¹¹ and those of Dantzler et al.⁹ The data of Zaalishvili²² show good agreement with the general trends observed for other methane-paraffin hydrocarbon systems.

The data of Zaalishvili are ranked as 'A' quality for the following reasons: 1) Zaalishvili's data agree with the general trends of B_{12} versus $\frac{1}{T}$ and convergence of B_{12} values at high temperatures as shown for other methane-paraffin hydrocarbon binaries; 2) the data of Zaalishvili are internally consistent; and 3) the data of Zaalishvili have been shown to be of good quality for other binaries in the present study (see Table 3). The data of Dantzler et al. and the data of Wormald et al. are ranked as 'B' quality while the data of Pecsok and Windsor and the datum of Massoudi are ranked as 'C' quality for the following reasons: 1) these authors have reported data which disagree with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative $dB_{12}/d(\frac{1}{T})$) for other methane-paraffin hydrocarbon binaries; 2) these authors have reported values which disagree with a smooth curve constructed from the reliable data of Zaalishvili by more than 5% for the 'B' quality data and more than 10% for the 'C' quality data; and 3) these authors have reported data with large uncertainties,¹ ± 42 cm³/mol for the data of Pecsok and Windsor, ± 13 cm³/mol for the data of Dantzler et al., ± 12 cm³/mol for the data of Massoudi and King, and ± 20 cm³/mol for the data of Wormald. The datum reported by Mason and Eakin¹³ at 298.15 K (77° F; $\frac{1}{T}=0.0034$) is considered to of 'C' quality because the datum has a reported uncertainty of ± 11 cm³/mol.

4.1.6 CH₄-i-C₅H₁₂

As shown in Figure 12, Mason and Eakin,¹³ and Pecsok and Windsor²¹ reported data for the CH₄-i-C₅H₁₂ binary system. The B_{12} data of Pecsok and Windsor for the CH₄-i-C₅H₁₂ binary at 298 K (77° F) and 323 K (122° F) are of 'C' quality because a smooth curve which can be constructed by considering these data is in disagreement with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for methane binaries with other paraffin hydrocarbons, and the general trend of decreasing B_{12} values for i-C₅H₁₂ binaries with other paraffin hydrocarbons of increasing chain length as shown in Figure 13. The datum of Mason and Eakin¹³ at 289 K (61° F) is considered to be of 'A' quality ranking despite the lack of other CH₄-i-C₅H₁₂ binary data with which to compare at a similar temperature. The reason for this assessment is because of the agreement of the datum of Mason and Eakin with the general trends for other methane-paraffin hydrocarbon binaries, and the reported uncertainty of $\pm 5 \text{ cm}^3/\text{mol}$ for this datum.

4.1.7 CH₄-neo-C₅H₁₂

For the CH₄-neo-C₅H₁₂ system the Data of Strein et al.,²³ having a reported accuracy of $\pm 1\%$ and good internal consistency as shown in Figure 14, are considered reliable. Further verification of the accuracy of the data of Strein et al. comes from the agreement of the general trends of this data with the trends shown for the quality data of the CH₄-n-C₄H₁₀, CH₄-i-C₄H₁₀ and CH₄-n-C₅H₁₂ binary systems as shown in Figure 15. On the other hand, the data of Hamann et al.²⁴ and the data of Bellm et al.²⁵ disagree significantly with the general trends exhibited by the other binaries in that the data of Hamann et al. and of Bellm et al. do not converge with the other data at higher temperatures (lower values of $\frac{1}{T}$). Therefore, the data of Hamann et al. and of Bellm et al. are considered to be of lower quality. Further evidence that the data of Bellm et al. are of low quality is that these data received a low quality ranking by Dymond and Smith.¹ Also, the data of Hamann et al. show a large amount of scatter which is an indication that these data are in error. In addition, the data of Bellm et al. and of Hamann et al. deviate by as much as 41% and 11% respectively from the data of Strein et al.

The lower temperature data of Baughman et al.²⁶ have a reported uncertainty of 10% and have a significantly different slope from that of an extrapolation of the data of Strein to lower temperatures. Therefore, these data are considered to be of lower quality.

Based on the above considerations the data of Strein et al. are ranked as 'A' quality and the data of Bellm et al., of Hamann et al. and of Baughman et al. are ranked as 'C' quality.

4.1.8 CH₄-CO

The single available datum of Mason and Eakin¹³ at 289 K (61° F; $\frac{1}{T}=0.0035$) for the CH₄-CO binary is considered to be of 'A' quality based on the reported accuracy of $\pm 5 \text{ cm}^3/\text{mol}$ and the fact that in most cases the data of Mason and Eakin have been proven to be of 'A' quality when reported to have an accuracy of $\pm 5 \text{ cm}^3/\text{mol}$.

4.1.9 CH₄-CO₂

Three sources report B_{12} information for the CH₄-CO₂ binary system covering the 289 to 511 K (100 to 460° F; $\frac{1}{T}$ =0.0032 to 0.002) temperature range. These are: Zaalishvili,²² Katayama et al.⁸ and Mason and Eakin.¹³ The data of Zaalishvili are considered reliable for the following reasons: 1) the internal consistency of Zaalishvili's data as shown in Figure 16; 2) the internal consistency of Zaalishvili's data for other binary systems for which Zaalishvili measured data (ie. the C₂H₆-CO₂ and the CH₄-n-C₅H₁₂ binary systems); and 3) the agreement of the general trends for B_{12} versus $\frac{1}{T}$ (and the temperature derivative $dB_{12}/d(\frac{1}{T})$) between Zaalishvili's CH₄-CO₂ data and the data for other CO₂ binary systems with paraffin hydrocarbons as shown in Figure 17.

The datum of Katayama et al. and the datum of Mason and Eakin are also considered to be of good quality because these data agree with an extrapolation of Zaalishvili's data to lower temperatures (larger values of $\frac{1}{T}$) within 5 cm³/mol, and have reported accuracies of ±0.4 and ±5 cm³/mol respectively. On the basis of the above considerations, Zaalishvili's data, the datum of Katayama et al. and the datum of Mason and Eakin all are given 'A' rankings.

4.1.10 CH₄-N₂

The data Roe and Saville²⁷ for the CH₄-N₂ binary system over the 155 to 292 K (-181 to 66° F; $\frac{1}{T}$ =0.0065 to 0.0034) temperature range are considered to be extremely reliable for the following reasons: 1) the reported accuracy of Roe's data is within 2%; 2) a plot of Roe's data in Figure 18 reveals that the data are internally consistent; and 3) a comparison of Roe's data with the reliable data for similar nitrogen-paraffin hydrocarbon systems in Figure 19 reveals that all three of these systems show similar trends of convergence of B_{12} values at higher temperatures. Convergence of B_{12} values at higher temperatures for similar systems is a common trend as is shown for other similar binary systems discussed in other sections of this report. Roe's data have, therefore, been given a quality ranking of 'A'.

The datum of Mason and Eakin¹³ at 289 K (60.53; $\frac{1}{T}$ =0.0035) deviates from the datum of Roe at a similar temperature of 292 K (66° F; $\frac{1}{T}$ =0.0034) by 2.3 cm³/mol and, therefore, is of the same quality as the data of Roe. The datum of Mason and Eakin is, therefore, given a quality ranking of 'A'.

4.1.11 CH₄-H₂

Interaction second virial coefficient data reported by Mueller et al.,²⁸ the datum of Mason and Eakin,¹³ and the datum reported by Vilcu et al.⁶ for the CH₄-H₂ binary covering the 144 to 298 K (-200 to 77° F; $\frac{1}{T}$ =0.0069 to 0.0034) temperature range as shown in Figure 20 are ranked as 'A' quality. The 'A' ranking for the data of Mueller et al. and the datum of Vilcu et al. is based on the internal consistency of the plotted data as shown in Figure 20 and on the fact that the data of Mueller et al. were reported by

Dymond and Smith¹ to be accurate within $1 \text{ cm}^3/\text{mol}$. The datum of Mason and Eakin is ranked as 'A' quality since it agrees with an extrapolation of a smooth curve constructed from the data of Mueller et al. within $2 \text{ cm}^3/\text{mole}$. Correlated values of Vilcu et al. at 298 K (77°F ; $\frac{1}{T}=0.0034$) are in good agreement with a smooth curve constructed from the data of Mueller et al. and datum of Vilcu et al. collectively. Much of the needed higher temperature values can be derived from PVT measurements of Solbrig and Ellington²⁹ which cover the 138 to 423 K temperature range.

4.1.12 $\text{CH}_4\text{-H}_2\text{O}$

The single available data set of Rigby et al.³⁰ for the $\text{CH}_4\text{-H}_2\text{O}$ binary system covering the 298 to 511 K (77 to 460°F ; $\frac{1}{T}=0.0034$ to 0.002) temperature region as shown in Figure 21 is ranked as 'B' quality based on the following: 1) these data have a reported uncertainty of $\pm 15 \text{ cm}^3/\text{mol}$;¹ and 2) although there is scatter in the data, all points deviate by less than about $5 \text{ cm}^3/\text{mol}$ from a smooth curve which can be constructed from the data.

4.1.13 $\text{CH}_4\text{-H}_2\text{S}$

There are no available interaction second virial coefficient values for the $\text{CH}_4\text{-H}_2\text{S}$ binary; however, the volumetric data of Reamer et al.,³¹ covering the 280 to 400 K temperature range, may be used in deriving interaction second virial coefficients of accuracy sufficient for design purposes.

4.1.14 $\text{CH}_4\text{-He}$

The single available interaction second virial coefficient data set of Bell and Dunlop³² for the $\text{CH}_4\text{-He}$ system covering the 292 to 320 K (66 to 116°F ; $\frac{1}{T}=0.0034\text{-}0.0031$) temperature range is plotted in Figure 22. Although the data of Bell and Dunlop are sparse and there are no other $\text{CH}_4\text{-He}$ binary data with which to compare, the data are considered to be of 'A' quality based on the following reasons: 1) Bell and Dunlop report an uncertainty for their data of $\pm 0.3 \text{ cm}^3/\text{mol}$; and 2) other measurements made by Bell and Dunlop for the $\text{N}_2\text{-He}$ binary system compare well with other quality data for the $\text{N}_2\text{-He}$ binary system (see discussion for the $\text{N}_2\text{-He}$ binary system data).

4.1.15 $\text{CH}_4\text{-NH}_3$

The only available information for $\text{CH}_4\text{-NH}_3$ binary system are the smoothed values of Lee et al.³³ covering the 423 to 573 K (302 to 572°F ; $\frac{1}{T}=0.0024$ to 0.0017) temperature range as shown in Figure 23. These values are considered to be at least of 'B' quality since the data of Lee et al. for both the $\text{CH}_4\text{-NH}_3$ and the $\text{N}_2\text{-NH}_3$ binaries are internally consistent. The procurement of additional data may be helpful in verifying the quality of these data.

4.2 Ethane Binaries

4.2.1 $C_2H_6-C_3H_8$

Two sources, Dantzler et al.⁹ and Mason and Eakin,¹³ have reported $C_2H_6-C_3H_8$ interaction second virial coefficients covering the 288 to 373 K (59 to 212° F; $\frac{1}{T}=0.0035$ to 0.0027) temperature range. The datum of Mason and Eakin is in good agreement with the data of Dantzler et al. deviating from an extrapolation of the data of Dantzler et al. by only 3% as shown in Figure 24. The data for these two investigators are considered to be of 'A' quality for the following reasons: 1) the datum of Mason and Eakin is in good agreement with an extrapolation of the data of Dantzler et al.; 2) the data of Dantzler et al. are internally consistent for the $C_2H_6-C_3H_8$ binary as well as for all of the other binary systems of concern in the present study (see discussions for the following binary systems: $CH_4-C_2H_6$, $CH_4-C_3H_8$, $CH_4-n-C_4H_{10}$, $CH_4-n-C_5H_{12}$, $C_2H_6-C_3H_8$, $C_2H_6-n-C_5H_{12}$); 3) the data of Dantzler et al. have been shown to be of good quality for several other systems of concern in the present study (see Table 2); and 4) a smooth curve constructed from considering the data of Dantzler et al. and the datum of Mason and Eakin collectively agrees with the general trends of B_{12} versus $\frac{1}{T}$ (as well as for the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for other ethane-paraffin hydrocarbon binaries, and with the general trends of decreasing B_{12} values for interactions of ethane with paraffin-hydrocarbons of increasing chain length as shown in Figure 25.

4.2.2 $C_2H_6-n-C_4H_{10}$

Of three sources reporting B_{12} information for the $C_2H_6-n-C_4H_{10}$ binary system, the data of Dantzler et al.⁹ and the datum of Mason and Eakin¹³ covering the 288 to 373 K (59 to 212° F; $\frac{1}{T}=0.0035$ to 0.0027) temperature range are of better overall quality than the data of Wormald et al.¹¹ which exist over the same temperature range. The reliability of the data of Dantzler et al. and the datum of Mason and Eakin is based on the following reasons: 1) the data of Dantzler et al. as shown in Figure 26 are internally consistent. The datum of Mason and Eakin is consistent with an extrapolation of the data of Dantzler down to lower temperatures; 2) the data of Dantzler et al. have been shown to be of good quality for several other systems considered in the present study (see Table 2); and 3) a smooth curve constructed from considering the data of Dantzler et al. and the datum of Mason and Eakin collectively agrees with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for other ethane-paraffin hydrocarbon binary systems, and with the general trend of decreasing values for B_{12} for ethane binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 27. Based on the above the data of Dantzler et al. and the datum of Mason and Eakin are considered to be of 'A' quality.

One of the three points reported by Wormald et al., the datum at 333 K (140° F; $\frac{1}{T}=0.003$), shows a 16% deviation from a smooth curve constructed by considering the data of Dantzler et al. and the datum of Mason and Eakin collectively. Therefore, this

datum is of 'C' quality. However, the datum at 304.5 K (88° F; $\frac{1}{T}=0.0033$) deviates from the smooth curve by only 4% while the datum at 363 K (194° F; $\frac{1}{T}=0.0028$) falls on the curve. Therefore, these two points are of 'A' quality.

4.2.3 C₂H₆-n-C₅H₁₂

Three sources, Dantzler et al.;⁹ Massoudi and King;²⁰ and Pecsok and Windsor,²¹ report data for the C₂H₆-n-C₅H₁₂ binary covering the temperature range of 288 to 373 K (59 to 212° F; $\frac{1}{T}=0.0035$ to 0.0027). The data of Dantzler et al. are ranked as 'A' quality for the following reasons: 1) the data of Dantzler et al. are internally consistent as shown in Figure 28; 2) the data of Dantzler et al. have been shown to be of good quality for several other binary systems considered in the present study (See Table 2); and 3) the data of Dantzler et al. are in agreement with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative $dB_{12}/d(\frac{1}{T})$) and the general trend of decreasing B_{12} values for ethane binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 29.

The datum of Massoudi and King and the datum of Pecsok and Windsor at 298 K (77° F; $\frac{1}{T}=0.0034$), on the other hand, are ranked as 'C' quality for the following reasons: 1) the datum of Massoudi and King has a reported accuracy of about 8% and deviates from the smooth curve constructed from the data of Dantzler et al. by about 14%; 2) the data of Massoudi and King have been shown to be of low quality for the n-C₅H₁₂-CO₂ and the n-C₅H₁₂-N₂ binary systems; and 3) the datum of Pecsok and Windsor has a reported error of about $\pm 41\%$ ¹ and is in disagreement with the 298 K datum of Dantzler et al. by about 8%.

4.2.4 C₂H₆-CO₂

The data of Zaalishvili²² for the C₂H₆-CO₂ binary system which cover the temperature range of 311 to 500 K (100 to 460° F; $\frac{1}{T}=0.0032$ to 0.002) are given an 'A' quality ranking for the following reasons: 1) Zaalishvili's data show good internal consistency for the C₂H₆-CO₂ binary data as shown in Figure 30; 2) there is good internal consistency for other binary data which Zaalishvili has measured (ie., for the CH₄-CO₂ and CH₄-n-C₅H₁₂ binary systems); 3) Zaalishvili's C₂H₆-CO₂ interaction second virial coefficient data show consistent trends for B_{12} versus $\frac{1}{T}$ (and for the temperature derivative $dB_{12}/d(\frac{1}{T})$) with those of other reliable paraffin hydrocarbon-carbon dioxide data as shown in Figure 31.

It should be mentioned that the general trend for the second derivative of Zaalishvili's data, $d^2B_{12}/d(\frac{1}{T})^2$, is not in agreement with the general trends for the second derivatives for other paraffin hydrocarbon-carbon dioxide binaries. The second derivative normally is negative as indicated by a curve which is slightly concave downward. This is not the case for Zaalishvili's data as shown in Figure 31. However, when considering a smooth curve which could be constructed by considering the datum of Katayama et al.⁸ at 298 K (77° F; $\frac{1}{T}=0.0034$) the datum of Mason and Eakin¹³ at 289 K (61° F; $\frac{1}{T}=0.0035$) and the data of Zaalishvili collectively, none of the points deviate from the curve by more than 5%.

The datum of Katayama et al. and the datum of Mason and Eakin show agreement with each other and with the general trends of B_{12} versus $\frac{1}{T}$ expected for the C_2H_6 - CO_2 binary when comparisons are made with other paraffin hydrocarbon-carbon dioxide binary systems in Figure 31. Also, the reported accuracies of the datum of Katayama et al. and the datum of Mason and Eakin of ± 7 and $\pm 5 \text{ cm}^3/\text{mol}$ respectively indicate these data are of high quality and therefore are given an 'A' ranking.

4.2.5 $C_2H_6-N_2$

For the two sources, Gunn¹² (Gunn's data are derived from the PVT data of Sage and Lacey¹⁹); and Mason and Eakin,¹³ reporting B_{12} information covering the 277 to 511 K (39 to 460°F; $\frac{1}{T}=0.0036$ to 0.02) temperature range for the $C_2H_6-N_2$ binary system, the data generally agree as shown in Figure 32. An exception to this agreement is Gunn's datum at 311 K (100°F; $\frac{1}{T}=0.0032$) which deviates by about 6 cm^3/mol from a curve which could be constructed by considering the datum of Mason and Eakin and the data of Gunn collectively.

It is possible, of course, that Gunn's datum at 311 K is more accurate while the two surrounding data of Gunn and the datum of Mason and Eakin are less accurate. However, it is more reasonable to assume that only Gunn's 311 K datum is inaccurate because of the good agreement of the remaining values with a curve which could be constructed through these values. All of the values do, however, agree with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for the similar binary systems CH_4-N_2 and $n-C_4H_{10}-N_2$ as shown in Figure 33. The agreement with these general trends combined with the fact that Gunn's data and the data of Mason and Eakin have been determined to be reliable for other binary systems, indicates that Gunn's values and the datum of Mason and Eakin are of 'A' quality.

4.2.6 $C_2H_6-H_2$

The datum of Mason and Eakin¹³ at 289 K (61°F; $\frac{1}{T}=0.0035$) is the only available interaction second virial coefficient information for the $C_2H_6-H_2$ binary. The datum of Mason and Eakin is considered to be of 'A' quality because the datum has a reported uncertainty of $\pm 5 \text{ cm}^3/\text{mol}$ and the majority of the data of Mason and Eakin in this report have been shown to be of good quality. Additional interaction second virial coefficient data may be useful in the future for the $C_2H_6-H_2$ binary system for situations where syngas is mixed with natural gas. However, at present such information is of low priority to the gas industry and needed information can be derived from PVT data or calculated from available correlations.

4.2.7 $C_2H_6-H_2O$

Two sources, Coan and King;³⁴ and Rigby et al.,³⁰ report interaction second virial coefficients for the $C_2H_6-H_2O$ binary system covering the temperature range of 298 to 444

K (77 to 340° F ; $\frac{1}{T}=0.0034-0.0023$) as shown in Figure 34. Both data sets are in disagreement with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for the data set of Rigby et al.³⁰ for the $\text{CH}_4\text{-H}_2\text{O}$ binary (see Figure 35). Therefore, the existing $\text{C}_2\text{H}_6\text{-H}_2\text{O}$ data is considered to be of 'C' quality overall. Individual points may be of better than 'C' quality, but without additional data it is difficult to determine which are more accurate.

4.2.8 $\text{C}_2\text{H}_6\text{-H}_2\text{S}$

The interaction second virial coefficient data of Khoury and Robinson³⁵ covering the 323 to 398 K (122 to 257° F ; $\frac{1}{T}=0.0031$ to 0.0025) temperature range as shown in Figure 36 are the only available interaction second virial coefficient data for the $\text{C}_2\text{H}_6\text{-H}_2\text{S}$ binary system. The data of Khoury and Robinson are internally consistent and therefore are considered to be of at least of 'B' quality. However, since there are no other interaction second virial coefficient values with which to compare for the $\text{C}_2\text{H}_6\text{-H}_2\text{S}$ binary system or for other H_2S binaries with paraffin hydrocarbons, assessment of the quality of the data of Khoury and Robinson is difficult and the 'B' quality ranking is somewhat uncertain.

4.3 Propane Binaries

4.3.1 $\text{C}_3\text{H}_8\text{-n-C}_4\text{H}_{10}$

Two sources, Dantzler et al.⁹ and Mason and Eakin,¹³ have reported interaction second virial coefficients for the $\text{C}_3\text{H}_8\text{-n-C}_4\text{H}_{10}$ binary covering the 288 to 373 K (59 to 212° F ; $\frac{1}{T}=0.0035$ to 0.0027) temperature range as shown in Figure 37. These data are of 'A' quality and sufficient to meet most gas industry needs. This quality ranking was given for the following reasons: 1) a curve constructed from the data of Dantzler et al. and the datum of Mason and Eakin collectively shows good internal consistency; 2) the curve which can be constructed by considering the data of Dantzler et al. and the datum of Mason and Eakin collectively is consistent with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for other propane-paraffin hydrocarbon binary systems data, and with the trend of decreasing values of B_{12} for propane binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 38; and 3) the data of Dantzler et al. for other binary systems have been shown for all other binaries considered in the present study to be of good quality (see Table 2).

4.3.2 $\text{C}_3\text{H}_8\text{-n-C}_5\text{H}_{12}$

The single available data set of Dantzler et al.⁹ for the $\text{C}_3\text{H}_8\text{-n-C}_5\text{H}_{12}$ binary system covering the 298 to 373 K (77 to 212° F ; $\frac{1}{T}=0.0034-0.0027$) is considered to be of 'A' quality based on the following: 1) the data are internally consistent as shown in Figure 39; 2) the data of Dantzler et al. have been shown to be of good quality for all other

binary systems considered in the present study for which Dantzler et al. have measured data; and 3) the data of Dantzler et al. are consistent with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) and the trend of decreasing B_{12} values for propane binaries with other paraffin hydrocarbons of increasing chain length as shown in Figure 40.

4.3.3 C_3H_8 -i- C_5H_{12}

No interaction second virial coefficient information is available for the C_3H_8 -i- C_5H_{12} system; however, the PVTx data measured over the 273 to 573 K (32 to 572°F) range by Vaughn and Collins³⁶ should be useful for the determination of fairly reliable B_{12} values. If these values prove to be accurate they will be sufficient for most needs.

4.3.4 C_3H_8 - CO_2

Four sources, Gunn¹² (Gunn's data are derived from the PVT data of Sage and Lacey¹⁹); Bougard and Jadot;³⁷ Mason and Eakin¹³; and Sie et al.³⁸ have reported interaction second virial coefficients for the C_3H_8 - CO_2 binary covering the temperature range of 288 to 511 K (59 to 460 °F; $\frac{1}{T}=0.0035$ to 0.002). Of the B_{12} 's for these four sources all with the exception of the datum of Sie et al. and the 344 K (150°F; $\frac{1}{T}=0.0029$) datum of Gunn, generally agree with the trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for all of the data and values considered collectively (see Figure 41). The datum of Sie et al. disagrees by about 30% from a smooth curve which would result from considering the others' B_{12} data collectively. The 344 K datum of Gunn disagrees from the smooth curve by about 10%. Further observations through the comparison with similar paraffin hydrocarbon-carbon dioxide binary systems in Figure 42, show that the data from all sources with the exception of the datum of Sie et al. agree with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) and of decreasing B_{12} values with increasing hydrocarbon chain length.

Based on the above considerations the datum of Sie et al. is given a quality ranking of 'C'. The remaining data are given a quality ranking of 'A' based on the following reasons: 1) the data are in agreement within 5% with a smooth curve constructed from all of the data with the exception of 344 K datum of Gunn; 2) Gunn's data have been determined to be of high accuracy for other natural gas binaries; and 3) the datum of Bougard has a reported accuracy of $\pm 4\%$ and the datum of Mason and Eakin has a reported accuracy of $\pm 5 \text{ cm}^3/\text{mol}$.¹

4.3.5 C_3H_8 - N_2

The datum of Mason and Eakin¹³ is the only available B_{12} information for the C_3H_8 - N_2 binary. The quality of the datum of Mason and Eakin is difficult to assess without other C_3H_8 - N_2 binary B_{12} information with which to compare. An examination of a plot of all paraffin hydrocarbon-nitrogen binary systems data in Figure 43 does reveal,

however, that the datum of Mason and Eakin is in the correct range. As shown in Figure 43 the datum of Mason and Eakin falls between the B_{12} 's for the $C_2H_6-N_2$ and the $n-C_4H_{10}-N_2$ binary systems. B_{12} 's generally decrease for nitrogen binaries with paraffin hydrocarbons of increasing chain length; therefore the location of the datum of Mason and Eakin is where it would be expected. Also the datum of Mason and Eakin has a reported uncertainty of $\pm 5 \text{ cm}^3/\text{mol}$. Therefore the datum is considered to be of 'A' quality.

One could, therefore, make crude estimates of interaction second virial coefficient values for the $C_3H_8-N_2$ binary system by generating a line between the point where the B_{12} values for the $C_2H_6-N_2$ and $n-C_4H_{10}-N_2$ binary systems converge down to the datum of Mason and Eakin. B_{12} information gained from generating values by this method may be helpful in estimating properties for design purposes. However, where accurate properties prediction is important, these generated values should not be relied upon. In such cases it is recommended that one determine B_{12} values through the use of a correlation or from existing PVT data until more accurate B_{12} information for the $C_3H_8-N_2$ binary system is made available.

4.3.6 $C_3H_8-H_2$

The datum of Mason and Eakin¹³ at 289 K ($61^\circ F$; $\frac{1}{T}=0.0035$) is the only available interaction second virial coefficient information for the $C_3H_8-H_2$ binary system. The datum of Mason and Eakin is ranked as 'A' quality because the datum has a reported uncertainty of $\pm 5 \text{ cm}^3/\text{mol}$ and much of the other data of Mason and Eakin have been shown to be of 'A' quality. Additional interaction second virial coefficient information may be useful for situations where syngas is mixed with natural gas. However, at present such information is of low priority to the gas industry. Where values are desired these can be derived from available PVT data or calculated with a correlation.

