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U.S. DEPARTMENT OF COMMERCE /National Bureau of Standards

Four Mathematical Models for the Prediction of LNG Densities

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Four Mathematical Models for the Prediction of LNG Densities

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FOUR MATHEMATICAL MODELS FOR THE PREDICTION OF LNG DENSITIES*

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Four mathematical models of the equation of state for LNG like mixtures are presented. The four models include an extended corresponding states model, a cell model, a hard sphere model and a revised Klosek and McKinley model. Each of the models has been optimized to the same experimental data set which included data for pure nitrogen, methane, ethane, propane, iso and normal butane, iso and normal pentane and mixtures thereof. For LNG like mixtures (mixtures of the orthobaric liquid state at temperatures of 120 K or less and containing at least 60% methane, less than 4% nitrogen, less than 4% each of iso and normal butane and less than 2% total of iso and normal pentane), all of the models are estimated to predict densities to within 0.1% of the true value. The revised Klosek and McKinley model is valid only for mixtures within the range of temperature and composition specified above while the other three models are valid for a broader range of pressure, temperature and composition. The experimental PVTx data set used in the optimization together with comparisons are given and listings of computer programs for each of the models are included.

Key words: Cell model; comparisons; computer programs; corresponding states; equation of state; hard sphere; LNG; mixtures; PVTx data; revised Klosek and McKinley.

1. INTRODUCTION

The purpose of this report is to present in final form the four mathematical models which were optimized to the experimentally determined orthobaric liquid

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PVTx data of Miller and Hiza [25], Haynes and Hiza [12], Haynes, et al. [11], Hiza, et al. [14], Hiza and Haynes [15] and Haynes [9]. Interim results of the project have been reported by Haynes, et al. [13] and by McCarty [23]. The models reported in these two interim publications differ only slightly from those presented here. A companion archival document, McCarty [24] with the same results but in much less detail has been submitted to the Journal of Chemical Thermodynamics. The intent of the documentation here is more in the vein of a user's handbook. The above experimental data are for the liquid phase of nitrogen, methane, ethane, propane, normal and isobutane and various mixtures thereof.

The goal of the project (the project included the above referenced experimental work) was to produce one or more computer models which would predict the density of LNG to within 0.1% of the true value from a knowledge of the temperature, pressure and composition of the LNG. At the beginning of this study LNG was defined as mixtures of the above components (N_2, CH_4, C_2H_6, C_3H_8, n and iC_4H_{10}) and only the saturated liquid between 95 - 150 K was to be considered. Near the end of the project n and iC_5H_{12} were added to the list of allowable components but no experimental PVTx of pure n and iC_5H_{12} or binary systems containing n and iC_5H_{12} were measured as part of the project. The inclusion of these two components is based on data from Orrit, et al. [29] and Orrit, et al. [30].

Four models were considered: the extended corresponding states model, a hard sphere model, a cell model and a Revised Klosek and McKinley model. With the exception of the Revised Klosek and McKinley model only pure fluid and binary system data were used to optimize the models. In the case of the Revised Klosek and McKinley model multicomponent PVTx data were used in the optimization process.

Over a normal range of LNG composition and temperature all four of the models predict densities which agree to within 0.1% of experiment. This is true of all of the experimental PVTx measurements on LNG like mixtures made as part of this project.

No equation or mathematical model based on experimental data can be more accurate than the original data and therefore the extent to which the original goal of 0.1% accuracy has been met depends entirely upon the accuracy of the experimental data referenced above.

There is no reason to doubt the experimental data and therefore there is every reason to believe that the goal of the project has been achieved.

2. EXTENDED CORRESPONDING STATES

The thermodynamic equations for the extended corresponding states method are developed in a paper by Rowlinson and Watson [35] and only a very brief description will be given here. Leach [20] developed transformation functions for hydrocarbons which are called shape factor functions. Mollerup [27] and Mollerup and Rowlinson [26] combined the earlier work with the equation of state for methane by Goodwin [8] to produce a computer program to calculate the density of LNG mixtures, which was further modified by Mollerup [28].

The computer program in Appendix F for the calculation of LNG densities based on the extended corresponding states method is an extensive revision of the Mollerup program. Earlier revisions were reported by McCarty [23] and Haynes, Hiza and McCarty [13].

The extended corresponding states method is defined by the following equations:

$$Z_i[P,T] = Z_o[P\, h_{ii,o}/f_{ii,o},\ T/f_{ii,o}] \qquad (1)$$

$$G_i[P,T] = f_{ii,o}\, G_o[P\, h_{ii,o}/f_{ii,o},\ T/f_{ii,o}] - RT\, \ell n(h_{ii,o}) \qquad (2)$$

where Z is the compressibility factor, G is the Gibbs free energy, P is pressure and T is temperature. The subscript o denotes the reference fluid and the subscript i denotes the fluid for which properties are to be obtained via the equation of state for the reference fluid and the transformation functions $f_{ii,o}$ and $h_{ii,o}$. The double subscript ii is introduced now to allow extension of the method to mixtures. The two defining eqs (1) and (2) are necessary since there are two transformation functions. In this case the equation of state for methane by McCarty [22] was chosen for the reference fluid. During the course of the study it was necessary to modify the equation of state by McCarty [22] to give a realistic vapor liquid phase boundary down to a temperature of 43 K. This modification was necessary to accommodate the very low reduced temperatures of the heavier hydrocarbons and was accomplished without changing the performance of the equation of state above the triple point of methane. The equation of state is given in Appendix B.

The $f_{ii,o}$ and $h_{ii,o}$ are defined as

$$f_{ii,o} = (T^c_{ii}/T^c_o)\ \theta_{ii,o}\ (T_r,V_r) \qquad (3)$$

and

$$h_{ii,o} = (V^c_{ii,o}/V^c_o)\ \phi_{ii,o}\ (T_r,V_r) \qquad (4)$$

where

$$\theta_{ii,o} = 1 + (w_i - w_o)[n_1 - n_2\, \ell n\, T_{r_i} + (n_3 - n_4/T_{r_i})(V_{r_i} - n_5)] \qquad (5)$$

and

$$\phi_{ii,o} = \frac{Z_o^c}{Z_i^c}\left[1 + (w_i - w_o)[n_6(V_{r_i} - n