4.3.7 C_3H_8-He

The scatter of as much as $8.5 \text{ cm}^3/\text{mol}$ shown in Figure 44 for the single available data set of Warowny and Stecki³⁹ for the C_3H_8-He binary system indicate that these data are of at best 'B' quality. No additional interaction second virial coefficient data procurement is recommended for the C_3H_8-He binary system, however, since the system is of low importance industrially and desired information may be derived from available PVT data or calculated with a correlation.

4.4 n-Butane Binaries

4.4.1 n-C₄H₁₀-i-C₄H₁₀

The single data set of Connolly⁴⁰ for the n-C₄H₁₀-i-C₄H₁₀ binary covering the temperature range of 344 to 445 K (160 to 341° F; $\frac{1}{T}=0.0029-0.0022$) is ranked as 'A' quality based on the following reasons: 1) The data of Connolly are internally consistent as shown in Figure 45; and 2) the data agree with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative ($dB_{12}/d(\frac{1}{T})$)) for n-butane binaries with other paraffin hydrocarbons and the trend of decreasing B_{12} values for n-butane binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 46; and 3) the data of Connolly are in close agreement (within 6%) with the second virial coefficients for like interactions of n-butane recommended by Dymond and Smith¹ which are considered to be accurate within 5%.

Also estimates of design quality data can be obtained from the recommended values of Dymond and Smith for the second virial coefficients for like interactions of n-butane at temperatures outside the range of Connolly's data.

4.4.2 n-C₄H₁₀-n-C₅H₁₂

Of two sources reporting data for the n-C₄H₁₀-n-C₅H₁₂ binary system covering the 298 to 373 K (77 to 212° F; $\frac{1}{T}=0.0034$ to 0.0027) temperature range as shown in Figure 47, the data of Dantzler et al.⁹ are of 'A' quality while the datum of Jessen and Lightfoot⁴¹ is of 'C' quality. Justification for the ranking of the data of Dantzler et al. is based on the following reasons: 1) the data of Dantzler et al. for the n-C₅H₁₂-n-C₄H₁₀ binary are internally consistent; 2) the data of Dantzler et al. have been determined to be of good quality for other systems considered in the present study for which Dantzler et al. have measured data; 3) the data of Dantzler et al. are in agreement with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) and the general trend of decreasing B_{12} values for n-pentane binaries with other paraffin hydrocarbons of increasing chain length as shown in Figure 48. The datum of Jessen and Lightfoot at 303 K (86° F; $\frac{1}{T}=0.0033$), on the other hand, is ranked as 'C' quality because it deviates from the smooth curve which can be constructed from the data of Dantzler by about 30% and the datum of Jessen and Lightfoot falls in the range of the recommended values by Dymond and Smith¹ for like interactions of n-pentane molecules. As shown in Figure 48 the values for like interactions of n-pentane molecules are consistently lower than the values of Dantzler for unlike interactions between n-pentane and n-butane molecules.

4.4.3 n-C₄H₁₀-CO₂

Of three sources reporting B_{12} 's for the n-C₄H₁₀-CO₂ binary system covering the temperature range of 288 to 478 K (59 to 401° F; $\frac{1}{T}=0.0035$ to 0.02), Gunn's data¹² which have been calculated from the volumetric data of Sage and Lacey¹⁹ and the datum of

Mason and Eakin¹³ are considered reliable, while the datum of Sie et al.³⁸ is of low quality. The reliability of Gunn's data is based on the following: 1) the internal consistency of Gunn's n-C₄H₁₀-CO₂ data as shown in Figure 49 as well as the internal consistency of Gunn's data calculated from the volumetric data of Sage and Lacey for the C₃H₈-CO₂ binary system;¹⁹ and 2) the agreement of Gunn's data with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for other paraffin hydrocarbon-carbon dioxide binaries and the general trend of decreasing B_{12} 's for carbon dioxide binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 50. Gunn's data are, therefore, ranked as 'A' quality.

The unreliability of the datum of Sie et al. is based on the fact that the datum is in disagreement with an extrapolation of Gunn's data by about 30% as shown in Figure 49 and on the fact that the datum of Sie et al. falls approximately in the range of where data for binary interactions of carbon dioxide with propane are expected (see Figure 50). The datum of Sie et al. is, therefore, ranked as 'C' quality.

The reliability of the datum of Mason and Eakin¹³ is difficult to assess because there are no other n-C₄H₁₀-CO₂ data in a similar temperature range with which to compare. However, the datum has a reported accuracy of $\pm 5 \text{ cm}^3/\text{mol}$. Also, the datum of Mason and Eakin follows the general trend of decreasing values for binaries of carbon dioxide with paraffin hydrocarbons of increasing chain length as shown in Figure 50. The data of Mason and Eakin have also been determined to be of 'A' quality for many of the other systems in this work. Therefore, the datum of Mason and Eakin for the n-C₄H₁₀-CO₂ system is likely to be of 'A' quality, but certainly of at least 'B' quality.

The datum of Mason and Eakin may be used to aid in making crude estimations of B_{12} 's through interpolation between the datum of Mason and Eakin (289 K) and the data of Gunn (378 to 478 K). However, for accurate B_{12} values between 288 to 378 K it is recommended that additional B_{12} information be measured or that one rely on PVT data for the calculation of such information.

4.4.4 n-C₄H₁₀-N₂

Five sources, Cruikshank et al.;⁴² Gunn¹² (Gunn's data actually are data derived from the PVT data of Evans and Watson⁴³); Hicks et al.;⁴⁴ Mason and Eakin;¹³ and Young,⁴⁵ have reported interaction second virial coefficients for the n-C₄H₁₀-N₂ binary system as shown in Figure 51. The data of Gunn covering the 427 to 478 K (309 to 401 °F; $\frac{1}{T}$ =0.0023 to 0.0021) temperature range are ranked as 'A' quality based on the following: 1) Gunn's data have been determined to be of good quality in cases where Gunn reported data for other binaries considered in the present study; 2) the data of Gunn for the n-C₄H₁₀-N₂ binary are internally consistent as shown in Figure 51; and 3) as shown in Figure 52 Gunn's data for the n-C₄H₁₀-N₂ binary are consistent with the general trends for B_{12} versus $\frac{1}{T}$ (and for the temperature derivative, $dB_{12}/d(\frac{1}{T})$) and also for the general trends of decreasing B_{12} values for nitrogen binaries with paraffin hydrocarbons of increasing chain length.

The data of Cruikshank et al.; Hicks et al.; and Young covering the 303 to 333 K (86 to 140 °F; $\frac{1}{T}$ =0.0033 to 0.003) temperature range, are regarded as 'B' quality for the following

reasons: 1) the data are in agreement within $15 \text{ cm}^3/\text{mol}$; 2) the reported uncertainties for the data from these sources are larger than $5 \text{ cm}^3/\text{mol}$ but less than $15 \text{ cm}^3/\text{mol}$.¹ The uncertainty for the datum of Cruikshank et al. is $\pm 9 \text{ cm}^3/\text{mol}$, the uncertainty for the data of Hicks et al. is $\pm 12 \text{ cm}^3/\text{mol}$, and the uncertainty for the datum of Young is $\pm 12 \text{ cm}^3/\text{mol}$; and 3) the qualities of the data of these authors have been shown to be less than 'A' for other binary systems considered in the present study. For instance, the data of Cruikshank et al. for the n-C₅H₁₂-N₂ and the n-C₅H₁₂-H₂ binaries; the data of Hicks for the n-C₅H₁₂-N₂ binary; and the data of Young for the n-C₅H₁₂-N₂ and the i-C₅H₁₂-N₂ binaries all have been shown to be of low quality (see individual discussions for these particular binaries).

The datum of Mason and Eakin at 289 K (61° F; $\frac{1}{T}=0.0035$) has a reported uncertainty of $\pm 5 \text{ cm}^3/\text{mol}$.¹ The uncertainty of $5 \text{ cm}^3/\text{mol}$ would normally rank the datum of Mason and Eakin in the 'A' category. One cannot in this case make such a judgment with confidence, however, because there are no other quality data in the same temperature range with which to compare the datum of Mason and Eakin for the n-C₄H₁₀-N₂ binary. The datum of Mason and Eakin, however, follows the general trends of decreasing B_{12} values for nitrogen binaries with paraffin of increasing chain length. Based on these considerations the datum of Mason and Eakin is likely to be of 'A' quality, but certainly is of at least 'B' quality. Therefore, an interpolation between the datum of Mason and Eakin (289 K) and the data of Gunn (428 to 478 K) may be useful in providing crude estimates of B_{12} values in the temperature range of 289 to 438 K until additional quality data are obtained. (Note: Such an interpolation would give more credence to the data of Hicks et al.)

4.4.5 n-C₄H₁₀-H₂

The datum of Mason and Eakin¹³ for the n-C₄H₁₀-H₂ binary system is the only available interaction second virial coefficient information for this system. However, the datum of Mason and Eakin is ranked as 'A' quality based on the reported uncertainty of $\pm 5 \text{ cm}^3/\text{mol}$ ¹ and on the fact that the data of Mason and Eakin have been shown to be of good quality in most other cases in the present study.

4.4.6 n-C₄H₁₀-He

The large amount of scatter of as much as $10 \text{ cm}^3/\text{mol}$ in the data of Jones and Kay⁴⁶ covering the 373 to 498 K (212 to 439° F; $\frac{1}{T}=0.027$ to 0.002) temperature range as shown in Figure 53 for the n-C₄H₁₀-H₂ binary system is an indication that these data are at best of 'B' quality. Note that all the data fall within a $10 \text{ cm}^3/\text{mol}$ band. There are no other available data for the n-C₄H₁₀-He binary system. Other desired values can be derived from available PVT data or determined with the use of a correlation.

4.5 Isobutane Binaries

With the exception of the CH_4 - $i\text{-C}_4\text{H}_{10}$ and $n\text{-C}_4\text{H}_{10}$ - $i\text{-C}_4\text{H}_{10}$ binaries for which B_{12} data are available, there are no interaction second virial coefficient data for other isobutane binary systems which are of importance for the gas industries. It has been shown, however, in the discussions for the CH_4 - $i\text{-C}_4\text{H}_{10}$ and $n\text{-C}_4\text{H}_{10}$ - $i\text{-C}_4\text{H}_{10}$ binary data evaluations that these data could be closely approximated by the data for the CH_4 - $n\text{-C}_4\text{H}_{10}$ binary interaction second virial coefficient data and the second virial coefficient values for like interactions of $n\text{-C}_4\text{H}_{10}$ which were recommended by Dymond and Smith.¹ Thus, where $n\text{-C}_4\text{H}_{10}$ binary data exist for binaries of industrial importance, these data may be used to estimate the $i\text{-C}_4\text{H}_{10}$ binary system values. $i\text{-C}_4\text{H}_{10}$ binaries for which B_{12} values can be approximated by existing B_{12} data or values of corresponding $n\text{-C}_4\text{H}_{10}$ binaries are listed as follows: $i\text{-C}_4\text{H}_{10}$ - CH_4 , $i\text{-C}_4\text{H}_{10}$ - CO_2 , $i\text{-C}_4\text{H}_{10}$ - N_2 , $i\text{-C}_4\text{H}_{10}$ - C_2H_6 , $i\text{-C}_4\text{H}_{10}$ - C_3H_8 , $i\text{-C}_4\text{H}_{10}$ - $n\text{-C}_5\text{H}_{12}$, $i\text{-C}_4\text{H}_{10}$ - $i\text{-C}_5\text{H}_{12}$, and $i\text{-C}_4\text{H}_{10}$ -neo- C_5H_{12} . This list includes all i -butane binaries that are considered in this report to be of greatest importance to the gas industry.

4.6 n-Pentane Binaries

4.6.1 $n\text{-C}_5\text{H}_{12}$ - CO_2

Four sources, Vigdergauz and Semkin;⁴⁷ Massoudi and King;²⁰ Sie;³⁸ and Desty et al.⁴⁸ have reported data for the $n\text{-C}_5\text{H}_{12}$ - CO_2 binary covering the temperature range of 298 to 353 K (77 to 176° F; $\frac{1}{T}=0.0034$ -0.0028) as shown in Figure 54. The datum of Vigdergauz and Semkin, the datum of Sie, and the datum of Desty are of low quality because they are inconsistent with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative of $dB_{12}/d(\frac{1}{T})$), and with the general trend of decreasing B_{12} values with carbon dioxide binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 55. Further evidence indicating that these data are of low quality is given in the following statements: The data of Vigdergauz and Semkin were given a low ranking through the evaluations made by Dymond and Smith;¹ Sie's data have been shown to be of poor quality for other binary systems as discussed in the evaluations of the CO_2 - C_3H_8 and CO_2 - $n\text{-C}_4\text{H}_{10}$ binary systems; and the datum of Desty for the $n\text{-C}_5\text{H}_{12}$ - CO_2 binary system has an uncertainty of $\pm 17\%$.¹ Based on the above considerations the datum of Vigdergauz and Semkin, the datum of Sie and the datum of Desty are determined to be of 'C' quality.

The datum of Massoudi and King, on the other hand, is consistent with the general trend of decreasing B_{12} values for carbon dioxide binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 55 and the datum of Massoudi and King has a reported error of about 8% (1). The datum of Massoudi and King, therefore, is given a quality ranking of 'B'.

4.6.2 n-C₅H₁₂-N₂

Twelve sources have reported data for the n-C₅H₁₂-N₂ binary system. For these data there is a large amount of scatter as shown in Figure 56. Because the reported errors are greater than 10% or 15 cm^3/mol^1 for Desty et al.;⁴⁸ Letcher and Marsicano;⁴⁹ Vigdergauz and Semkin;⁴⁷ and Young,⁴⁵ these data are given a quality ranking of 'C'. Data reported by all other sources have accuracies which would normally give them a quality ranking of 'B' because these reported errors are less than 10% or 15 cm^3/mol^1 . These sources are Cruikshank et al. (1966);⁵⁰ Cruikshank et al. (1968);⁴² Everett;⁵¹ Gainey (1967);⁵² Gainey and Pecsok (1970);⁵³ Hicks and Young;⁴⁴ Leung and Eichinger;⁵⁴ and Massoudi and King.²⁰ It should be noted, however, that several of the reported data points are in considerable disagreement at 298 K (77° F; $\frac{1}{T}=0.0034$). Sources reporting the data in question are Cruikshank et al. (1966); Everett; Leung and Eichinger; and Massoudi and King. These sources have reported data at 298 K which range between 75 and 105 cm^3/mol , as shown in Figure 56. Since these data cover such a wide range the accuracy of these data are uncertain and therefore the 'B' ranking for these data is questionable.

4.6.3 n-C₅H₁₂-H₂

Four sources have reported interaction second virial coefficients for the n-C₅H₁₂-H₂ binary system as shown in Figure 57. The datum of Cruikshank et al.,⁵⁰ the datum of Desty et al.,⁴⁸ the datum of Everett⁵¹ and the datum of Mason and Eakin¹³ agree within 15 cm^3/mol over the 288 to 298 K (59 to 77° F; $\frac{1}{T}=0.0035$ to 0.0034) temperature range and, therefore, are of at least 'B' quality. The datum of Mason and Eakin, however, is considered to be of better quality than the others' data since it has a reported uncertainty of $\pm 5 \text{ cm}^3/\text{mol}^1$ and data of Mason and Eakin have been shown to be reliable in most other cases in the present study. Therefore, it is given an 'A' quality ranking.

The datum of Cruikshank et al. and the datum of Everett also have low reported uncertainties, but the data for these authors have been shown to be less reliable for other binaries in the present study. Therefore, the data of Cruikshank et al. and the datum of Everett are ranked as 'B' quality. The datum of Desty et al. is probably not of greater than 'B' quality because of the low ranking these authors' data received for other systems in this paper. Desired interaction second virial coefficient values for the n-C₅H₁₂-H₂ binary system may also be derived from available PVT data or determined with the use of a correlation.

4.7 Isopentane Binaries

4.7.1 i-C₅H₁₂-CO₂

The datum of Desty et al.⁴⁸ for the i-C₅H₁₂-CO₂ binary system at 298 K (77° F) is ranked as 'C' quality because it is in disagreement with the general trends of decreasing

B_{12} values for CO_2 binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 58. Figure 58 reveals that the datum of Desty et al. for the $i\text{-C}_5\text{H}_{12}\text{-CO}_2$ binary roughly falls in the region where one would expect to find data for the $\text{C}_3\text{H}_8\text{-CO}_2$ binary.

4.7.2 $i\text{-C}_5\text{H}_{12}\text{-N}_2$

For the $i\text{-C}_5\text{H}_{12}\text{-N}_2$ binary, Desty et al.,⁴⁸ Gainey and Pecsok,⁵³ and Young⁴⁵ measured data over the 298 to 338 K (77 to 149° F; $\frac{1}{T}=0.0034\text{--}0.003$) temperature region as shown in Figure 59. It is difficult to assess the quality of the datum of Gainey and Pecsok and the datum of Desty et al. because of the lack of other quality data for either the $i\text{-C}_5\text{H}_{12}\text{-N}_2$ binary or other paraffin hydrocarbon-nitrogen binaries. However, the reported uncertainties of $\pm 30\text{ cm}^3/\text{mol}$ for the datum of Desty et al. and of $6\text{ cm}^3/\text{mol}$ for the datum of Gainey and Pecsok would place these data in the 'C' and 'B' categories, respectively. Corresponding rankings for these authors for the $n\text{-C}_5\text{H}_{12}\text{-N}_2$ binary would support the above rankings.

The $i\text{-C}_5\text{H}_{12}\text{-N}_2$ binary data of Young are given a 'C' ranking because of their disagreement with the general trends of the B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for other nitrogen binaries with paraffin hydrocarbons. Note that the $\frac{1}{T}$ dependency of the B_{12} data of Young has a positive slope in Figure 60 while the $\frac{1}{T}$ dependency for all other N_2 binaries with paraffin hydrocarbons has a negative slope. Despite the lack of data for isopentane binary systems the need for binary interaction virial coefficients for isopentane binary systems can be supplemented with information for either $n\text{-pentane}$ binary systems or neopentane binary systems. A similar case was made for the isobutane binaries where it was proposed that isobutane binary information be supplemented with information for $n\text{-butane}$ binaries.

4.7.3 $i\text{-C}_5\text{H}_{12}\text{-H}_2$

The datum of Desty et al.⁴⁸ at 298 K (77° F; $\frac{1}{T}=0.0034$) is the only available interaction second virial coefficient information for the $i\text{-C}_5\text{H}_{12}\text{-H}_2$ binary system. The datum has a reported uncertainty of $\pm 30\text{ cm}^3/\text{mol}$ and, therefore, is given a 'C' ranking. Where other interaction second virial coefficient values are desired for this system these may be determined with the use of a correlation.

4.8 Neopentane Binaries

4.8.1 $\text{neo-C}_5\text{H}_{12}\text{-N}_2$

The reported uncertainty for the interaction second virial coefficient data of Baughman et al.²⁶ covering the 199 to 258 K (-101 to 5° F; $\frac{1}{T}=0.005$ to 0.0039) temperature range for the $\text{neo-C}_5\text{H}_{12}\text{-N}_2$ binary system is $\pm 10\%$, which places these data on the borderline between 'B' and 'C' quality. However, since the three methods of analysis used

by Baughman et al. show fairly consistent trends (see Figure 61) the data are given a 'B' quality ranking. In addition these data are in agreement with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for other binaries with paraffin hydrocarbons and the general trend of decreasing B_{12} values for nitrogen binaries with paraffin hydrocarbons of increasing chain length as shown in Figure 62. Where other interaction second virial coefficient values are desired for this binary, these can be calculated with the use of a correlation or estimated with the values for nitrogen binaries with other pentane isomers.

4.8.2 neo-C₅H₁₂-He

The uncertainty reported for the interaction second virial coefficient data of Baughman et al.²⁶ for the neo-C₅H₁₂-He binary system is $\pm 10\%$.¹ However, since a plot of the data of Baughman et al. in Figure 63 reveals a lack of consistency for the trend of B_{12} versus $\frac{1}{T}$ with that shown for other similar binaries, the data of Baughman are ranked as 'C' quality.

4.8.3 Other Neopentane Binaries

Interaction second virial coefficient information is not available for other neopentane binaries of importance to the gas industry. In light of this neopentane B_{12} information can be estimated from the binary interaction second virial coefficient data for the corresponding pentane isomers. The rationale for this has been discussed in detail in previous sections on isobutane and isopentane binary data.

4.9 CO Binaries

4.9.1 CO-CO₂

The single available data set of Cottrell et al.,⁵⁵ covering the 303 to 363 K (86 to 194° F; $\frac{1}{T}=0.0033$ to 0.0026) temperature range for the CO-CO₂ binary system is considered to be of 'B' quality (see Figure 64). The 'B' quality ranking is based on the fact that the CO-CO₂ binary interaction second virial coefficient data points measured by Cottrell et al., deviate from each other by less than 11 cm^3/mol and the data measured by either Cottrell, Hamilton and Taubinger, or Cottrell and Hamilton for all other binary systems in the present study are of good quality.

4.9.2 CO-H₂

Reuss and Beenakker;⁵⁶ Townend and Bhatt;⁵⁷ and Scott,⁵⁸ have reported 'A' quality interaction second virial coefficient data for the H₂-CO binary system. The data of Reuss and Beenakker covering the 36 to 60 K (-395 to to 352° F; $\frac{1}{T}=0.028$ to 0.017)

temperature range as shown in Figure 65 are given an 'A' quality ranking based on the internal consistency of these data and on the internal consistency of other data measured by Reuss and Beenakker for the N_2 - H_2 binary. There are no other low temperature interaction second virial coefficient data with which to compare the data of Reuss and Beenakker. However, the solid-vapor equilibrium measurements made by Van Itterbeek and Van Donimck⁵⁹ may be used to calculate interaction second virials with which to compare the existing data of Reuss and Beenakker. It should be mentioned that although the data of Reuss and Beenakker are given an 'A' ranking, low temperature data for the CO - H_2 binary system are of low importance to the syngas industry since all processing and transport of these syngases takes place at higher temperatures.

The data of Townend and Bhatt and the data of Scott are ranked as 'A' quality based on the good agreement (within $3 \text{ cm}^3/\text{mol}$) between the two data sets at 298 K (77° F ; $\frac{1}{T}=0.0034$).

4.10 Carbon Dioxide Binaries

4.10.1 CO_2 - CS_2

The only available information for CO_2 - CS_2 binary is the datum of Eucken and Bresler⁶⁰ at 273 K . Since there are no other data with which to compare the datum of Eucken and Bresler no data ranking was given for this binary system. Since binaries with CS_2 are of relatively low importance to the syngas industry no additional data procurement is recommended. However, available data may be used to test correlations which in turn may be useful in providing desired CS_2 binary data for industry.

4.10.2 CO_2 - N_2

Seven sources, Cottrell et al.;⁵⁵ Edwards and Roseveare;⁶¹ Gorski and Miller;⁶² Gunn;¹² Mason and Eakin;¹³ Pfefferle et al.;⁶³ and Yakimenko et al.⁶⁴ have reported interaction second virial coefficients covering the 110 to 298 K (-262 to 77° F ; $\frac{1}{T}=0.0091$ to 0.0034) temperature range for the CO_2 - N_2 binary system as shown in Figure 66. Of these data those reported by Gunn (Gunn's data actually were derived from the volumetric data of Tang¹⁶), the datum of Gorski and Miller and the datum of Pfefferle et al. are ranked as 'A' quality for the following reasons: 1) the reported values of Gunn are internally consistent as shown in Figure 67; 2) Gunn's value at 298 K (77° F ; $\frac{1}{T}=0.0034$) is in good agreement with the 298 K datum of Gorski and Miller and of Pfefferle et al. (Pfefferle has also reported quality data for the CO_2 -He and the N_2 -He binary systems); and 3) Gunn's values have been of high quality for other binaries in this study.

The data of Cottrell et al. are ranked as 'B' quality for the following reasons: 1) the data of Cottrell et al. disagree with a smooth curve constructed from Gunn's values by as much as $5 \text{ cm}^3/\text{mol}$; 2) a smooth curve constructed from the data of Cottrell et al. disagrees with the general trends of B_{12} versus $\frac{1}{T}$, (and the temperature derivative

$dB_{12}/d(\frac{1}{T})$) for smooth curves constructed for other B_{12} data of the present study: and 3) the reported uncertainty in the data of Cottrell et al. is as high as 15%.

The datum of Edwards and Roseveare and the datum of Mason and Eakin are ranked as 'A' quality because they agree within $5 \text{ cm}^3/\text{mol}$ of an extrapolation of a smooth curve constructed from other data determined to be reliable. The low temperature data of Yakimenko et al. are considered to be of at least 'B' quality because the data are internally consistent and appear to be in agreement with an extrapolation which could be made from the data at higher temperatures.

4.10.3 $\text{CO}_2\text{-H}_2$

Two sources have reported B_{12} data for the $\text{CO}_2\text{-H}_2$ binary system covering the 298 to 363 K (77 to 194° F; $\frac{1}{T}=0.034$ to 0.0028) temperature range as shown in Figure 68. The data of Cottrell et al.⁵⁵ are of 'A' quality while the datum of Edwards and Roseveare⁶¹ is of 'C' quality. The data of Cottrell et al. are ranked as 'A' quality for the following reasons: 1) the data of Cottrell et al. for the $\text{CO}_2\text{-H}_2$ binary are internally consistent as shown in Figure 68; 2) the data have reported uncertainties of less than $5 \text{ cm}^3/\text{mol}$;¹ and 3) other CO_2 binary measurements made by Cottrell and Hamilton;⁶⁵ or Cottrell, Hamilton and Taubinger⁵⁵ have also been determined to be of good quality as shown in the evaluations of the data for the $\text{CO}_2\text{-CO}$, $\text{CO}_2\text{-N}_2$ and $\text{CO}_2\text{-He}$ binary systems.

The datum of Edwards and Roseveare has been given a quality ranking of 'C' based on the fact that the datum deviates from a smooth curve constructed from the data of Cottrell et al. by about $32 \text{ cm}^3/\text{mol}$ as shown in Figure 68, and data measured by Edwards and Roseveare have been shown to deviate significantly from other quality data in other binary systems evaluations (see the evaluations for the $\text{N}_2\text{-He}$ and the $\text{CO}_2\text{-He}$ binary systems).

4.10.4 $\text{CO}_2\text{-H}_2\text{O}$

Of the two sources reporting data for the $\text{H}_2\text{O-CO}_2$ binary system covering the 298 to 373 K (77 to 212° F; $\frac{1}{T}=0.0034$ to 0.0027) temperature range, the data of Coan and King³⁴ are regarded as 'B' quality while the data of Pollitzer and Strebel⁶⁶ are regarded as 'C' quality. The ranking for the data of Coan and King is based on the following: 1) The data of Coan and King have a reported error of less than 10%;¹ 2) the data of Coan and King are internally consistent as shown in Figure 69; and 3) the data of Coan and King have been shown to be of 'B' quality for other binary systems of interest in the present study (see evaluations for the $\text{C}_2\text{H}_6\text{-H}_2\text{O}$ and $\text{N}_2\text{O-H}_2\text{O}$ binary systems).

The data of Pollitzer and Strebel are ranked as 'C' quality based on the fact that they show a large deviation from the data of Coan and King as shown in Figure 69. Also, when the two data points measured by Pollitzer and Strebel are considered collectively they disagree with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for all other quality interaction second virial coefficient data for the binary systems considered to be of importance in the present study.

4.10.5 CO₂-H₂S

No data are presently available for the CO₂-H₂S binary; however, the PVT data of Bierlein and Kay⁶⁷ covering the 274 to 354 *K* temperature region may be useful in calculating interaction second virial coefficients for this system.

4.10.6 CO₂-He

Of seven sources, Bell and Dunlop;³² Cottrell and Hamilton;⁶⁵ Edwards and Roseveare;⁶¹ Harper and Miller;⁶⁸ Linshits et al.;⁶⁹ Pfefferle et al.;⁶³ and Tsiklis et al.,^{70,71} reporting interaction second virial coefficient data for the CO₂-He binary covering the 292 to 423 *K* (66 to 302° *F*; $\frac{1}{T}$ =0.0034 to 0.0024) all data but those of Edwards and Roseveare are in fair agreement with a general trend of B_{12} versus $\frac{1}{T}$ as shown in Figure 70. Since the datum of Edwards and Roseveare disagrees by about 55 *cm*³/*mol* from the data of the other sources, it is given a 'C' quality ranking. An expanded plot of the data for the six sources excluding the datum of Edwards and Roseveare in Figure 71 reveals that a fair amount of scatter still exists for these data. Therefore, it is difficult to give an accurate ranking of the data for the remaining six sources. However, these data are of at least 'B' quality since they agree within 15 *cm*³/*mol*. Where interaction second virial coefficient values are desired for other temperature ranges for the CO₂-He binary system, these can be derived from available PVT data or calculated using a correlation.

4.11 CS₂ Binaries

CS₂ binaries which are of greatest importance to the gas industry are those of CS₂ with the major constituents of syngas. These binaries are CS₂-CO₂, CS₂-H₂, CS₂-CO, CS₂-H₂O, CS₂-CH₄ and CS₂-N₂. The only available data for CS₂ binaries are those of Eucken and Bresler⁶⁰ who have measured a single datum for each of the CS₂-CO₂, CS₂-N₂, and CS₂-H₂ binaries at 273 *K*. Since there are no other data with which to compare the data of Eucken and Bresler no data rankings are given for these binary system data.

4.12 COS Binaries

COS is a minor constituent of syngas product streams and at present there are no available data for COS binaries with syngas components. The COS binaries of highest importance to the gas industry are those binaries with the major constituents of syngas, COS-CO₂, COS-H₂, COS-CO, COS-H₂O, COS-CH₄ and COS-N₂. Where COS binary interaction second virial coefficient values are desired, these can be calculated using available correlations.

4.13 Nitrogen Binaries

4.13.1 N₂-H₂

Interaction second virial coefficient values for the N₂-H₂ binary system have been reported by eight sources, Brewer and Vaughn;⁷² Edwards and Roseveare;⁶¹ Lunbeck;⁷³ Michels and Wassenaar;⁷⁴ Ostronov et al.;⁷⁵ Verschoyle;⁷⁶ Vilcu and Gainer;⁷ and Zandbergen and Beenakker⁷⁷ covering the 148 to 323 *K* (-193 to 122° *F*; $\frac{1}{T}$ =0.0067 to 0.0031) temperature range as shown in Figures 72 and 73. Of the existing data for the N₂-H₂ binary system all the data with the exception of the data of Ostronov et al. are given an 'A' quality ranking.

The 'A' quality ranking for the data of Brewer and Vaughn and the data of Zandbergen and Beenakker at 270 *K* and below is based on the following reasons: 1) these data are internally consistent as shown in Figure 73; and 2) the data for these two sources agree within 1 *cm*³/*mol*.

The data of Brewer and Vaughn above 273 *K*, the datum of Edwards and Roseveare, the datum of Lunbeck, the datum of Michels and Wassenaar, the data of Verschoyle, and the datum of Vilcu and Gainer are ranked as 'A' quality because they are in agreement within at least 4 *cm*³/*mol* and they are in agreement with an extrapolation of the data of Zandbergen and Beenakker within at least 4 *cm*³/*mol*.

The data of Ostronov et al., are ranked as 'B' quality for the following reasons: 1) the data of Ostronov et al. deviate by as much as 9 *cm*³/*mol* from a smooth curve constructed by considering the data of Zandbergen and Beenakker and the data of Brewer and Vaughn collectively; and 2) the data of Ostronov et al. disagree with the general trends of *B*₁₂ versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) for the data of the remaining sources considered collectively. The higher temperature data of Ostronov et al., however, are in better agreement with the data from the other sources, and, are therefore, of better quality.

Data of Reuss and Beenakker⁵⁶ covering the lower temperature region of 36 to 60 *K* (-395 to -352° *F*; $\frac{1}{T}$ =0.028 to 0.017) shown in Figure 72 are of at least of 'B' quality since the data of Reuss and Beenakker are internally consistent, and appear to be in agreement with an extrapolation which could be made from a curve constructed from the higher temperature data.

4.13.2 N₂-H₂O

Of two sources, Rigby et al.;³⁰ and Richards et al.⁷⁸ who have reported interaction second virial coefficients for the N₂-H₂O binary system covering the 298 to 423 *K* (77 to 302° *F*; $\frac{1}{T}$ =0.0034 to 0.0024) temperature range as shown in Figure 74, both have reported 'A' quality data. The 'A' ranking is based on the following reasons: 1) the data for both sources are internally consistent as shown in Figure 74; 2) a continuous smooth curve can be constructed from considering both the data of Richards et al. and the data of Rigby

et al. collectively; 3) the data of Rigby et al. have a reported uncertainty of less than $6 \text{ cm}^3/\text{mol}$;¹ and 4) Richards et al. have shown with the use of a theoretical analysis using the Stockmayer potential that their data are in good agreement with the data of Rigby et al. Figure 74 shows that the 400 K (261° F; $\frac{1}{T}=0.0025$) datum of Richards et al. is in disagreement with a smooth curve which can be constructed from the data. However, the disagreement from the smooth curve for the 400 K datum is less than $4 \text{ cm}^3/\text{mol}$ and therefore, the 400 K datum is still considered to be of 'A' quality.

The existing data for the N₂-H₂O binary system may be supplemented with interaction second virial coefficients calculated from either the data for water solubility in compressed nitrogen of Bartlett⁷⁹ covering the 298 to 323 K temperature range and the measurements of the solubility of water in the compressed gas by Saddington and Krase.⁸⁰ Additional evaluation procedures would be necessary in order to assess the quality of B_{12} values derived from the data of Bartlett and the data of Saddington and Krase.

4.13.3 N₂-H₂S

No interaction second virial coefficient information is available for the N₂-H₂S binary. However, the mixture virial coefficients, B(M), given by Robinson et al.,⁸¹ for the N₂-H₂S binary system covering the 293 to 372 K temperature range are useful in calculating interaction second virial coefficients where such information is needed.

4.13.4 N₂-He

Ten sources report interaction second virial coefficient data for the N₂-He binary system covering the 90 to 748 K (-262 to 887° F; $\frac{1}{T}=0.011$ to 0.0013) temperature range as shown in Figure 75. These sources are: Bell and Dunlop;³² Brewer and Vaughn;⁷² Canfield et al.;⁸² Edwards and Roseveare;⁶¹ Hall and Canfield;⁸³ Knobler et al.;⁸⁴ Kramer and Miller;⁸⁵ Ku and Dodge;⁸⁶ Pfefferle et al.;⁶³ and Witonsky and Miller.⁸⁷

Six of the above sources, Canfield et al.; Brewer and Vaughn; Bell and Dunlop; Kramer and Miller; Ku and Dodge; and Pfefferle et al. report data covering the 133 to 373 K (-220 to 212° F; $\frac{1}{T}=0.0075$ to 0.0027) temperature region for the N₂-He binary system which are considered to be of 'A' quality. The 'A' ranking for the data of these six sources is based on the following reasons: 1) these data are in good agreement with a smooth curve constructed by considering all of the data collectively (having an average deviation of less than $1 \text{ cm}^3/\text{mol}$). The largest disagreement between any of the data points for these six sources are for the 273 and 223 K data of Canfield et al. and the 273 and 223 K data points of Brewer and Vaughn which disagree by about $1.5 \text{ cm}^3/\text{mol}$ from a smooth curve. However, all of the other data points are in considerably better agreement with the smooth curve; and 2) several of the authors have measured data for other systems for which their data received an 'A' quality ranking. For instance, the data of Brewer and Vaughn have been shown to be of 'A' quality for the N₂-H₂ binary and of 'A' quality for the H₂-He binary system; the data of Bell and Dunlop have been shown to be of 'A' quality for the CH₄-He binary system; and the data of Pfefferle et al. have been shown to be of 'A' quality for the CO₂-He binary system.

The datum of Edwards and Roseveare at 298 K (77° F; $\frac{1}{T}=0.0034$) and the data of Hall and Canfield covering the at 103 to 113 K (-274 to 256° F; $\frac{1}{T}=0.0097$ to 0.0088) temperature range are ranked as 'B' quality. The datum of Edwards and Roseveare has received a 'B' ranking because it deviates from other quality data by about 9 cm^3/mol . Although there are no other data with which to compare the data of Hall and Canfield, a 'B' ranking has been given to these data because they appear to be in fair agreement with an extrapolation which can be made from other quality high temperature data down to the lower temperature region. However, further data procurement may reveal that the data of Hall and Canfield are of better quality. The datum of Knobler et al. is ranked as 'C' quality because it deviates significantly from an extrapolation which can be made from quality higher temperature data down to lower temperatures.

The higher temperature data of Witonsky and Miller covering the 448 to 748 K (347 to 887° F; $\frac{1}{T}=0.0022$ to 0.0013) temperature range at first appear to be of low quality since a smooth curve constructed from the data of Witonsky and Miller disagrees with the general trends of B_{12} versus $\frac{1}{T}$ (and the temperature derivative, $dB_{12}/d(\frac{1}{T})$) that are shown for the quality data at lower temperatures. However, second virial coefficient theory shows that values asymptotically approach a single value as temperatures increase. This is most likely the case for the data of Witonsky and Miller since all of their data fall within a range of 3 cm^3/mol . It should also be mentioned that the apparent large positive slope for the data of Witonsky and Miller is deceiving since a plot of B_{12} versus inverse temperature tends to bring points closer together as temperature is increased (ie. at smaller values of $\frac{1}{T}$). For this reason the data of Witonsky and Miller are considered to be of at least 'B' quality. However, future procurement of additional data at high temperatures may be helpful in verifying the reliability of these data.

4.13.5 N₂-NH₃

The only available data for N₂-NH₃ binary system are the data of Lee et al.³³ covering the 318 to 673 K (113 to 752° F; $\frac{1}{T}=0.0031$ to 0.0015) temperature range as shown in Figure 76. These data are at least of 'B' quality since the data of Lee et al. for both the N₂-NH₃ and the CH₄-NH₃ binaries are internally consistent. The procurement of additional data may be helpful in verifying the quality of these data.

4.14 NO_x Binaries

4.14.1 N₂O-H₂O

The only existing interaction second virial coefficient data for the N₂O-H₂O binary are those of Coan and King³⁴ covering the 298 to 373 K (77 to 212° F; $\frac{1}{T}=0.0034$ to 0.0027) temperature range as shown in Figure 77. The data of Coan and King are given an 'A' quality ranking since the data are internally consistent, and these data have a reported accuracy of about 5%.

4.14.2 Other NO_x Binaries

The NO_x binaries of greater importance to the gas industries are those of N₂O, NO, NO₂, and N₂O with the major syngas components H₂, CO, H₂O, CO₂, CH₄ and N₂. However, the only available information is that which has already been discussed for the N₂O-H₂O system above. Other existing NO_x mixture data from which interaction second virial coefficient values can be derived are the data of Markham and Kobe⁸⁸ for the percentage volume change on mixing for the N₂O-CO₂ and the N₂O-N₂ binary systems at 298 K, the Joule-Thomson coefficients for the N₂O-N₂ and N₂O-CO₂ binary systems determined by Charnley et al.⁸⁹ covering the 298 to 318 K temperature range, and the volumetric behavior of the NO-NO₂ binary system reported by Selleck et al.⁹⁰ covering the 280 to 440 K temperature range.

4.15 Hydrogen Binaries

4.15.1 H₂-H₂O

With the exception of the data of Bartlett⁷⁹ for water solubility in compressed hydrogen which can be used to derive interaction second virial coefficients, no interaction second virial coefficient information is available for the H₂-H₂O binary system.

4.15.2 H₂-He

Figure 78 shows a plot of B_{12} versus $\frac{1}{T}$ for the H₂-He binary system data covering the 14 to 448 K (-434 to 347° F; $\frac{1}{T}=0.07$ to 0.0022) temperature range. This plot includes the data from six sources, Beenakker et al.;⁹¹ Brewer and Vaughn;⁷² Gibby et al.;⁹² Knobler et al.;⁸⁴ Tanner and Masson;⁹³ and Varekamp and Beenakker.⁹⁴ Two sources, Beenakker et al.; and Varekamp and Beenakker have also reported data for binaries of helium with hydrogen isotopes. As shown in Figure 78 the data for the isotopic binaries are in close agreement with the data for the nonisotopic binaries. Figure 78 also reveals that an extrapolation of the curve constructed from the low temperature data of Beenakker et al. at 20 K (-423° F; $\frac{1}{T}=0.05$) and the low temperature data of Varekamp and Beenakker covering the 14 to 17 K (-434 to to 429)° F; $\frac{1}{T}=0.07$ to 0.059) temperature range would be in fair agreement with the higher temperature data covering the 90 to 448 K (-298-347° F; $\frac{1}{T}=0.01$ to 0.0022) temperature range of the remaining data. Therefore, intermediate temperature values between 20 K and 248 K (-13° F; $\frac{1}{T}=0.0040$) may be estimated from the existing data. A quality ranking of 'A' is given to the data in the 148 to 448 K temperature range since these data agree within 2% (see Figure 79). The data for the lower temperature region (see Figure 80) agree within 5 cm³/mol from a curve which can be constructed by considering all of the low temperature data collectively and therefore are regarded as 'A' quality. The 90 K datum of Knobler et al. is difficult to assess because of the lack of similar data, but is regarded as being of 'B' quality since it is in agreement with an extrapolation of a smooth curve through the lower temperature range data (see

Figure 80). Note however, that the datum of Knobler et al. received a 'C' ranking for the N₂-He binary and therefore, the 'B' ranking in the present case is suspect. Additional data would help to clarify this question.

4.16 Hydrogen Cyanide Binaries

The HCN binaries of greater importance to the gas industries are those of HCN with the major syngas constituents, H₂, CO, H₂O, CO₂, CH₄ and N₂. At present there are no available interaction second virial coefficient data or values for these HCN binaries. It is recommended that desired values for these systems be derived from other existing thermodynamic data or calculated using an available correlation.

4.17 SO_x Binaries

At present there are no available interaction second virial coefficient data or values for SO_x binaries which are of importance to the gas industry. The most important SO_x binaries in the present study are those with the major constituents of syngas. These binaries are SO_x-H₂, SO_x-CO, SO_x-H₂O, SO_x-CO₂, SO_x-CH₄ and SO_x-N₂. It is recommended that desired values be derived from other existing thermodynamic data or calculated using an available correlation.

4.18 Ammonia Binaries

Since ammonia is a product of coal gasification processes, the ammonia binaries which are of importance to the gas industry are those binaries involving ammonia with the important constituents of syngas. These binaries include H₂-NH₃, CO-NH₃, H₂O-NH₃, CO₂-NH₃ and CH₄-NH₃. The only available data for any of these binaries are the data of Lee et al. for the CH₄-NH₃ and the N₂-NH₃ binaries covering the 423 to 573 K (302 to 572° F; $\frac{1}{T}$ =0.0024 to 0.0017) and the 318 to 673 K (113 to 752° F; $\frac{1}{T}$ =0.0031 to 0.0015) temperature ranges respectively as shown in Figures 23 and 76. These data have been discussed in previous sections. Data for ammonia binaries for other syngas components may be derived from correlations.

Acknowledgments

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Table 2
Interaction Second Virial Coefficient Data Ranges and Rank

System	Data Rank	Temperature Range (K)	Source
CH ₄ -C ₂ H ₆	A	298.15/373.15	Dantzler ⁹ (1968)
	X	273/323	Guggenheim ⁴ (1950)
	A [†]	273.15/323.15	Gunn ¹² (1958)
	X	215.00/323.20	Hayden ² (1975)
	C [‡]	215.00/273.15	Hoover ¹⁴ (1968)
	X	273.2/323.2	Huff ⁵ (1963)
	A	298	Katayama ⁸ (1980)
	A	288.7	Mason ¹³ (1961)
	A	272/323	Michels ¹⁰ (1939)
CH ₄ -C ₃ H ₈	A	241.1/266.7	Wormald ¹¹ (1979)
	B [†]	298.15/373.15	Dantzler ⁹ (1968)
	X	298.15/373.15	Dantzler ⁹ (1968)
	A [‡]	310.90/510.90	Gunn ¹² (1958)
	X	298.20/510.90	Hayden ² (1975)
	X	310.9/510.9	Huff ⁵ (1963)
	A	288.7	Mason ¹³ (1961)
CH ₄ -n-C ₄ H ₁₀	A	243.2/302.2	Wormald ¹¹ (1979)
	B [‡]	423.2/573.2	Beattie ¹⁷ (1941)
	B [‡]	298.15/373.15	Dantzler ⁹ (1968)
	X	423/573	Guggenheim ⁴ (1950)
	X	298.15/373.15	Dantzler ⁹ (1968)
	A [†]	344.30/510.90	Gunn ¹² (1958)
	X	423.20/510.90	Hayden ² (1975)
	X	344.3/510.9	Huff ⁵ (1963)
CH ₄ -i-C ₄ H ₁₀	A	228.70	Mason ¹³ (1961)
	A [‡]	277.0/394.3	Wormald ¹¹ (1979)
	A [†]	344.3/510.9	Gunn ¹² (1958)
	X	344.30/510.90	Hayden ² (1975)
	X	344.3/510.9	Huff ⁵ (1963)
CH ₄ -n-C ₅ H ₁₂	C	288.7	Mason ¹³ (1961)
	A	344.30/510.90	Olds ¹⁸ (1942)
	B	298.15/373.15	Dantzler ⁹ (1968)
	X	298.15/323.20	Hayden ² (1975)
	X	310.9/510.9	Huff ⁵ (1963)
CH ₄ -n-C ₅ H ₁₂	C [‡]	288.7	Mason ¹³ (1961)
	C	298.15	Massoudi ²⁰ (1973)
	C	298.2/323.2	Pecsok ²¹ (1968)
	B	318.5/403.5	Wormald ¹¹ (1979)
	A	310.9/510.8	Zaalishvili ²² (1956)

[†]Data derived from information reported by another author.

[‡]Data vary in quality or rankings somewhat uncertain.

Table 2. (continued)

System	Data Rank	Temperature Range (K)	Source
CH ₄ -i-C ₅ H ₁₂	A [‡]	288.7	Mason ¹³ (1961)
	C	298.2/323.2	Pecsok ²¹ (1968)
CH ₄ -neo-C ₅ H ₁₂	C	199.99/257.90	Baughman ²⁶ (1975)
	C	300/550	Bellm ²⁵ (1974)
	C	303.16/403.16	Hamann ²⁴ (1955)
	X	303.16/403.16	Hayden ² (1975)
	X	303.2/403.2	Huff ⁵ (1963)
	A	296.15/492.6	Strein ²³ (1971)
CH ₄ -CO	X	288.70	Hayden ² (1975)
	A [‡]	288.7	Mason ¹³ (1961)
CH ₄ -CO ₂	X	310.9/510.8	Hayden ² (1975)
	X	310.9/510.8	Huff ⁵ (1963)
	A	298	Katayama ⁸ (1980)
	A	288.7	Mason ¹³ (1961)
	A	310.9/510.8	Zaalishvili ²² (1956)
CH ₄ -N ₂	A	288.7	Mason ¹³ (1961)
	A	155.88/291.40	Roe ²⁷ (1980)
CH ₄ -H ₂	X	288.7	Hayden ² (1975)
	A	288.7	Mason ¹³ (1961)
	A	144.3/283.2	Mueller ²⁸ (1961)
	A	273.15	Vilcu ⁶ (1977)
	X	273.15	Vilcu ⁶ (1977)
CH ₄ -H ₂ O	X	298.15/510.90	Hayden ² (1975)
	B	298.15/510.90	Rigby ³⁰ (1968)
CH ₄ -He	A	292.9/320.0	Bell ³² (1981)
CH ₄ -NH ₃	X	423.00/523.00	Hayden ² (1975)
	B [‡]	423.00/523.00	Lee ³³ (1970)
C ₂ H ₆ -C ₃ H ₈	A	298.15/373.15	Dantzler ⁹ (1968)
	X	298.20/373.20	Hayden ² (1975)
	A	288.7	Mason ¹³ (1961)
C ₂ H ₆ -n-C ₄ H ₁₀	A	298.15/373.15	Dantzler ⁹ (1968)
	X	298.20/373.20	Hayden ² (1975)
	A	288.7	Mason ¹³ (1961)
	C [‡]	304.5/363.2	Wormald ¹¹ (1979)
C ₂ H ₆ -n-C ₅ H ₁₂	A	298.15/373.15	Dantzler ⁹ (1968)
	X	298.20/373.20	Hayden ² (1975)
	C	298.15	Massoudi ²⁰ (1973)
	C	298.2	Pecsok ²¹ (1968)

Table 2. (continued)

System	Data Rank	Temperature Range (K)	Source
C ₂ H ₆ -CO ₂	X	310.9/510.8	Hayden ² (1975)
	X	310.9/510.8	Huff ⁵ (1963)
	A	298	Katayama ⁸ (1980)
	A	288.7	Mason ¹³ (1961)
	A [†]	310.9/510.8	Zaalishvili ²² (1956)
C ₂ H ₆ -N ₂	A ^{†,‡}	277.6/510.9	Gunn ¹² (1958)
	X	277.6/510.9	Hayden ² (1975)
	X	277.6/510.8	Huff ⁵ (1963)
	A	288.7	Mason ¹³ (1961)
C ₂ H ₆ -H ₂	X	288.2	Hayden ² (1975)
	A [†]	288.7	Mason ¹³ (1961)
C ₂ H ₆ -H ₂ O	C	298.15/373.15	Coan ³⁴ (1971)
	X	298.15/444.30	Hayden ² (1975)
	C	377.60/444.30	Rigby ³⁰ (1968)
C ₂ H ₆ -H ₂ S	X	323.20/398.20	Hayden ² (1975)
	B [†]	323.15/398.15	Khoury ³⁵ (1971)
C ₃ H ₈ -n-C ₄ H ₁₀	A	298.15/373.15	Dantzler ⁹ (1968)
	X	298.20/373.20	Hayden ² (1975)
	A	288.7	Mason ¹³ (1961)
C ₃ H ₈ -n-C ₅ H ₁₂	A	298.15/373.15	Dantzler ⁹ (1968)
	X	298.20/373.20	Hayden ² (1975)
C ₃ H ₈ -CO ₂	A	300.5	Bougard ³⁷ (1976)
	A ^{†,‡}	310.9/510.9	Gunn ¹² (1958)
	X	310.90/510.80	Hayden ² (1975)
	X	310.9/510.9	Huff ⁵ (1963)
	A	288.7	Mason ¹³ (1961)
	C	313.2	Sie ³⁸ (1966)
C ₃ H ₈ -N ₂	A [†]	288.7	Mason ¹³ (1961)
C ₃ H ₈ -H ₂	X	288.20	Hayden ² (1975)
	A [†]	288.7	Mason ¹³ (1961)
C ₃ H ₈ -He	B [†]	393.18/423.00	Warowny ³⁹ (1978)
n-C ₄ H ₁₀ -i-C ₄ H ₁₀	A	344.26/444.26	Connolly ⁴⁰ (1962)
	X	344.30/444.30	Hayden ² (1975)
n-C ₄ H ₁₀ -n-C ₅ H ₁₂	A	298.15/373.15	Dantzler ⁹ (1968)
	X	298.20/373.20	Hayden ² (1975)
	C	303.15	Jessen ⁴¹ (1938)

Table 2. (continued)

System	Data Rank	Temperature Range (K)	Source
n-C ₄ H ₁₀ -CO ₂	A [†]	377.6/477.6	Gunn ¹² (1958)
	X	377.60/477.60	Hayden ² (1975)
	X	377.6/477.6	Huff ⁵ (1963)
	A [‡]	288.7	Mason ¹³ (1961)
	C	313.2	Sie ³⁸ (1966)
n-C ₄ H ₁₀ -N ₂	B	308.2	Cruikshank ⁴² (1968)
	A [†]	427.6/477.6	Gunn ¹² (1958)
	X	427.60/477.60	Hayden ² (1975)
	X	427.6/477.6	Huff ⁵ (1963)
	B	303.2/323.2	Hicks ⁴⁴ (1968)
	A [‡]	288.7	Mason ¹³ (1961)
n-C ₄ H ₁₀ -H ₂	B	333.2	Young ⁴⁵ (1968)
	X	288.20	Hayden ² (1975)
n-C ₄ H ₁₀ -He	A [‡]	288.7	Mason ¹³ (1961)
	B [‡]	373.3/498.3	Jones ⁴⁶ (1967)
n-C ₅ H ₁₂ -CO ₂	C	298.2	Desty ⁴⁸ (1962)
	B	298.15	Massoudi ²⁰ (1973)
	C	313.2	Sie ³⁸ (1966)
	C	353.2	Vigdergauz ⁴⁷ (1971)
n-C ₅ H ₁₂ -N ₂	B [‡]	298.2	Cruikshank ⁵⁰ (1966)
	B	308.15	Cruikshank ⁴² (1968)
	C	298.2	Desty ⁴⁸ (1962)
	B [‡]	298.2	Everett ⁵¹ (1965)
	B	313.2	Gainey ⁵² (1967)
	B	313.2	Gainey ⁵³ (1970)
	X	298.20	Hayden ² (1975)
	B	313.2/333.2	Hicks ⁴⁴ (1968)
	C	308.15	Letcher ⁴⁹ (1974)
	B [‡]	298.16	Leung ⁵⁴ (1974)
	B [‡]	298.15	Massoudi ²⁰ (1973)
	C	353.2	Vigdergauz ⁴⁷ (1971)
	C	328.2/338.2	Young ⁴⁵ (1968)
n-C ₅ H ₁₂ -H ₂	B	298.2	Cruikshank ⁵⁰ (1966)
	B	298.2	Desty ⁴⁸ (1962)
	B	298.2	Everett ⁵¹ (1965)
	X	298.20	Hayden ² (1975)
	A [‡]	288.7	Mason ¹³ (1961)
i-C ₅ H ₁₂ -CO ₂	C	298.2	Desty ⁴⁸ (1962)

Table 2. (continued)

System	Data Rank	Temperature Range (K)	Source
i-C ₅ H ₁₂ -N ₂	C [†]	298.2	Desty ⁴⁸ (1962)
	B [†]	313.2	Gainey ⁵³ (1970)
	C	328.2/338.2	Young ⁴⁵ (1968)
i-C ₅ H ₁₂ -H ₂	C [†]	298.2	Desty ⁴⁸ (1962)
neo-C ₅ H ₁₂ -N ₂	B	199.99/257.86	Baughman ²⁶ (1975)
neo-C ₅ H ₁₂ -He	C	199.99/230.00	Baughman ²⁶ (1975)
CO-CO ₂	B	303.2/363.2	Cottrell ⁵⁵ (1956)
	X	303.20/363.20	Hayden ² (1975)
CO-H ₂	X	272/290	Guggenheim ⁴ (1950)
	X	36.00/60.00	Hayden ² (1975)
	A	36/60	Reuss ⁵⁶ (1956)
	A	298.2	Scott ⁵⁸ (1929)
	A	273.2/298.2	Townend ⁵⁷ (1932)
CO ₂ -CS ₂	-	273.2	Eucken ⁶⁰ (1928)
CO ₂ -N ₂	B	303.2/363.2	Cottrell ⁵⁵ (1956)
	A	298.2	Edwards ⁶¹ (1942)
	A	305.15	Gorski ⁶² (1953)
	A [†]	298.15/398.15	Gunn ¹² (1958)
	X	298.20/363.20	Hayden ² (1975)
	X	298.2/398.2	Huff ⁵ (1963)
	A	228.7	Mason ¹³ (1961)
	A	303.2	Pfefferle ⁶³ (1944)
B [†]	110.0/125.0	Yakimenko ⁶⁴ (1977)	
CO ₂ -H ₂	X	303.20/363.20	Hayden ² (1975)
	A [†]	303.2/363.2	Cottrell ⁵⁵ (1956)
	C	298.2	Edwards ⁶¹ (1942)
CO ₂ -H ₂ O	X	298.20/373.20	Hayden ² (1975)
	B	298.15/373.15	Coan ³⁴ (1971)
	C	323/343.2	Pollitzer ⁶⁶ (1924)
CO ₂ -He	B [†]	292.9/320.0	Bell ³² (1981)
	B [†]	303.2/363.2	Cottrell ⁶⁵ (1956)
	C	298.2	Edwards ⁶¹ (1942)
	B [†]	303.2	Harper ⁶⁸ (1957)
	X	303.20/363.20	Hayden ² (1975)
	B [†]	323.2	Linshits ⁶⁹ (1975)
	B [†]	303.2	Pfefferle ⁶³ (1944)
	B [†]	373.2	Tsiklis ⁷⁰ (1974a)
	B [†]	423.2	Tsiklis ⁷¹ (1974b)

Table 2. (continued)

System	Data Rank	Temperature Range (K)	Source
CS ₂ -N ₂	-	272.2	Eucken ⁶⁰ (1928)
CS ₂ -H ₂	-	273.2	Eucken ⁶⁰ (1928)
N ₂ -H ₂	A	148.15/323.15	Brewer ⁷² (1969)
	A	298.2	Edwards ⁶¹ (1942)
	X	273/293	Guggenheim ⁴ (1950)
	X	148.20/293.20	Hayden ² (1975)
	A	298.16	Lunbeck ⁷³ (1951)
	A	298.16	Michels ⁷⁴ (1949)
	B [†]	198.15/273.15	Ostronov ⁷⁵ (1967)
	B [†]	36/60	Reuss ⁵⁶ (1956)
	A	273.2/293.2	Verschoye ⁷⁶ (1926)
	A	298.16	Vilcu ⁷ (1974)
	X	298.16	Vilcu ⁷ (1974)
A	170/270	Zandbergen ⁷⁷ (1967)	
N ₂ -H ₂ O	X	298.20/373.20	Hayden ² (1975)
	A	373.15/423.15	Richards ⁷⁸ (1979)
	A	298.15/373.15	Rigby ³⁰ (1968)
N ₂ -He	A	292.9/321.7	Bell ³² (1981)
	A	148.15/323.15	Brewer ⁷² (1969)
	A	133.15/273.15	Canfield ⁸² (1963)
	B	298.2	Edwards ⁶¹ (1942)
	B [†]	103.15/113.15	Hall ⁸³ (1969)
	X	148.20/293.20	Hayden ² (1975)
	C	90	Knobler ⁸⁴ (1959)
	A	303.20	Kramer ⁸⁵ (1957)
	A	311.7/373.2	Ku ⁸⁶ (1967)
A	303.2	Pfefferle ⁶³ (1944)	
B [†]	448.2/748.2	Witonsky ⁸⁷ (1963)	
N ₂ -NH ₃	X	318.00/723.00	Hayden ² (1975)
	B [†]	318.00/723.00	Lee ³³ (1970)
N ₂ O-H ₂ O	X	298.20/373.20	Hayden ² (1975)
	A	298.15/373.15	Coan ³⁴ (1971)
H ₂ -He	A	20.4	Beenakker ⁹¹ (1959)
	A	148.15/323.15	Brewer ⁷² (1969)
	A	298.2/448.2	Gibby ⁹² (1929)
	X	298/448	Huft ⁵ (1963)
	X	90.00/323.20	Hayden ² (1975)
	B [†]	90	Knobler ⁸⁴ (1959)
	A	298.2	Tanner ⁹³ (1930)
	A	14/17	Varekamp ⁹⁴ (1959)

Table 3
Recommended Unlike Interaction Second Virial Coefficients

System	Temperature Range (K)	Recommended B_{12} Values	Data Rank
CH ₄ -C ₂ H ₆	241.1/266.7	Wormald ¹¹	A
	273/323	Michels ¹⁰	A
	288.7	Mason ¹³	A
	298	Katayama ⁸	A
	298.15/373.15	Dantzler ⁹	A
CH ₄ -C ₃ H ₈	243.2/302.2	Wormald ¹¹	A
	310.9/510.9	Gunn ¹²	A
CH ₄ -n-C ₄ H ₁₀	277.0/394.3	Wormald ¹¹	A
	344.30/510.90	Gunn ¹²	A
	423.2/573.2	Beattie ¹⁷	B
CH ₄ -i-C ₄ H ₁₀	344.3/510.9	Olds ¹⁸	A
CH ₄ -n-C ₅ H ₁₂	298.15/373.15	Dantzler ⁹	B
	310.9/510.8	Zaalishvili ²²	A
CH ₄ -i-C ₅ H ₁₂	288.7	Mason ¹³	A
CH ₄ -neo-C ₅ H ₁₂	296.15/492.6	Strein ²³	A
CH ₄ -CO	288.7	Mason ¹³	A
CH ₄ -CO ₂	288.7	Mason ¹³	A
	298	Katayama ⁸	A
	310.9/510.8	Zaalishvili ²²	A
CH ₄ -N ₂	155.88/291.40	Roe ²⁷	A
CH ₄ -H ₂	144.3/283.2	Mueller ²⁸	A
	288.7	Mason ¹³	A
CH ₄ -H ₂ O	298.15/510.9	Rigby ³⁰	B
CH ₄ -He	292.9/320.0	Bell ³²	A
CH ₄ -NH ₃	423.00/523.00	Lee ³³	B
C ₂ H ₆ -C ₃ H ₈	288.7	Mason ¹³	A
	298.15/373.15	Dantzler ⁹	A
C ₂ H ₆ -n-C ₄ H ₁₀	288.7	Mason ¹³	A
	298.15/373.15	Dantzler ⁹	A
C ₂ H ₆ -n-C ₅ H ₁₂	298.15/373.15	Dantzler ⁹	A
C ₂ H ₆ -CO ₂	288.7	Mason ¹³	A
	298	Katayama ⁸	A
	310.9/510.9	Zaalishvili ²²	A
C ₂ H ₆ -N ₂	277.6/510.9	Gunn ¹²	A
C ₂ H ₆ -H ₂	288.7	Mason ¹³	A
C ₂ H ₆ -H ₂ S	323.15/398.15	Khoury ³⁵	B

Table 3. (continued)

System	Temperature Range (K)	Recommended B_{12} Values	Data Rank
C_3H_8 -n- C_4H_{10}	288.7	Mason ¹³	A
	298.15/373.15	Dantzler ⁹	A
C_3H_8 -n- C_5H_{12}	298.15/373.15	Dantzler ⁹	A
C_3H_8 -CO ₂	288.7	Mason ¹³	A
	300.5	Bougard ³⁷	A
	310.9/510.9	Gunn ¹²	A
C_3H_8 -N ₂	288.7	Mason ¹³	A
C_3H_8 -H ₂	288.7	Mason ¹³	A
C_3H_8 -He	393.18/423.00	Warowny ³⁹	B
n- C_4H_{10} -i- C_4H_{10}	344.26/444.26	Connolly ⁴⁰	A
n- C_4H_{10} -n- C_5H_{12}	298.15/373.15	Dantzler ⁹	A
n- C_4H_{10} -CO ₂	288.7	Mason ¹³	A
	377.6/477.6	Gunn ¹²	A
n- C_4H_{10} -N ₂	288.7	Mason ¹³	A
	303.2/323.2	Hicks ⁴⁴	B
	427.6/477.6	Gunn ¹²	A
n- C_4H_{10} -H ₂	288.7	Mason ¹³	A
n- C_4H_{10} -He	373.3/498.3	Jones ⁴⁶	B
n- C_5H_{12} -CO ₂	298.15	Massoudi ²⁰	B
	298.15	Massoudi ²⁰	B
n- C_5H_{12} -N ₂	298.16	Leung ⁵⁴	B
	298.2	Cruikshank ⁵⁰	B
	298.2	Everett ⁵¹	B
	308.15	Cruikshank ⁴²	B
	313.2	Gainey ⁵²	B
	313.2	Gainey ⁵³	B
	313.2/333.2	Hicks ⁴⁴	B
	298.2	Mason ¹³	A
n- C_5H_{12} -H ₂	298.2	Cruikshank ⁵⁰	B
	298.2	Desty ⁴⁸	B
	298.2	Everett ⁵¹	B
	298.2	Everett ⁵¹	B
i- C_5H_{12} -N ₂	313.2	Gainey ⁵³	B
neo- C_5H_{12} -N ₂	199.99/257.86	Baughman ²⁶	B
CO-CO ₂	303.2/363.2	Cottrell ⁵⁵	B
CO-H ₂	36/60	Reuss ⁵⁶	A
	273.2/298.2	Townend ⁵⁷	A

Table 3. (continued)

System	Temperature Range (K)	Recommended B_{12} Values	Data Rank
CO ₂ -N ₂	110.0/125.0	Yakimenko ⁶⁴	B
	288.7	Mason ¹³	A
	298.15/398.15	Gunn ¹²	A
CO ₂ -H ₂	303.2/363.2	Cottrell ⁵⁵	A
CO ₂ -H ₂ O	298.15/373.15	Coan ³⁴	B
CO ₂ -He	292.9/320.0	Bell ³²	B
	303.2/363.2	Cottrell ⁵⁵	B
	373.2	Tsiklis ⁷⁰	B
	423.2	Tsiklis ⁷¹	B
N ₂ -H ₂	36/60	Reuss ⁵⁶	B
	148.15/323.15	Brewer ⁷²	A
	170/270	Zandbergen ⁷⁷	A
N ₂ -H ₂ O	298.15/373.15	Rigby ³⁰	A
	373.15/423.15	Richards ⁷⁸	A
N ₂ -He	103.15/113.15	Hall ⁸³	B
	133.15/273.15	Canfield ⁸²	A
	148.15/323.15	Brewer ⁷²	A
	311.7/373.2	Ku ⁸⁶	A
	448.2/748.2	Witonsky ⁸⁷	B
N ₂ -NH ₃	318.00/723.00	Lee ³³	B
H ₂ O-N ₂ O	298.15/373.15	Coan ³⁴	A
H ₂ -He	14/17	Varekamp ⁹⁴	A
	20.4	Beenakker ⁹¹	A
	90	Knobler ⁸⁴	B
	148.15/323.15	Brewer ⁷²	A
	298.2/448.2	Gibby ⁹²	A

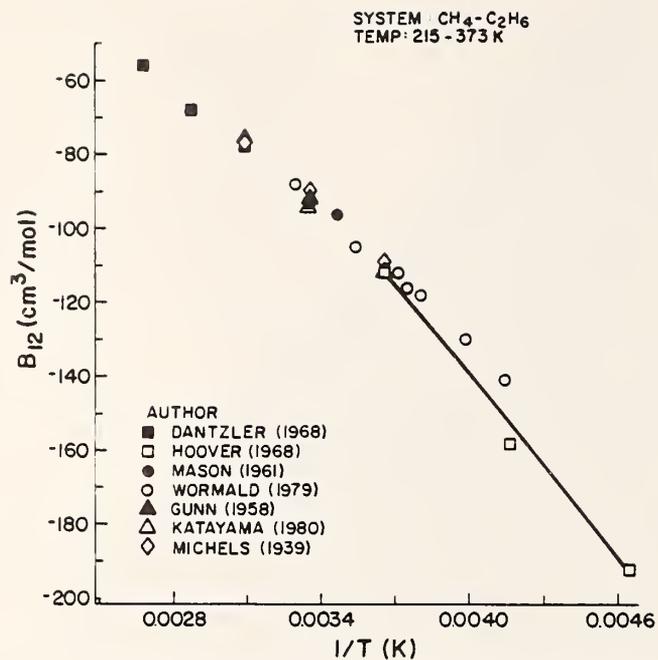


Figure 3. Temperature dependence of interaction second virial coefficient for the CH₄-C₂H₆ system.

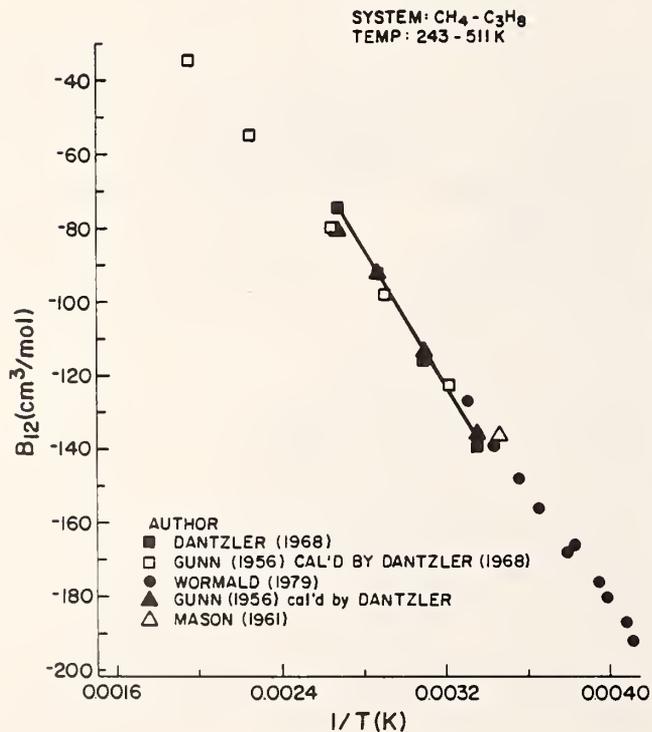


Figure 4. Temperature dependence of interaction second virial coefficient for the CH₄-C₃H₈ system.

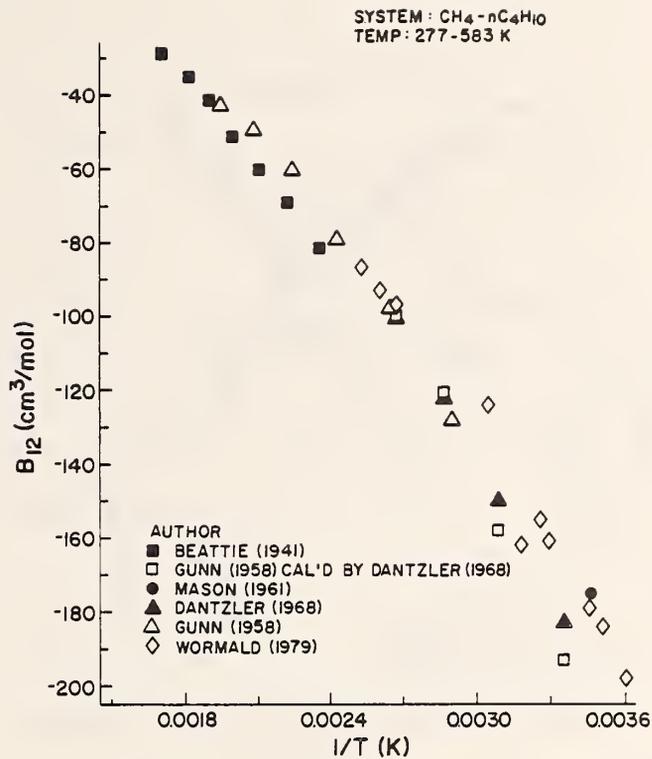


Figure 5. Temperature dependence of interaction second virial coefficient for the CH₄-n-C₄H₁₀ system.

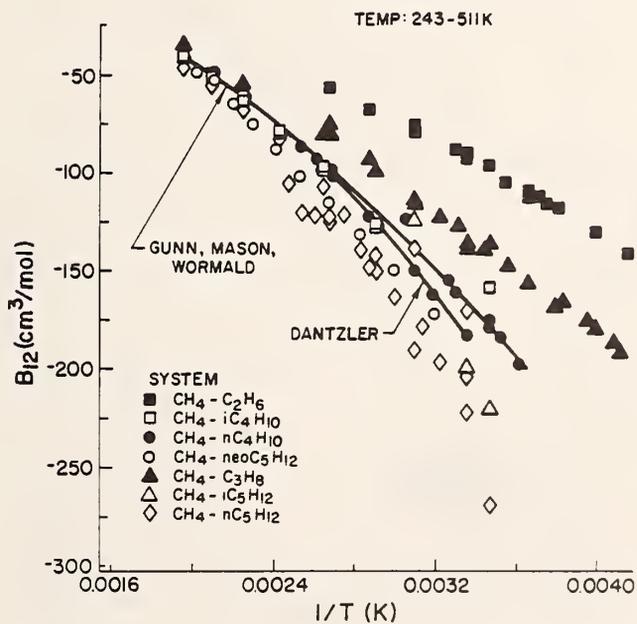


Figure 6. Temperature dependence of interaction second virial coefficient for methane-paraffin hydrocarbon binary systems.

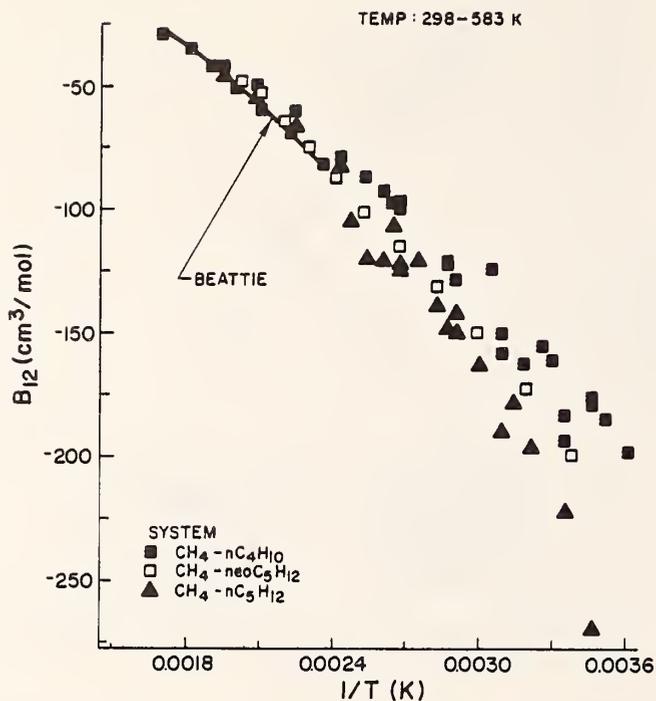


Figure 7. Temperature dependence of interaction second virial coefficient for methane-paraffin hydrocarbon binary systems.

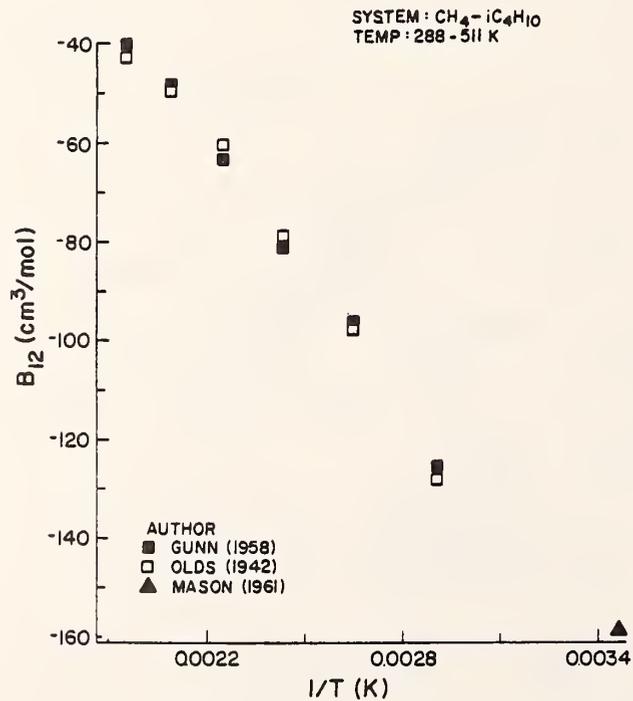


Figure 8. Temperature dependence of interaction second virial coefficient for the CH₄-i-C₄H₁₀ system.

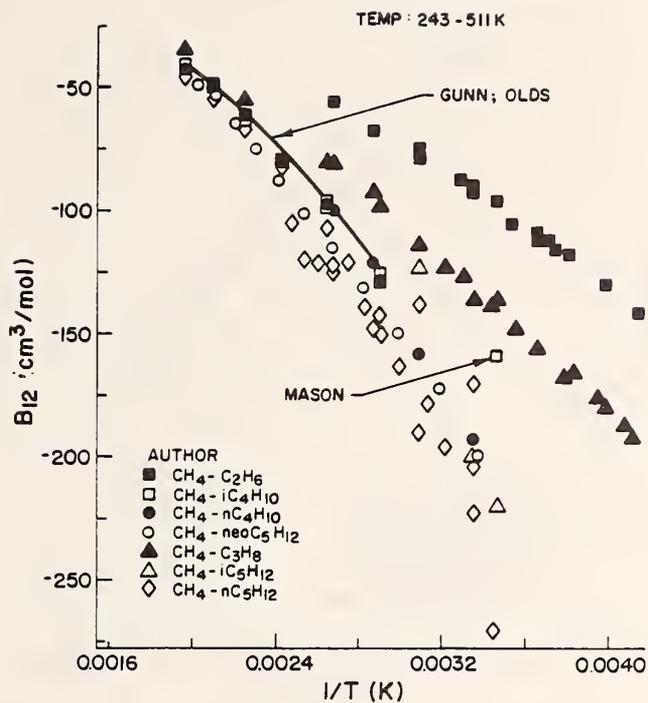


Figure 9. Temperature dependence of interaction second virial coefficient for methane-paraffin hydrocarbon binary systems.

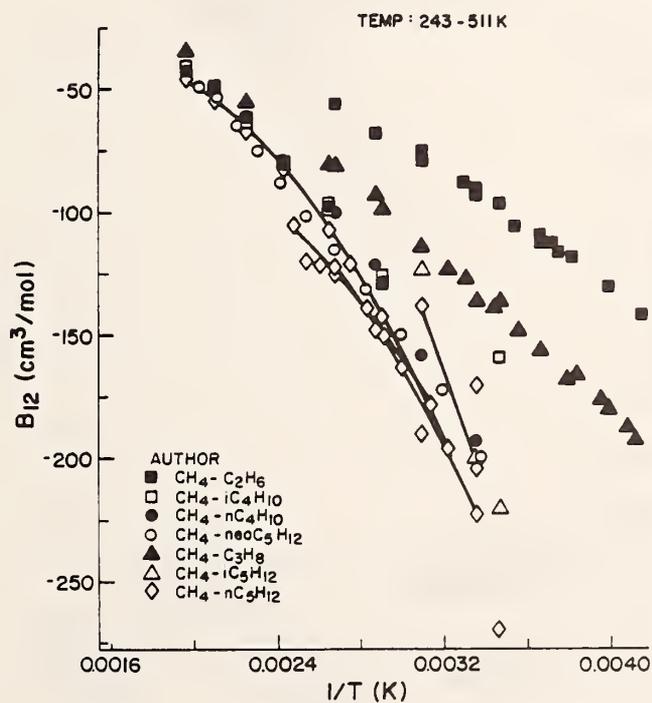


Figure 10. Temperature dependence of interaction second virial coefficient for methane-paraffin hydrocarbon binary systems.

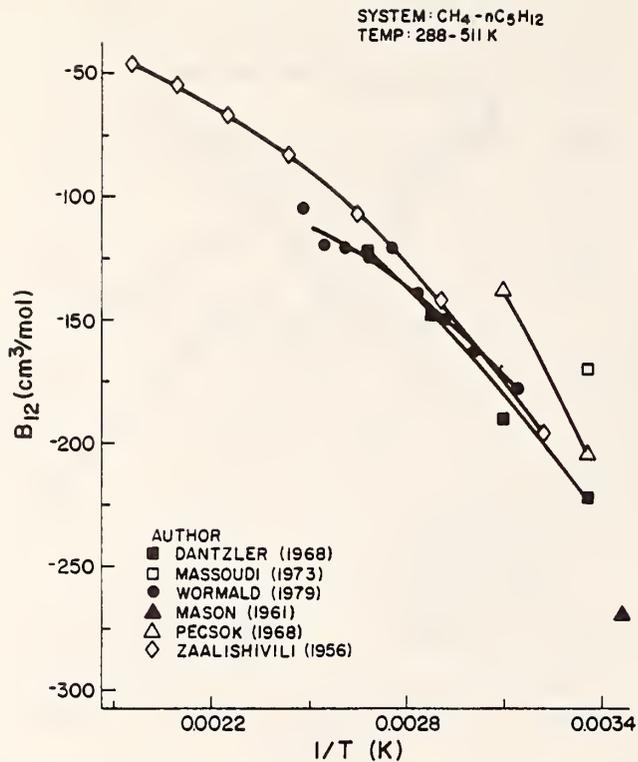


Figure 11. Temperature dependence of interaction second virial coefficient for the CH₄-n-C₅H₁₂ system.

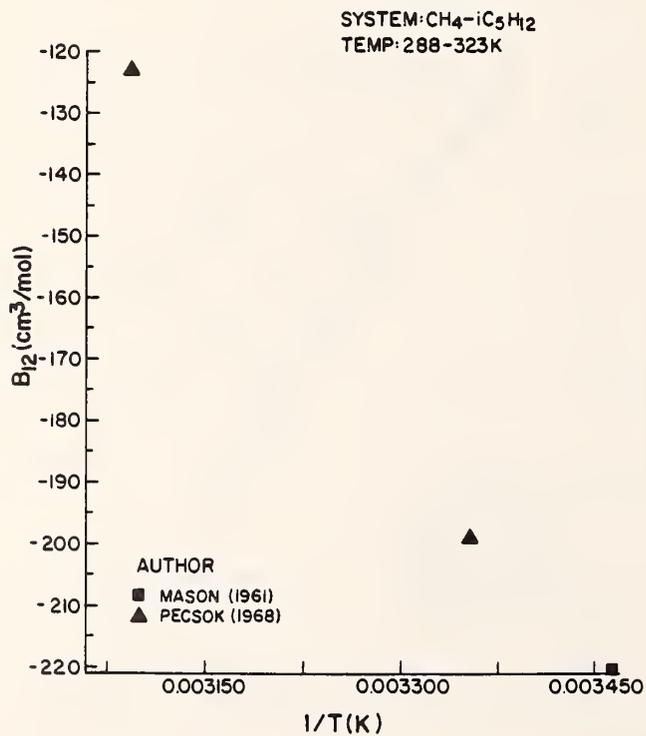


Figure 12. Temperature dependence of interaction second virial coefficient for the CH₄-i-C₅H₁₂ system.

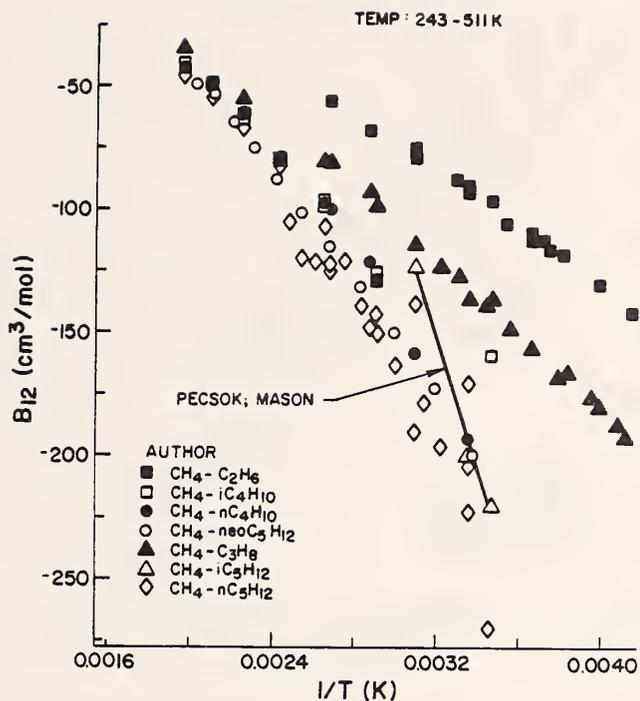


Figure 13. Temperature dependence of interaction second virial coefficient for methane-paraffin hydrocarbon binary systems.

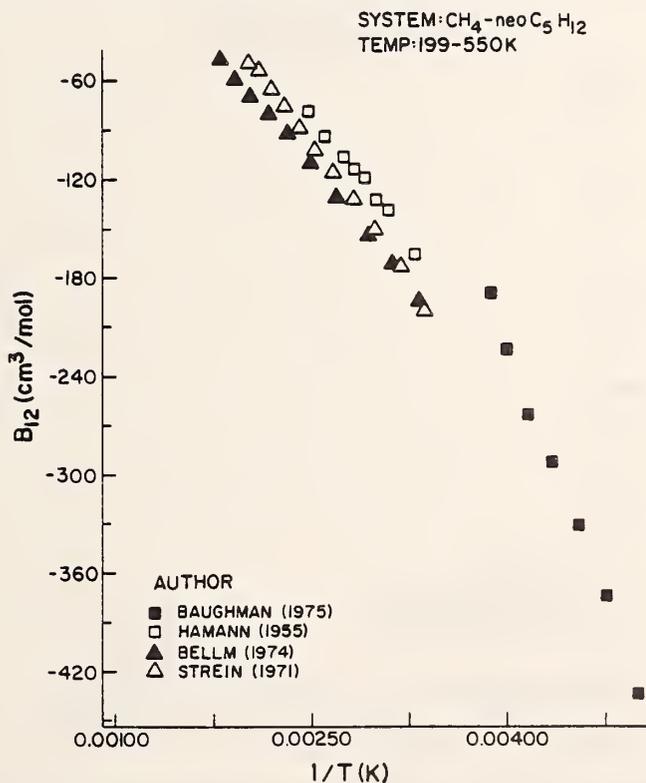


Figure 14. Temperature dependence of interaction second virial coefficient for the CH₄-neo-C₅H₁₂ system.

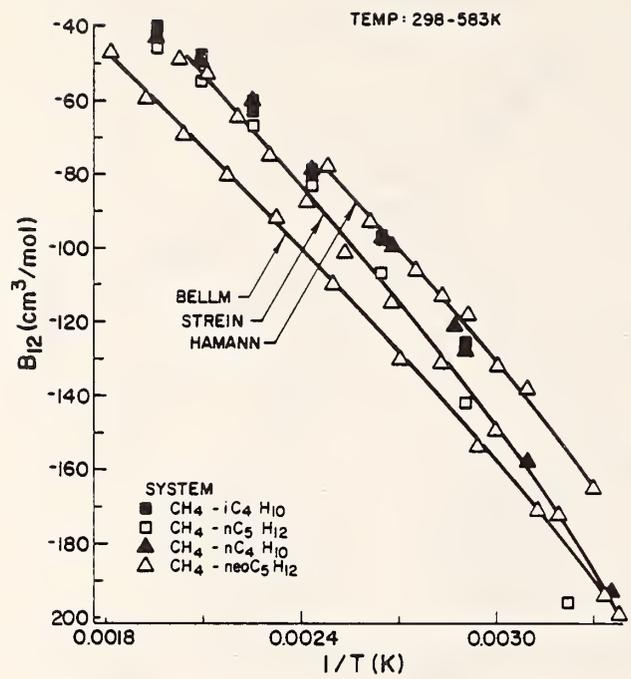


Figure 15. Temperature dependence of interaction second virial coefficient for methane-paraffin hydrocarbon binary systems.

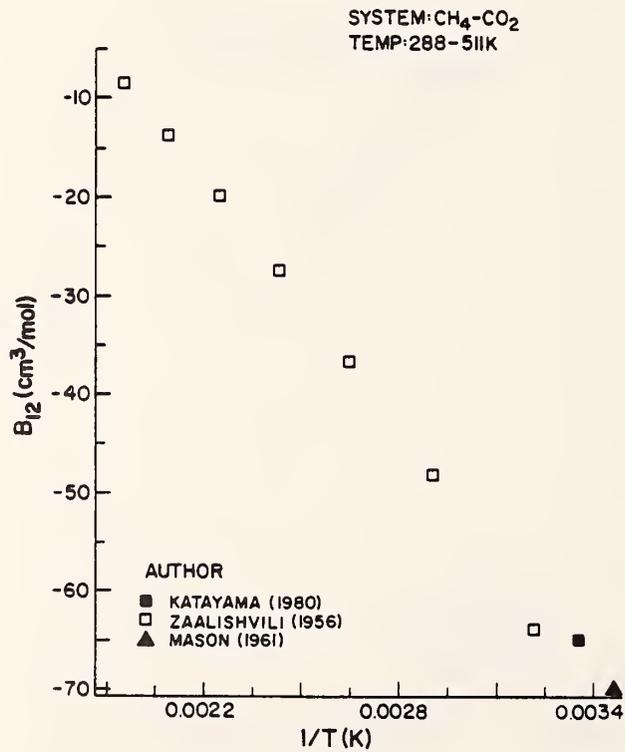


Figure 16. Temperature dependence of interaction second virial coefficient for the CH₄-CO₂ system.

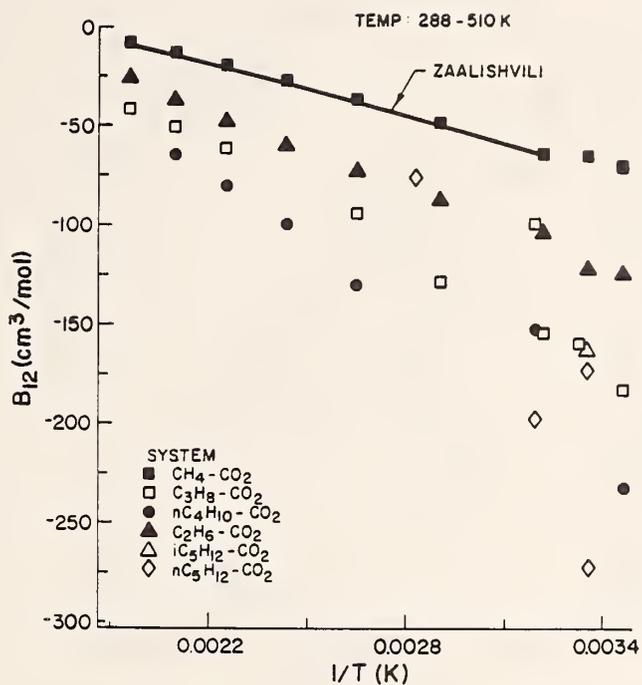


Figure 17. Temperature dependence of interaction second virial coefficient for carbon dioxide-paraffin hydrocarbon binary systems.

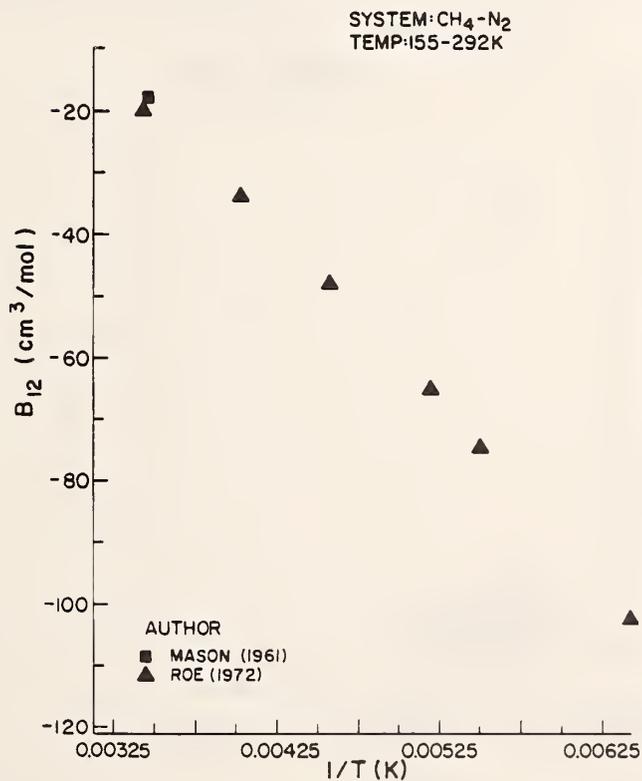


Figure 18. Temperature dependence of interaction second virial coefficient for the $\text{CH}_4 - \text{N}_2$ system.

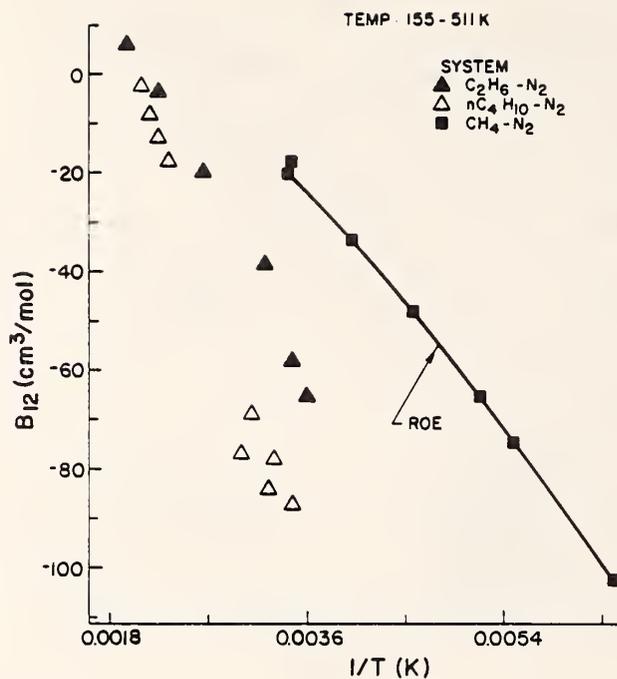


Figure 19. Temperature dependence of interaction second virial coefficient for nitrogen-paraffin hydrocarbon binary systems.

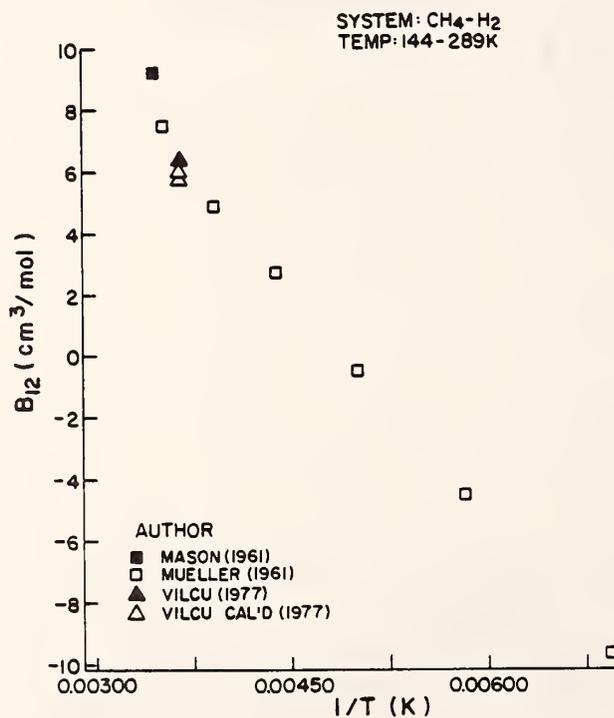


Figure 20. Temperature dependence of interaction second virial coefficient for the $\text{CH}_4\text{-H}_2$ system.

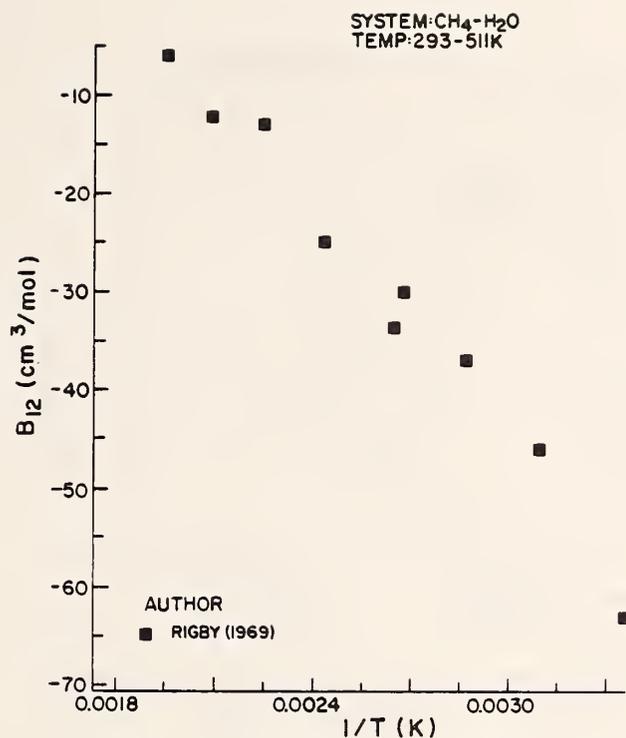


Figure 21. Temperature dependence of interaction second virial coefficient for the CH₄-H₂O system.

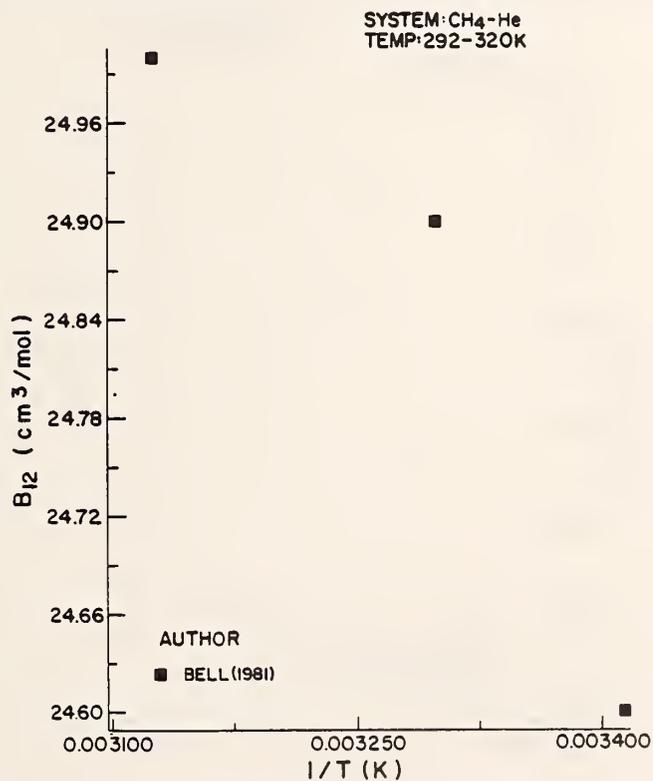


Figure 22. Temperature dependence of interaction second virial coefficient for the CH₄-He system.

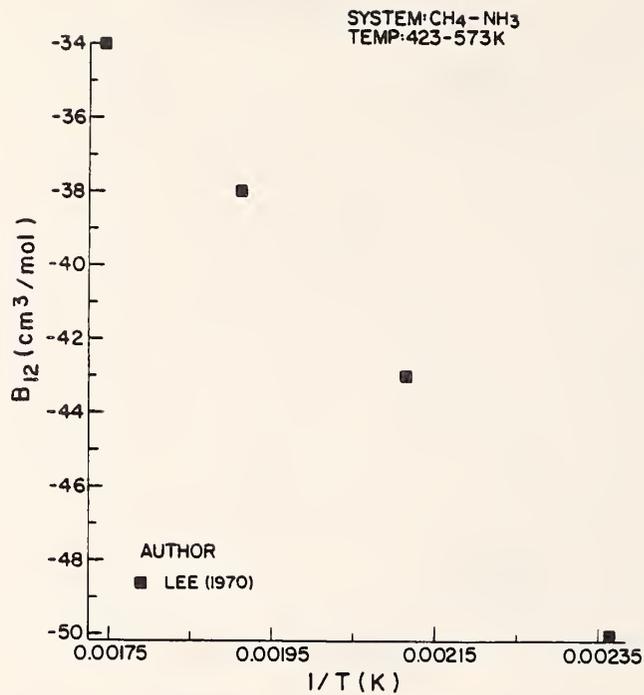


Figure 23. Temperature dependence of interaction second virial coefficient for the CH₄-NH₃ system.

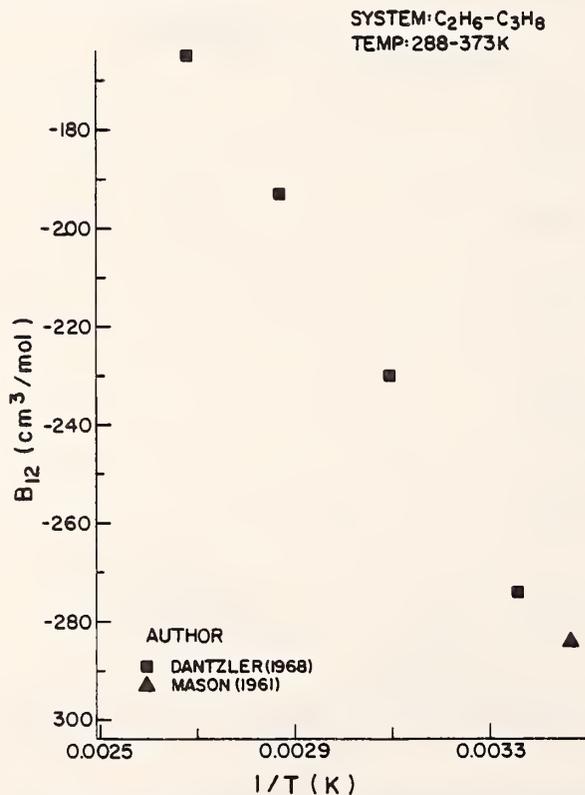


Figure 24. Temperature dependence of interaction second virial coefficient for the C₂H₆-C₃H₈ system.

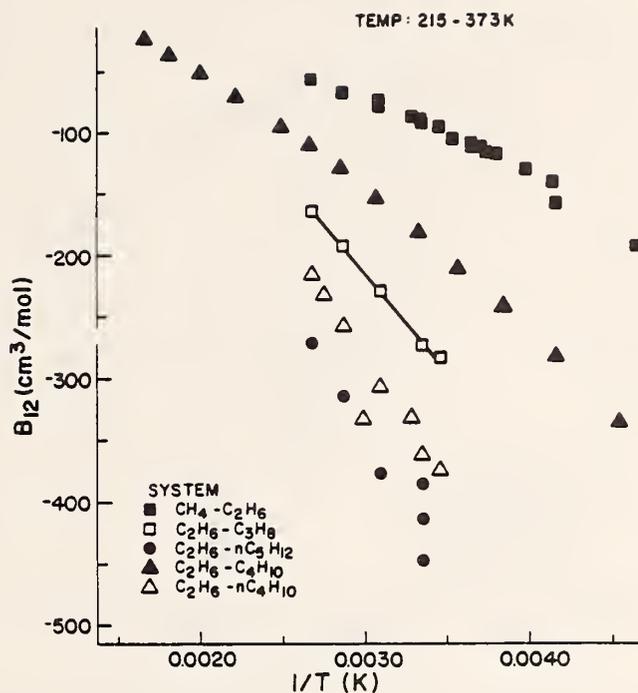


Figure 25. Temperature dependence of interaction second virial coefficient for ethane-paraffin hydrocarbon binary systems.

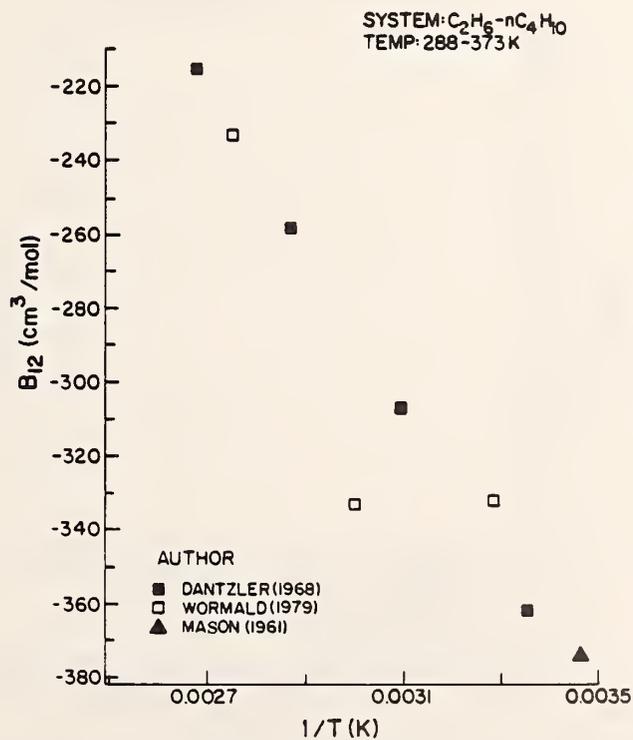


Figure 26. Temperature dependence of interaction second virial coefficient for the C₂H₆-nC₄H₁₀ system.

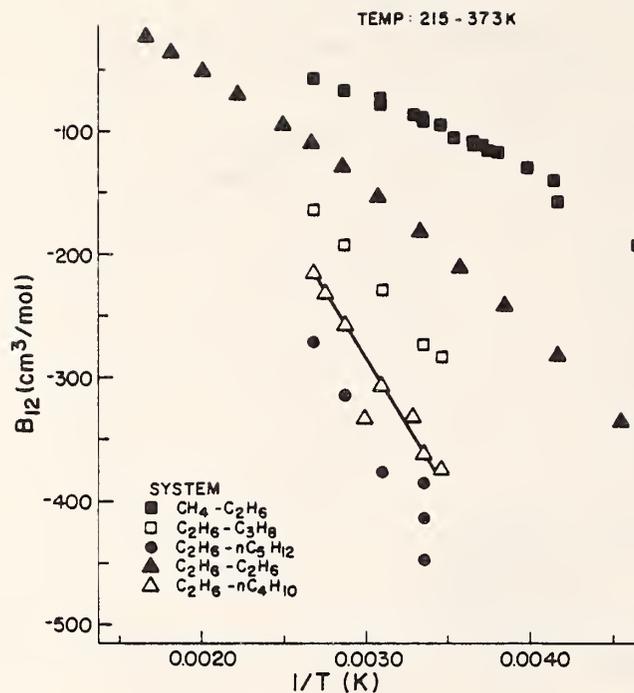


Figure 27. Temperature dependence of interaction second virial coefficient for ethane-paraffin hydrocarbon binary systems.

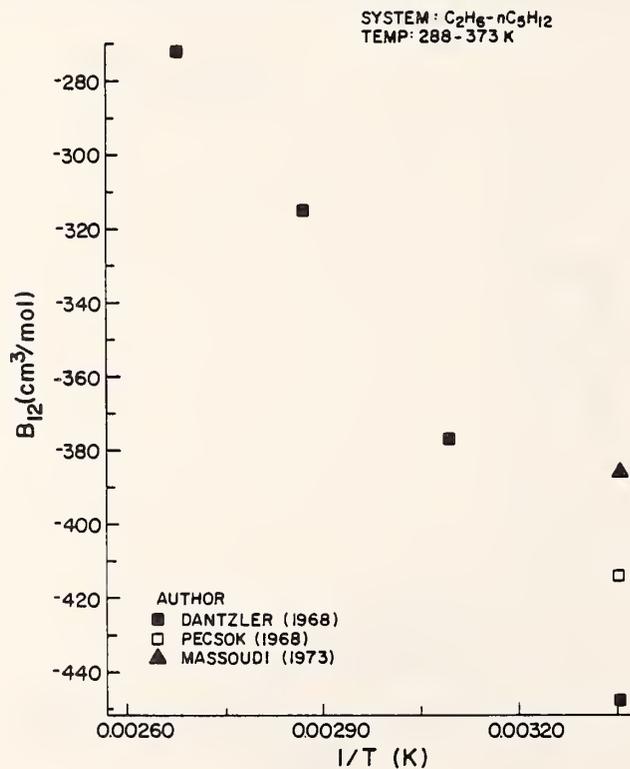


Figure 28. Temperature dependence of interaction second virial coefficient for the C₂H₆-n-C₅H₁₂ system.

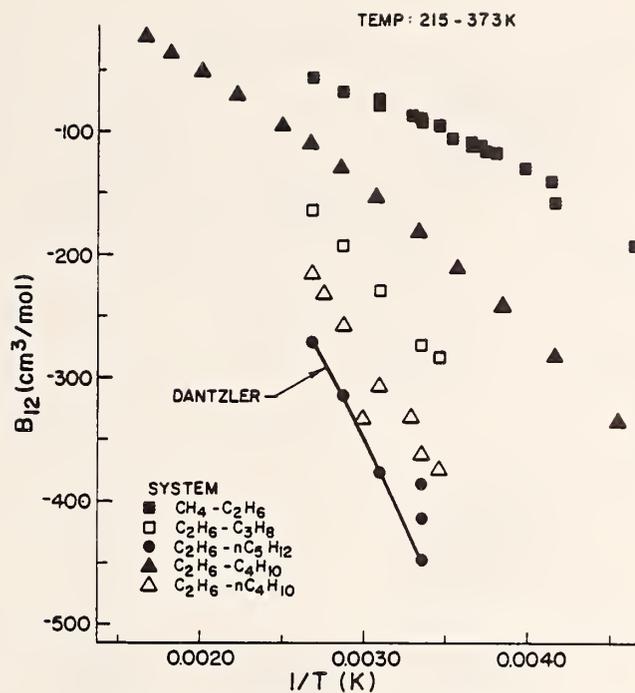


Figure 29. Temperature dependence of interaction second virial coefficient for ethane-paraffin hydrocarbon binary systems.

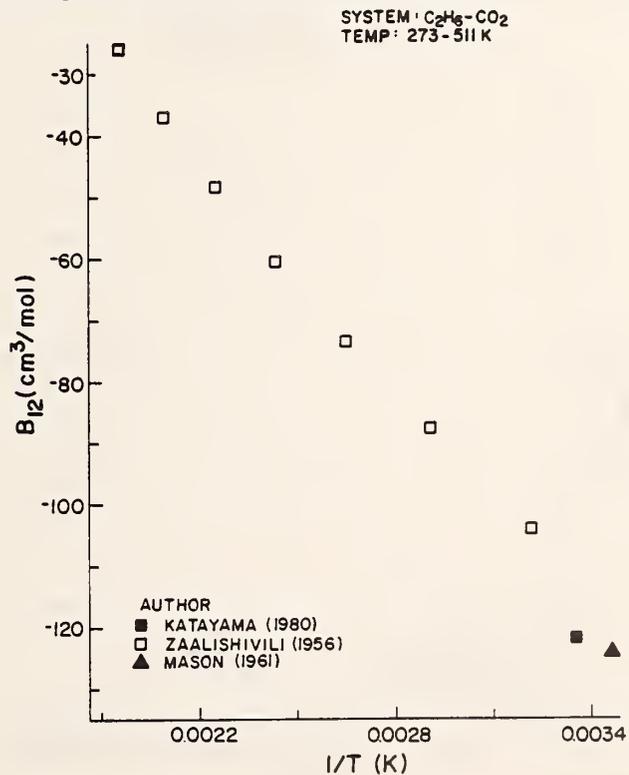


Figure 30. Temperature dependence of interaction second virial coefficient for the $\text{C}_2\text{H}_6 - \text{CO}_2$ system.

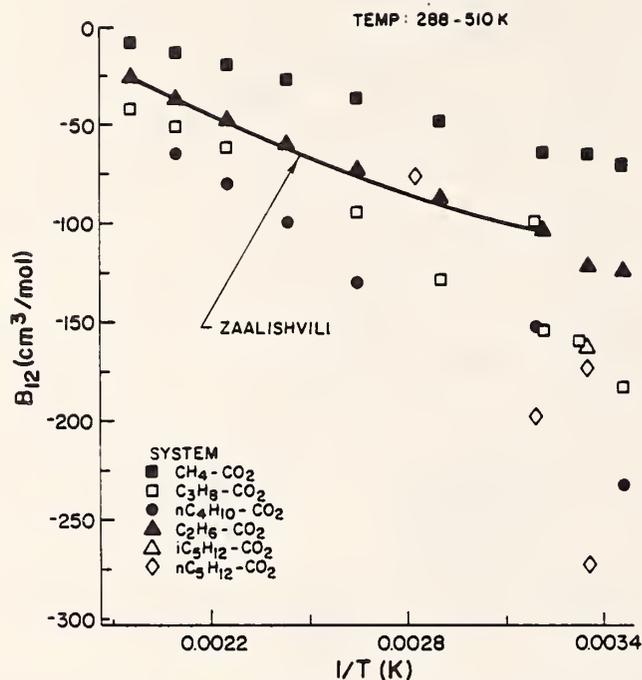


Figure 31. Temperature dependence of interaction second virial coefficient for carbon dioxide-paraffin hydrocarbon binary systems.

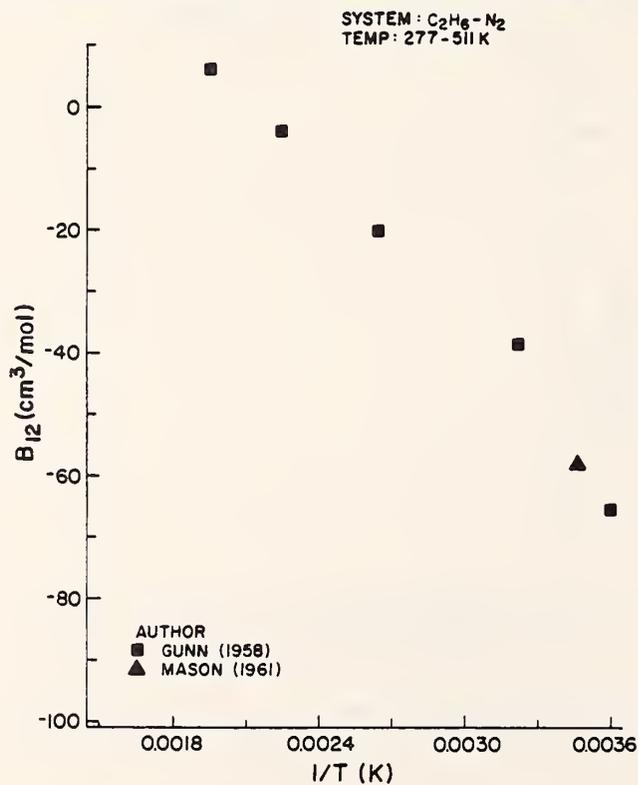


Figure 32. Temperature dependence of interaction second virial coefficient for the C₂H₆-N₂ system.

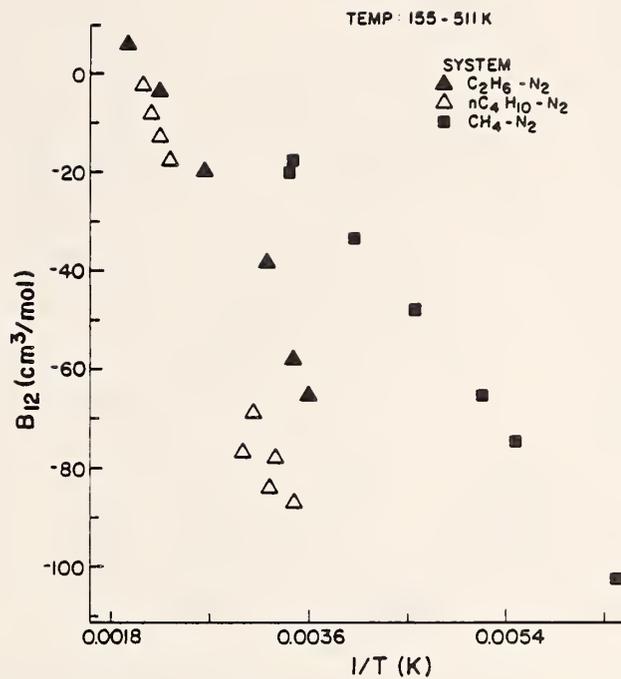


Figure 33. Temperature dependence of interaction second virial coefficient for nitrogen-paraffin hydrocarbon binary systems.

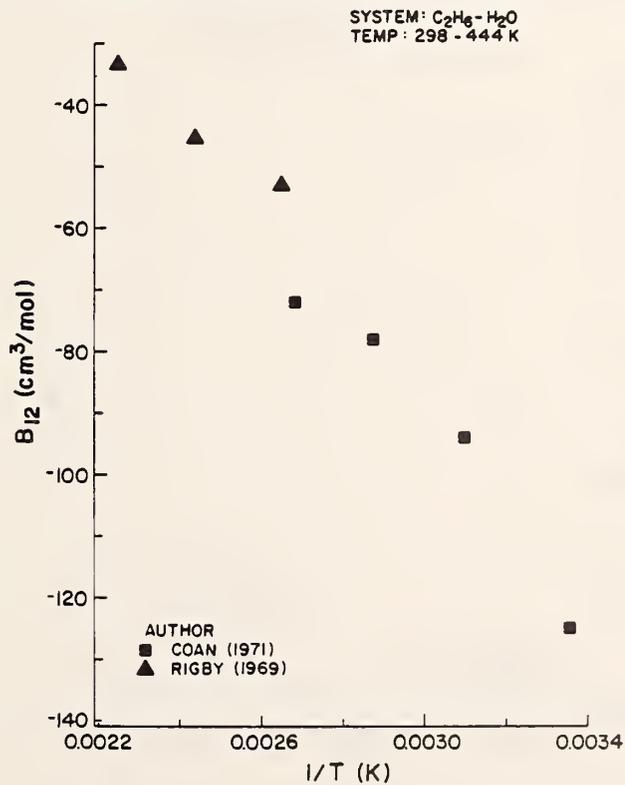


Figure 34. Temperature dependence of interaction second virial coefficient for the C₂H₆-H₂O system.

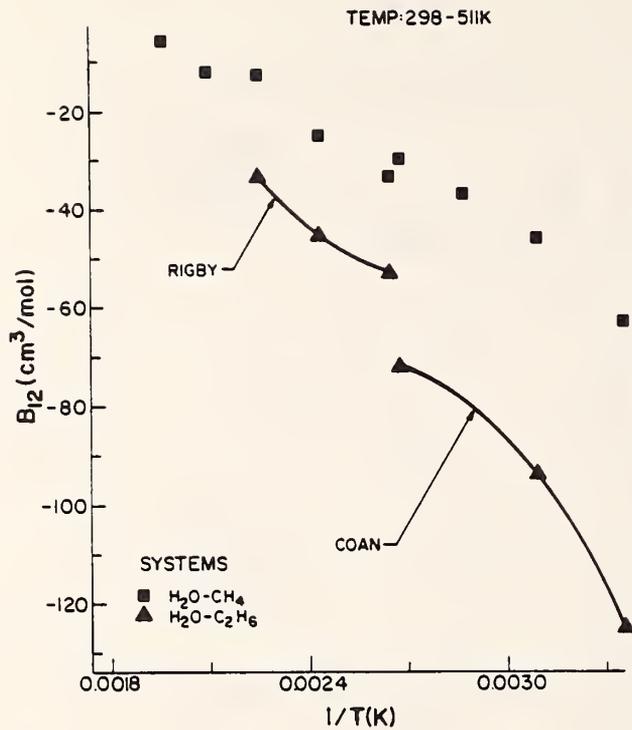


Figure 35. Temperature dependence of interaction second virial coefficient for water-paraffin hydrocarbon binary systems.

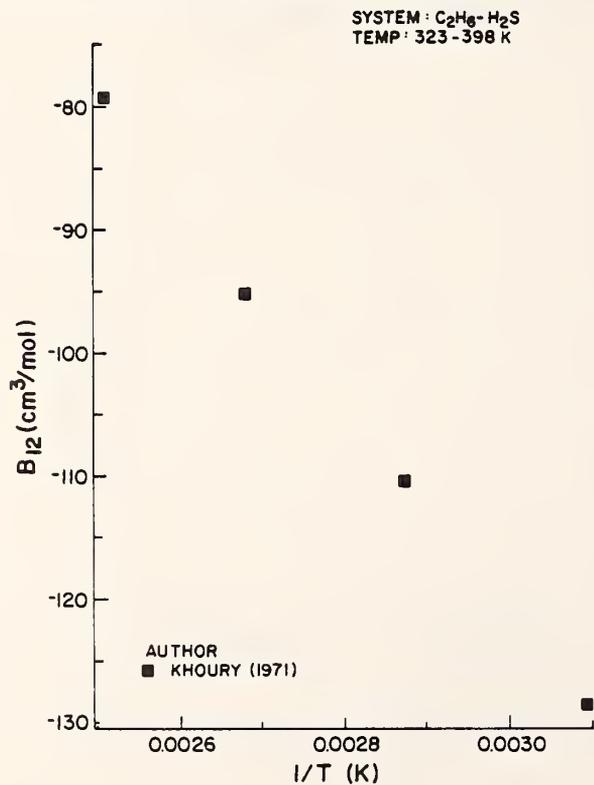


Figure 36. Temperature dependence of interaction second virial coefficient for the C₂H₆-H₂S system.

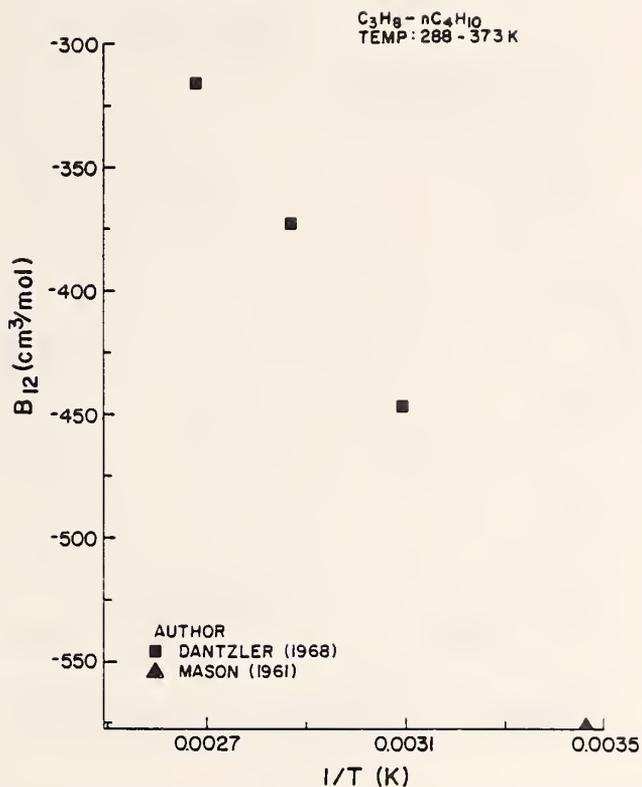


Figure 37. Temperature dependence of interaction second virial coefficient for the $C_3H_8-nC_4H_{10}$ system.

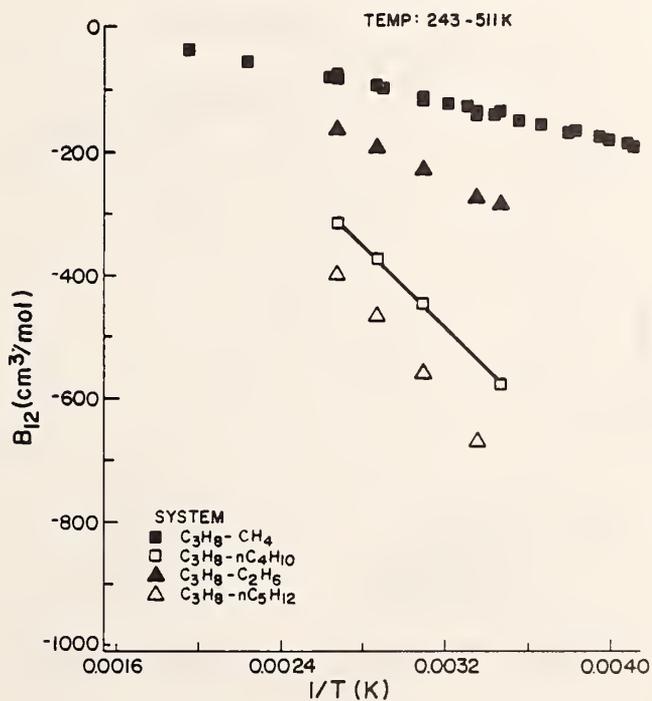


Figure 38. Temperature dependence of interaction second virial coefficient for propane-paraffin hydrocarbon binary systems.

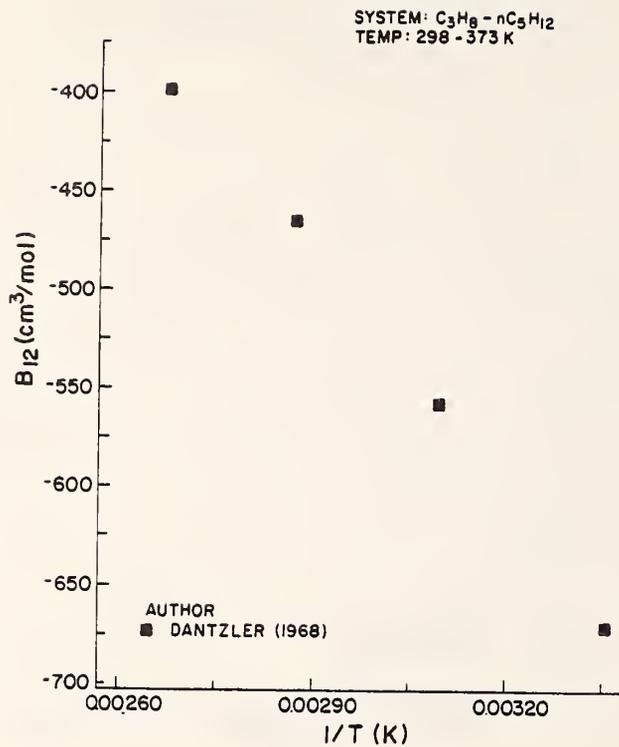


Figure 39. Temperature dependence of interaction second virial coefficient for the C₃H₈-nC₅H₁₂ system.

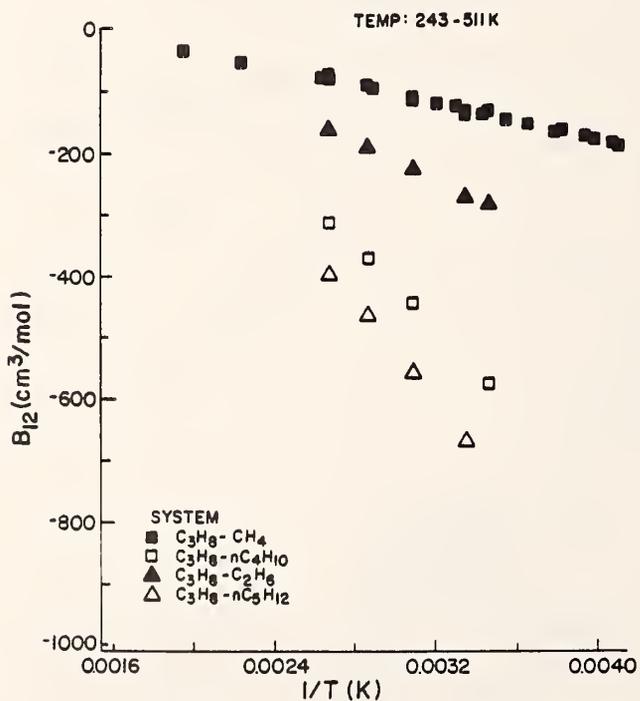


Figure 40. Temperature dependence of interaction second virial coefficient for propane-paraffin hydrocarbon binary systems.

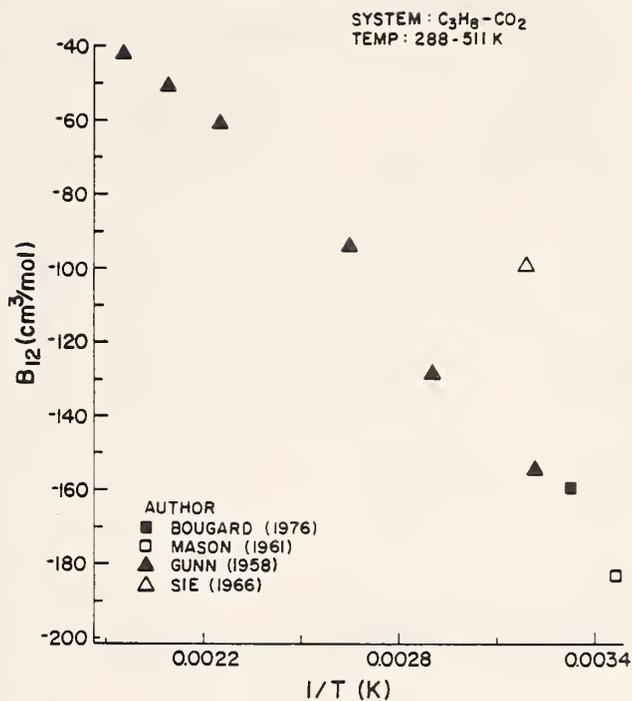


Figure 41. Temperature dependence of interaction second virial coefficient for the C₃H₈-CO₂ system.

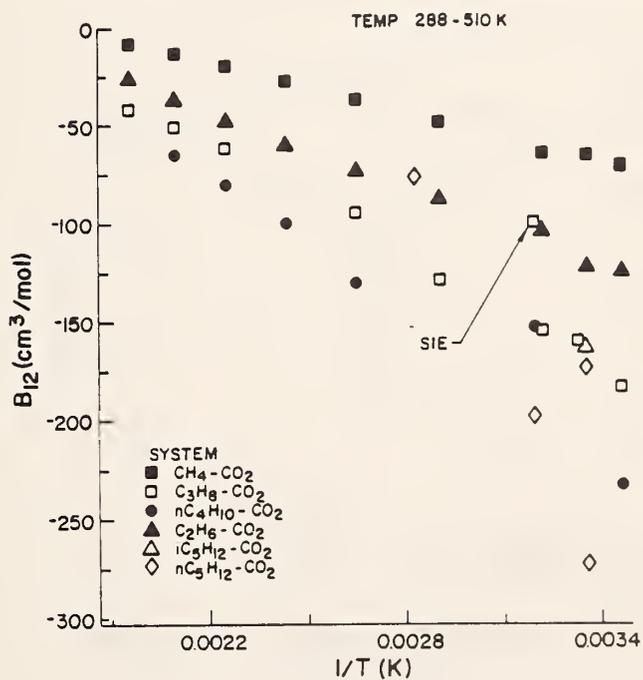


Figure 42. Temperature dependence of interaction second virial coefficient for carbon dioxide-paraffin hydrocarbon binary systems.

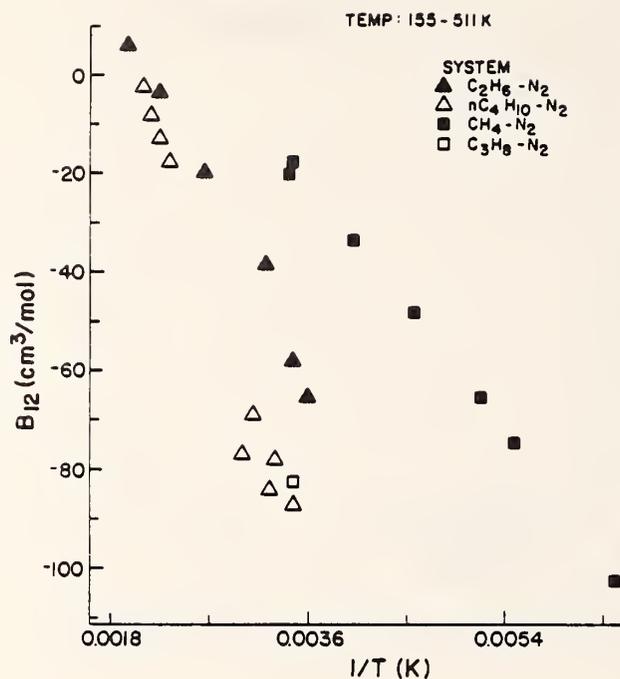


Figure 43. Temperature dependence of interaction second virial coefficient for nitrogen-paraffin hydrocarbon binary systems.

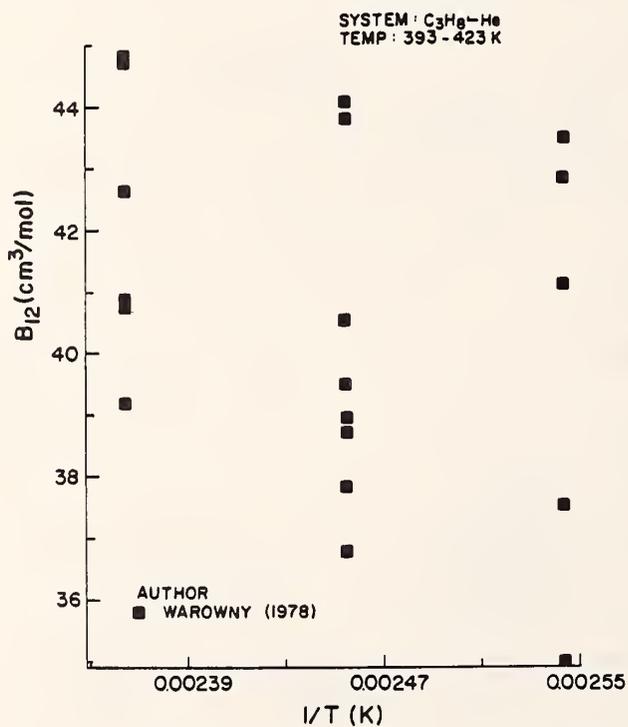


Figure 44. Temperature dependence of interaction second virial coefficient for the C₃H₈-He system.

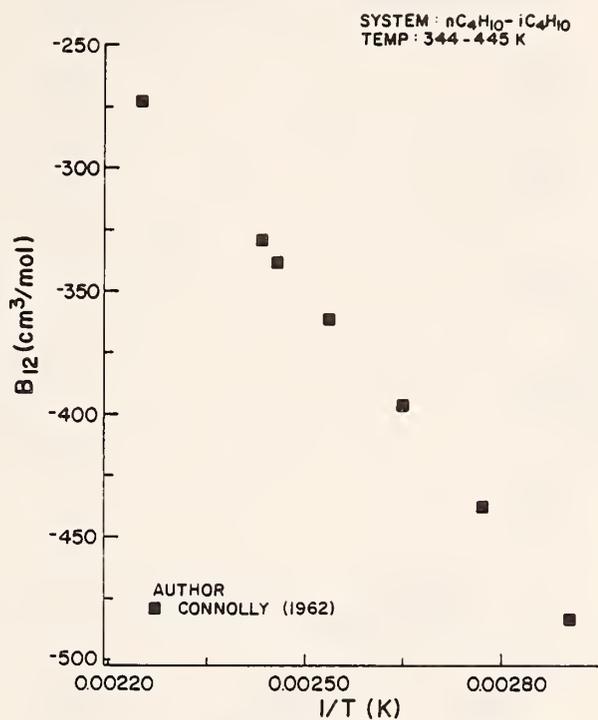


Figure 45. Temperature dependence of interaction second virial coefficient for the n-C₄H₁₀-i-C₄H₁₀ system.

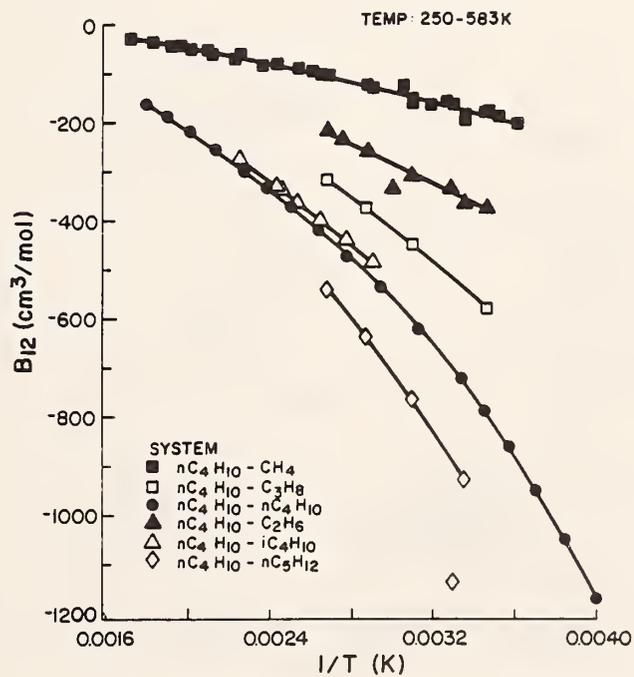


Figure 46. Temperature dependence of interaction second virial coefficient for n-butane-paraffin hydrocarbon binary systems.

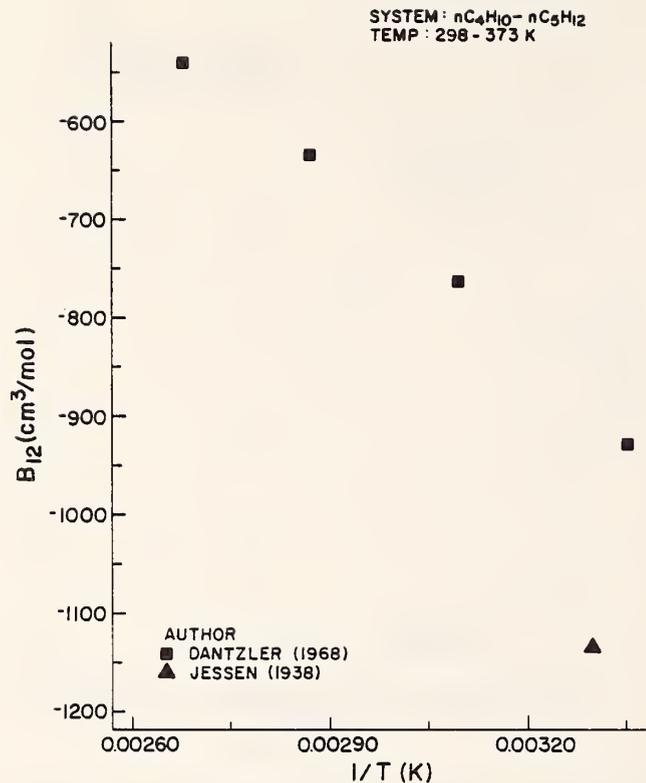


Figure 47. Temperature dependence of interaction second virial coefficient for the n-C₄H₁₀-n-C₅H₁₂ system.

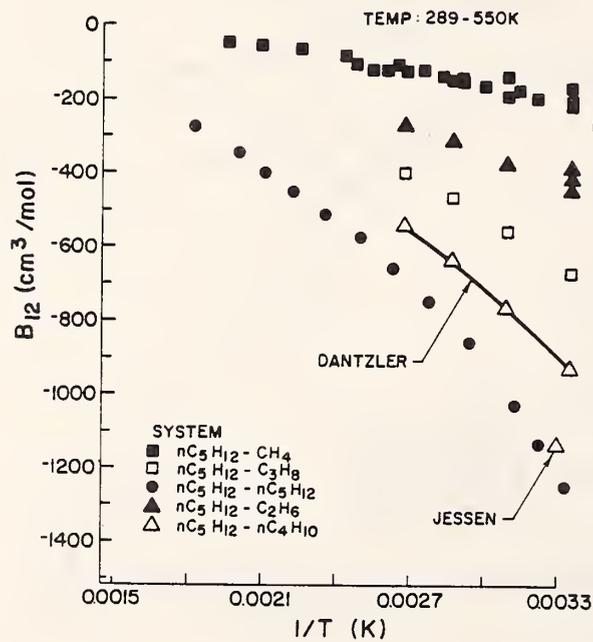


Figure 48. Temperature dependence of interaction second virial coefficient for n-pentane-paraffin hydrocarbon binary systems.

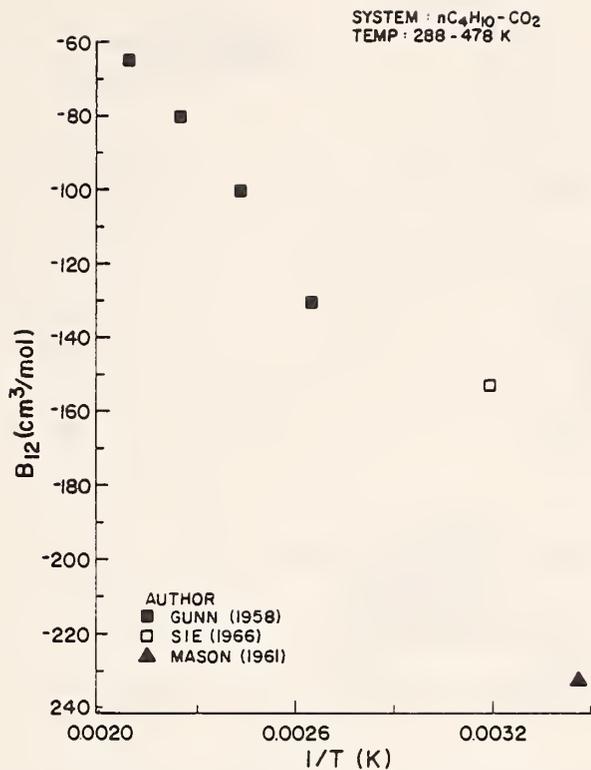


Figure 49. Temperature dependence of interaction second virial coefficient for the n-C₄H₁₀-CO₂ system.

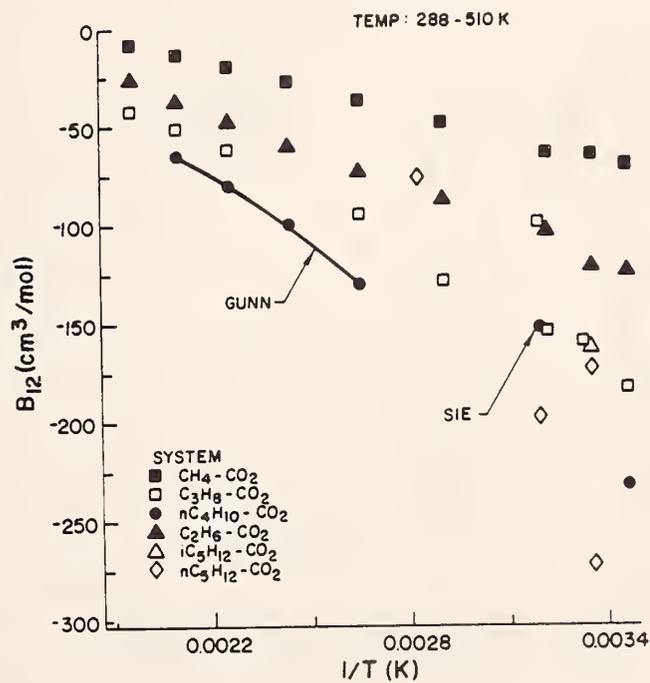


Figure 50. Temperature dependence of interaction second virial coefficient for carbon dioxide-paraffin hydrocarbon binary systems.

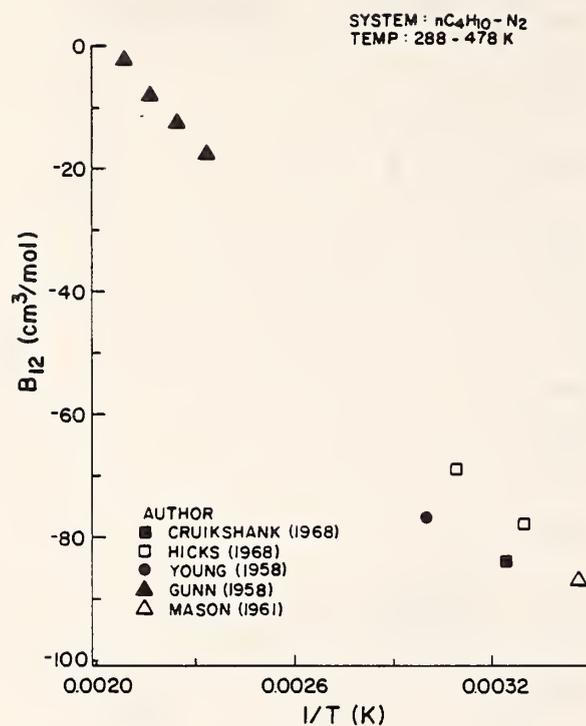


Figure 51. Temperature dependence of interaction second virial coefficient for the n-C₄H₁₀-N₂ system.

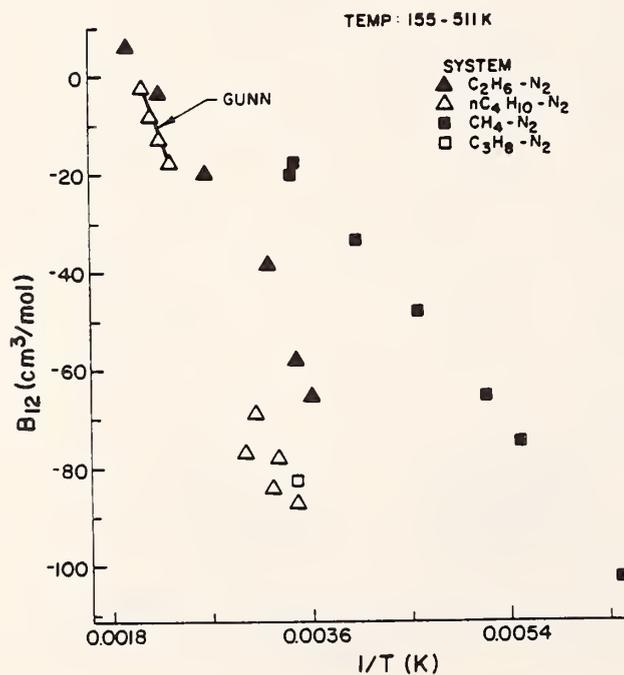


Figure 52. Temperature dependence of interaction second virial coefficient for nitrogen-paraffin hydrocarbon binary systems.

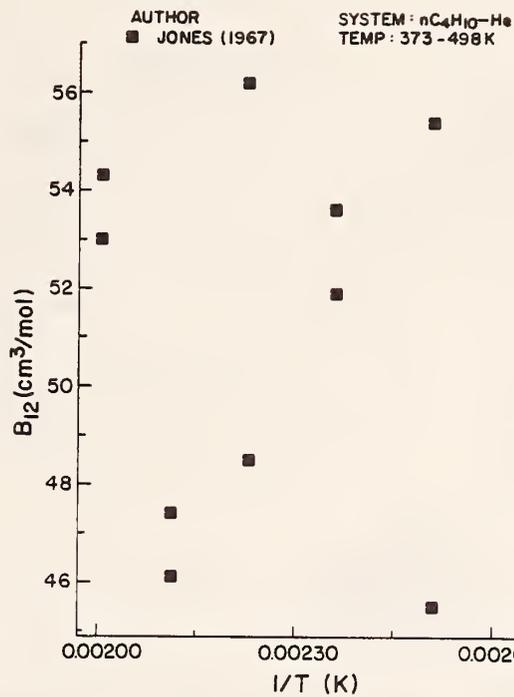


Figure 53. Temperature dependence of interaction second virial coefficient for the n-C₄H₁₀-He system.

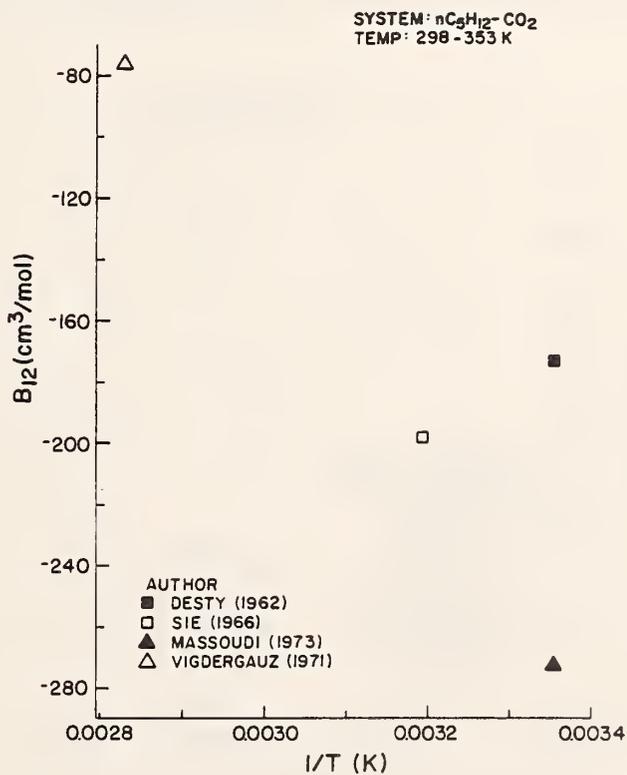


Figure 54. Temperature dependence of interaction second virial coefficient for the n-C₅H₁₂-CO₂ system.

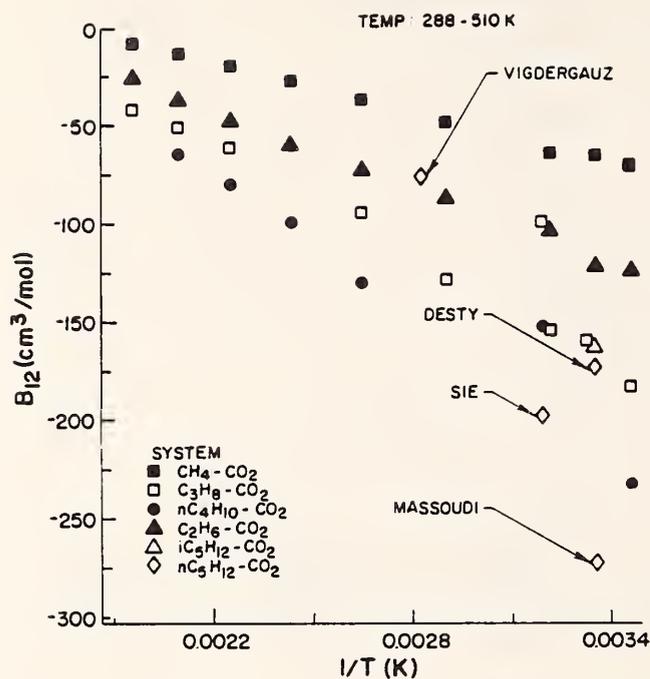


Figure 55. Temperature dependence of interaction second virial coefficient for carbon dioxide-paraffin hydrocarbon binary systems.

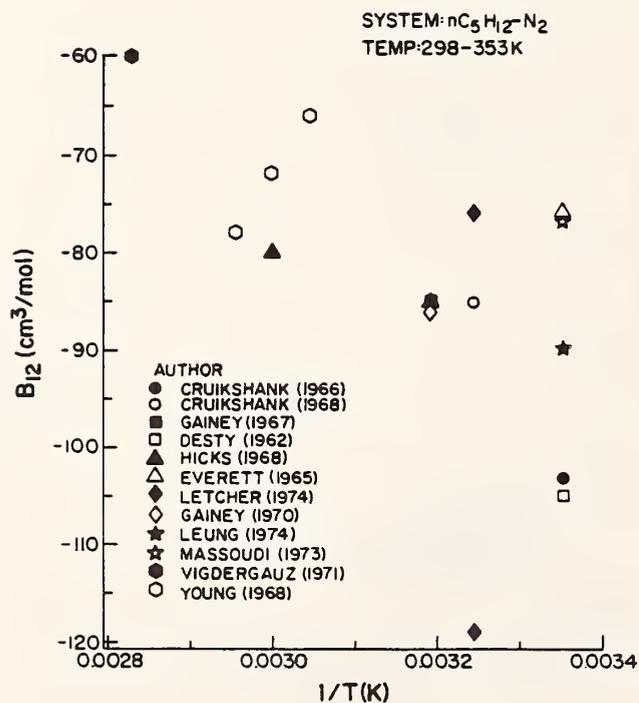


Figure 56. Temperature dependence of interaction second virial coefficient for the n-C₅H₁₂-N₂ system.

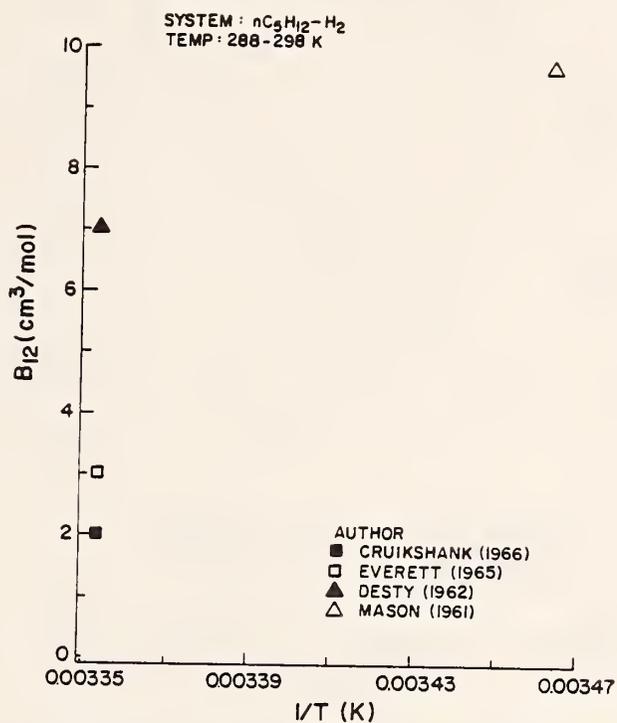


Figure 57. Temperature dependence of interaction second virial coefficient for the n-C₅H₁₂-H₂ system.

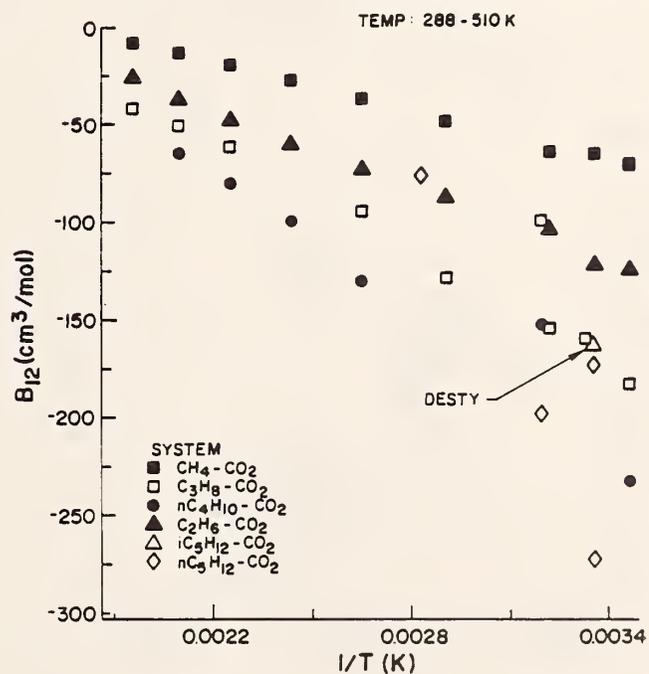


Figure 58. Temperature dependence of interaction second virial coefficient for carbon dioxide-paraffin hydrocarbon binary systems.

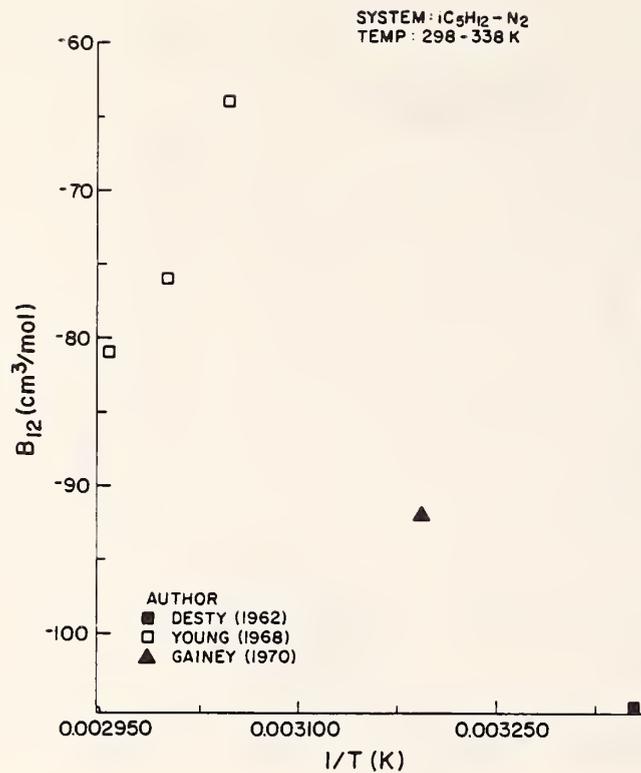


Figure 59. Temperature dependence of interaction second virial coefficient for the $i\text{-C}_5\text{H}_{12}\text{-N}_2$ system.

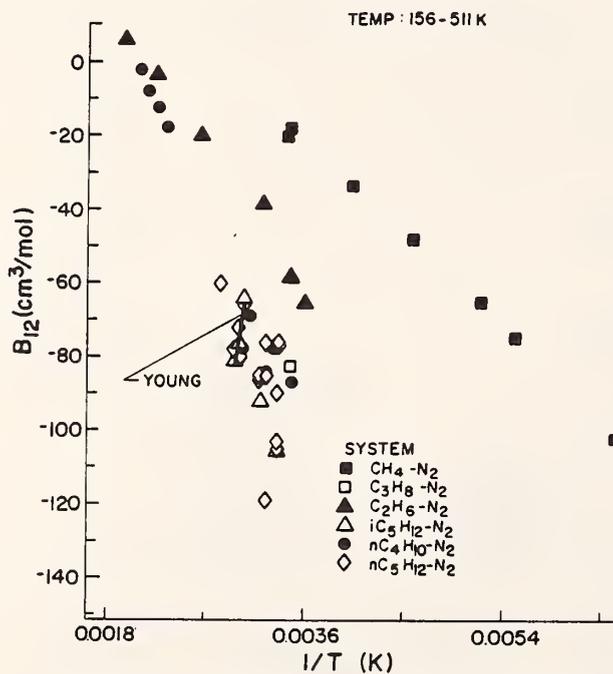


Figure 60. Temperature dependence of interaction second virial coefficient for nitrogen-paraffin hydrocarbon binary systems.

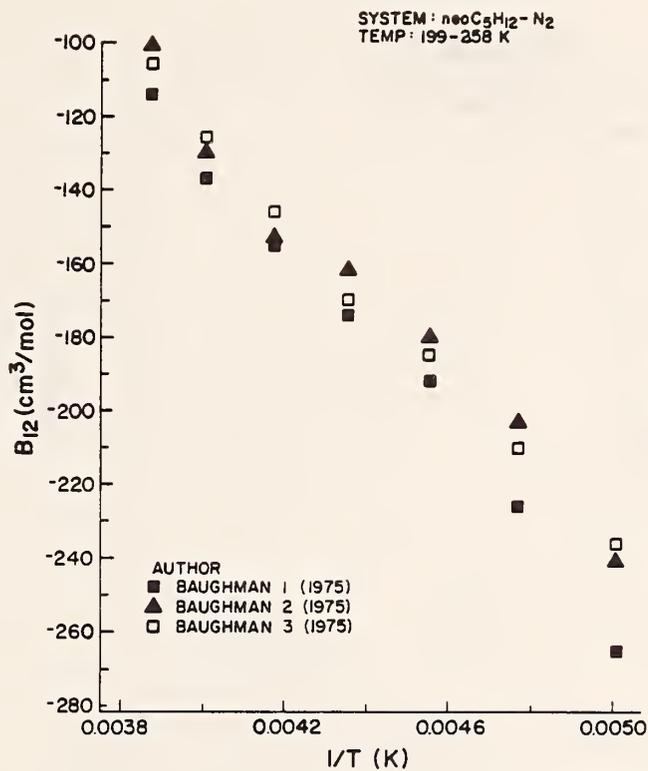


Figure 61. Temperature dependence of interaction second virial coefficient for the neo-C₅H₁₂-N₂ system.

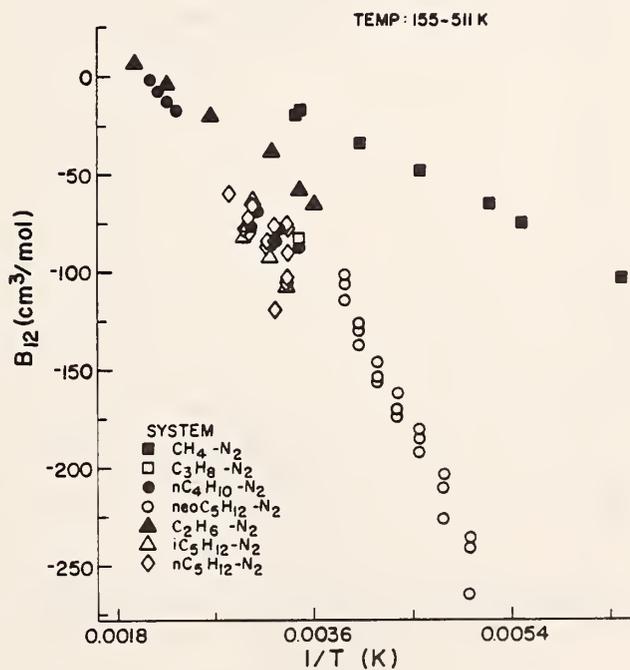


Figure 62. Temperature dependence of interaction second virial coefficient for nitrogen-paraffin hydrocarbon binary systems.

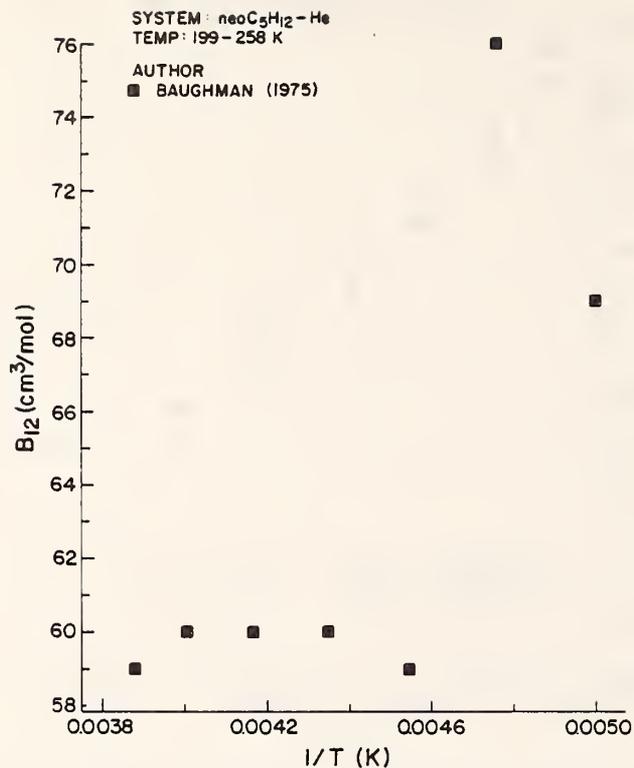


Figure 63. Temperature dependence of interaction second virial coefficient for the neo-C₅H₁₂-He system.

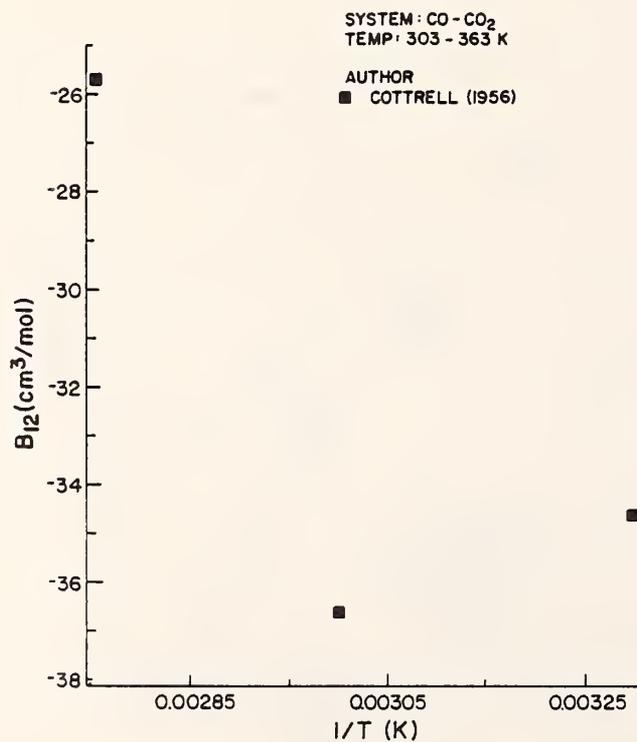


Figure 64. Temperature dependence of interaction second virial coefficient for the CO-CO₂ system.

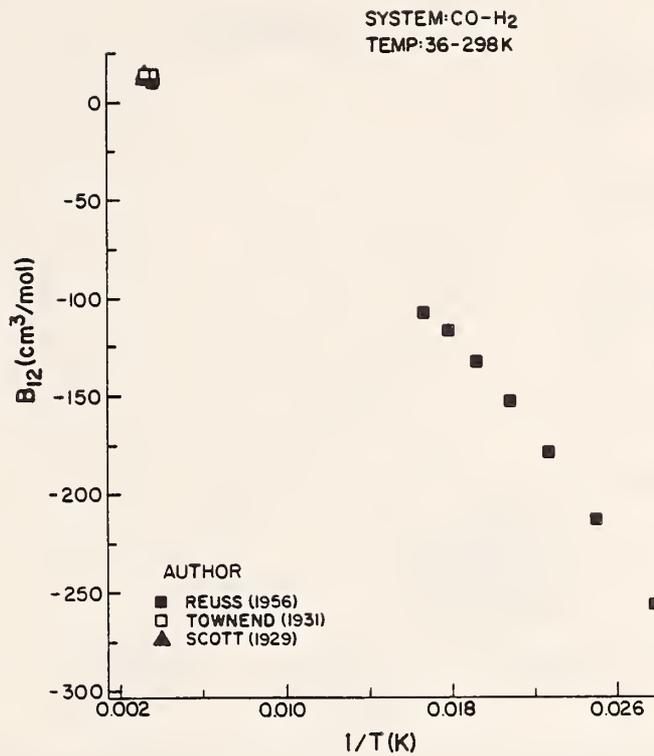


Figure 65. Temperature dependence of interaction second virial coefficient for the CO-H₂ system.

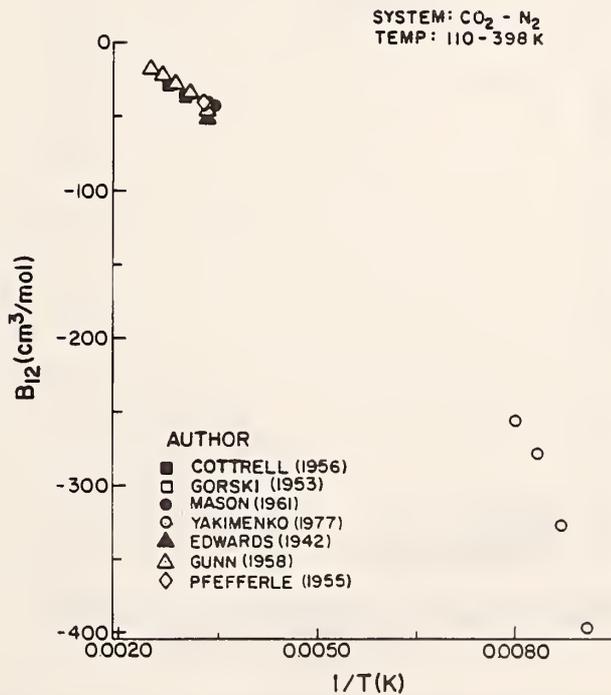


Figure 66. Temperature dependence of interaction second virial coefficient for the CO₂-N₂ system.

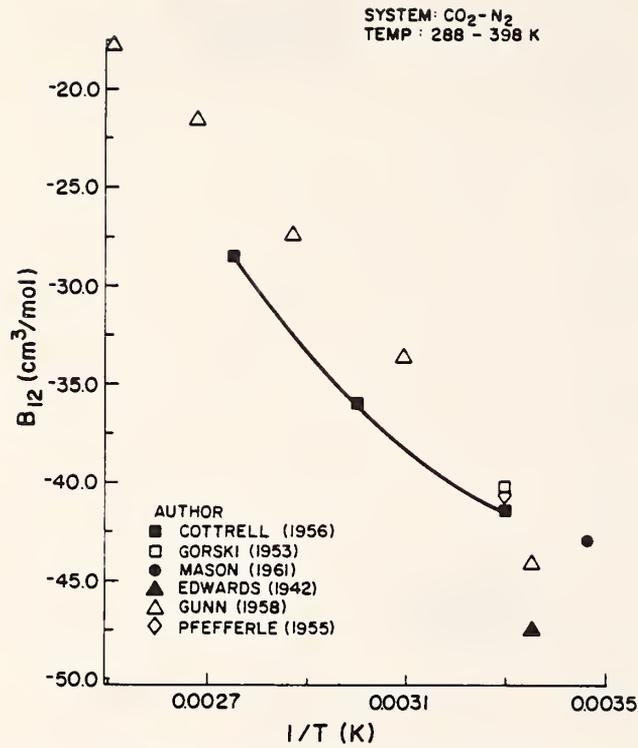


Figure 67. Temperature dependence of interaction second virial coefficient for the CO₂-N₂ system.

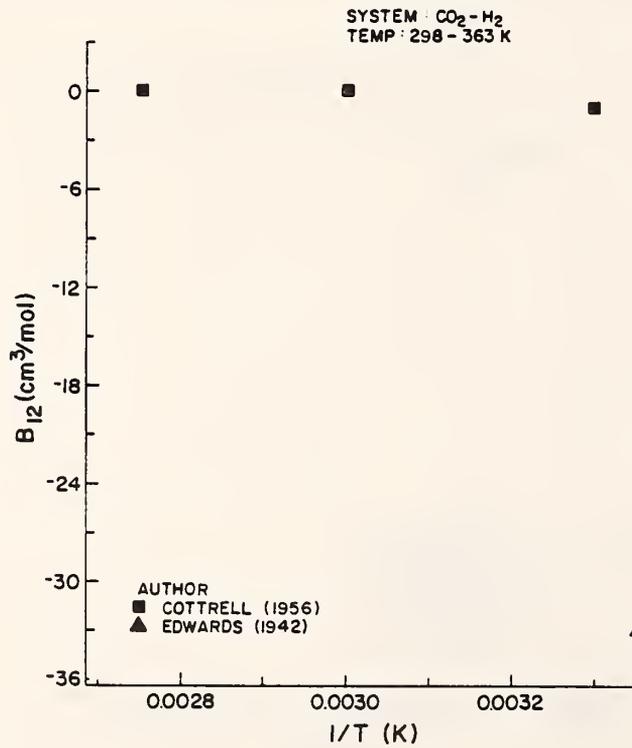


Figure 68. Temperature dependence of interaction second virial coefficient for the CO₂-H₂ system.

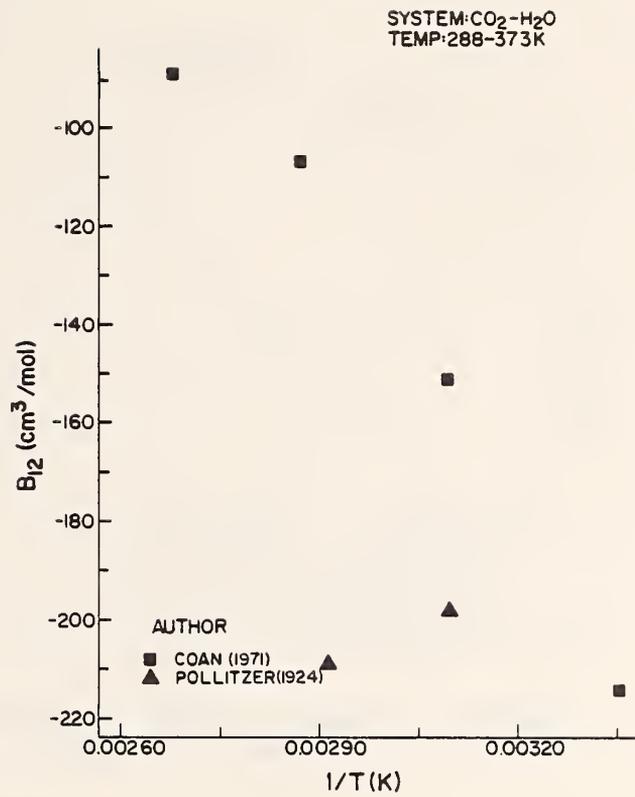


Figure 69. Temperature dependence of interaction second virial coefficient for the $\text{CO}_2\text{-H}_2\text{O}$ system.

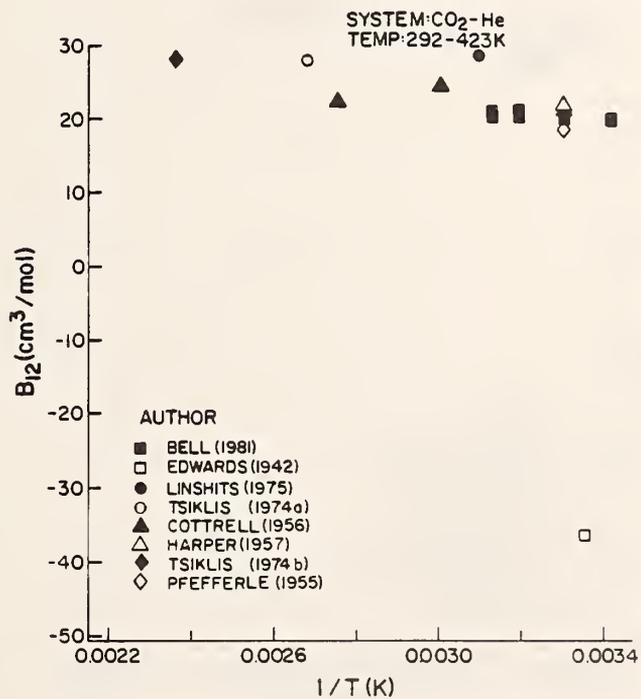


Figure 70. Temperature dependence of interaction second virial coefficient for the $\text{CO}_2\text{-He}$ system.

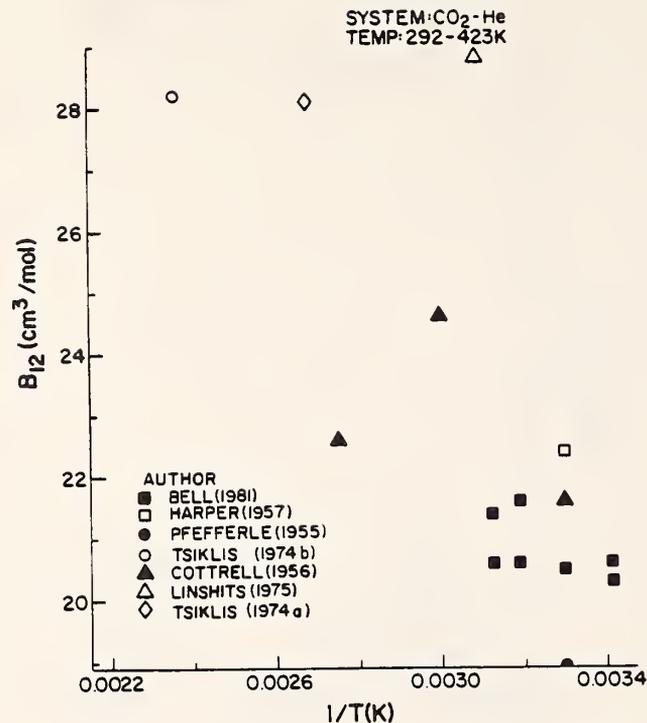


Figure 71. Temperature dependence of interaction second virial coefficient for the CO₂-He system (datum on Edwards and Roseveare deleted).

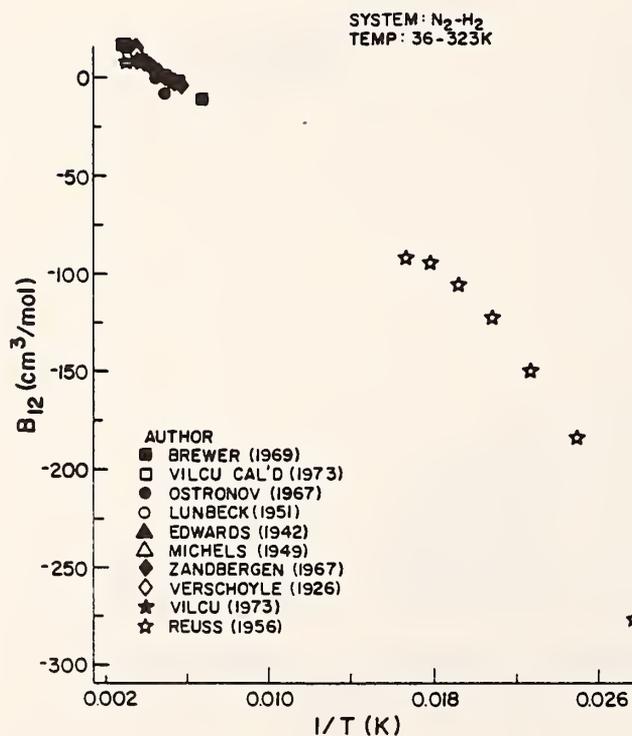


Figure 72. Temperature dependence of interaction second virial coefficient for the N₂-H₂ system (36-323K temperature range).

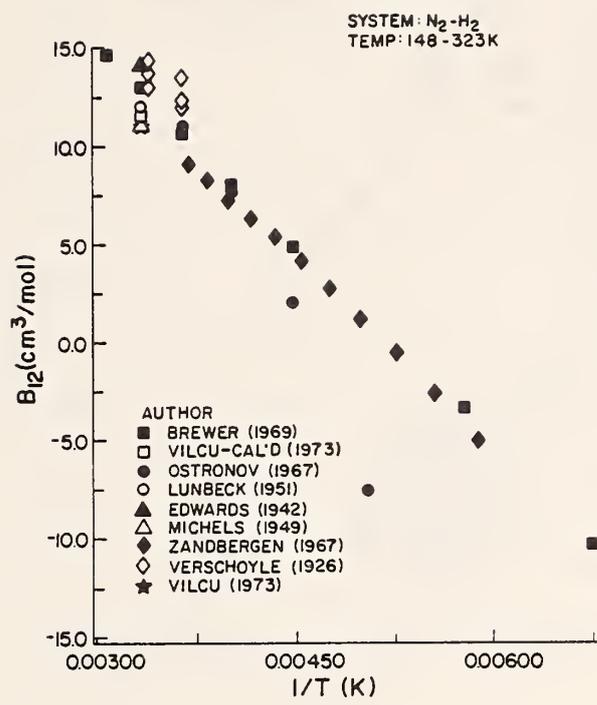


Figure 73. Temperature dependence of interaction second virial coefficient for the N₂-H₂ system (148-323K temperature range).

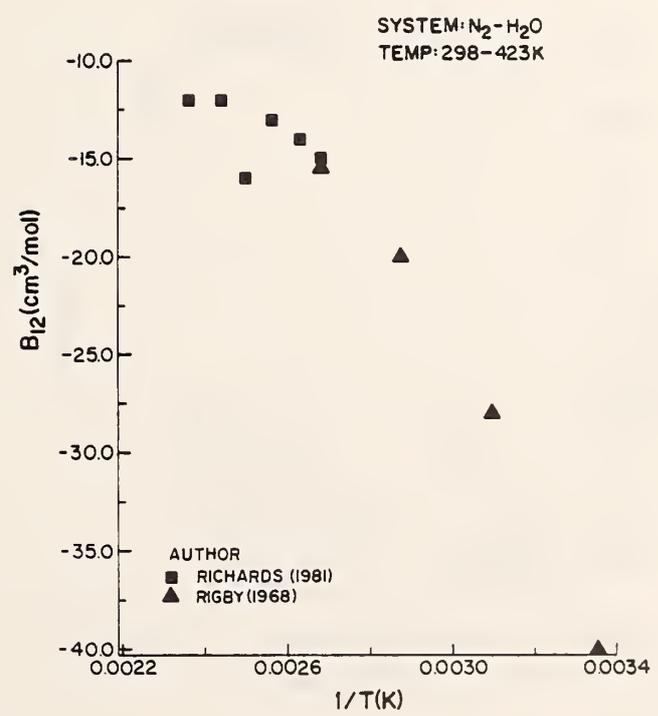


Figure 74. Temperature dependence of interaction second virial coefficient for the N₂-H₂O system.

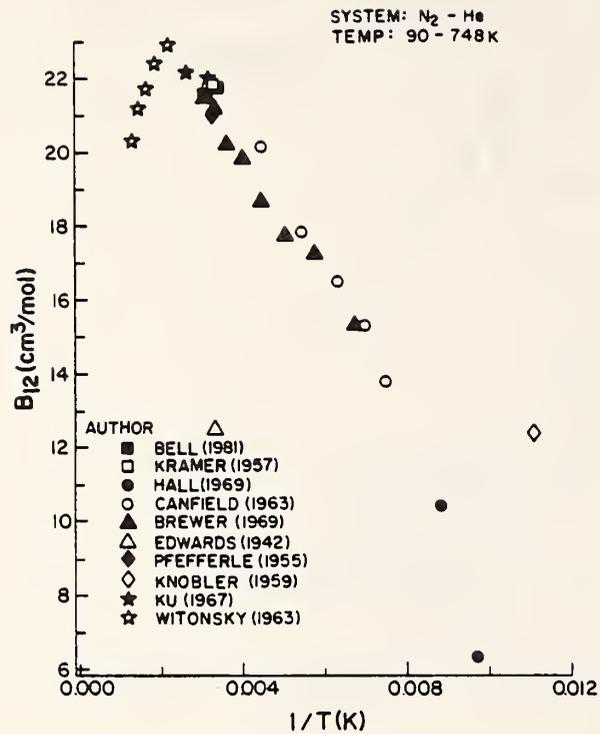


Figure 75. Temperature dependence of interaction second virial coefficient for the N₂-He system.

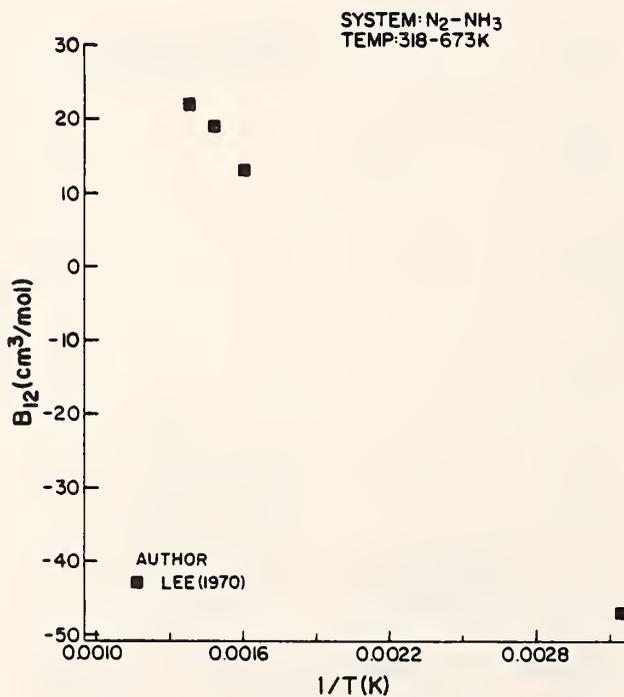


Figure 76. Temperature dependence of interaction second virial coefficient for the N₂-NH₃ system.

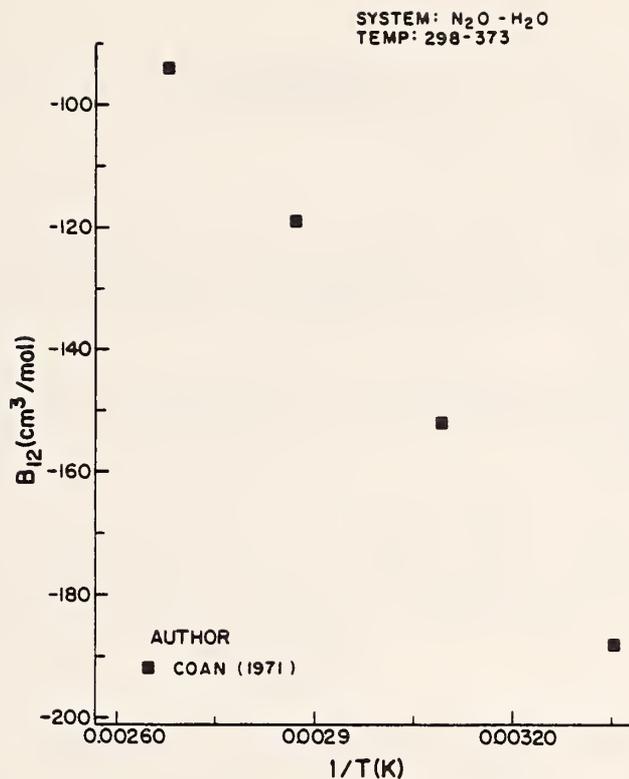


Figure 77. Temperature dependence of interaction second virial coefficient for the N₂O-H₂O system.

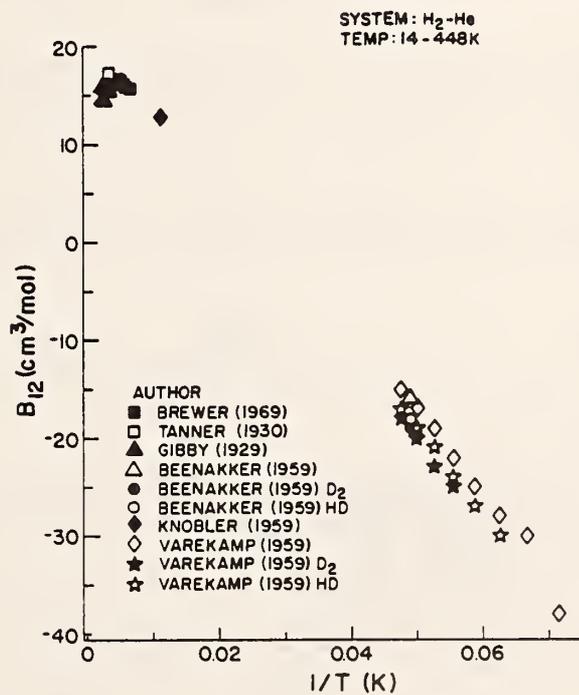


Figure 78. Temperature dependence of interaction second virial coefficient for the H₂-He system (14-448K temperature range).

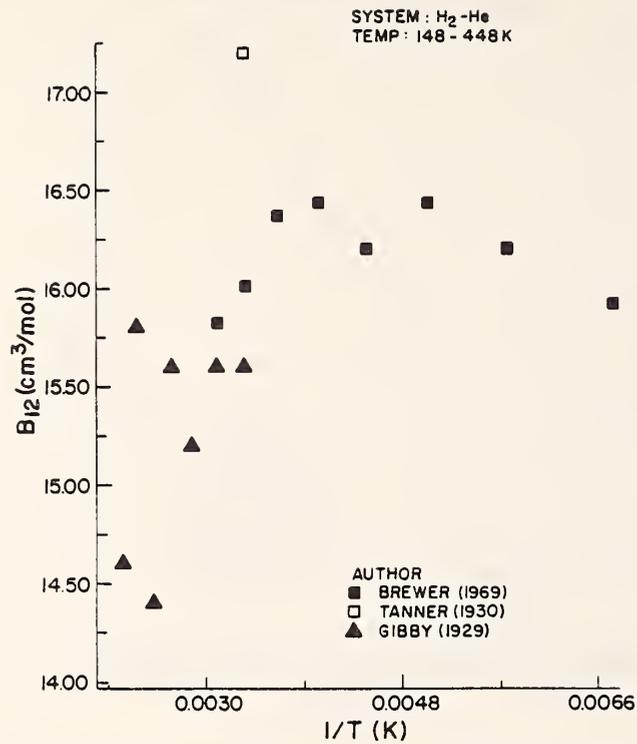


Figure 79. Temperature dependence of interaction second virial coefficient for the H₂-He system (148-448K temperature range).

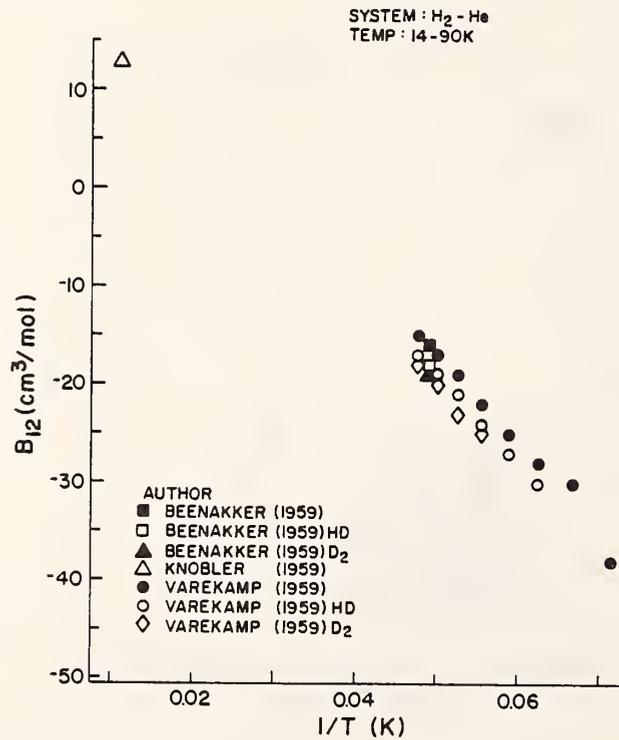


Figure 80. Temperature dependence of interaction second virial coefficient for the H₂-He system (14-90K temperature range).

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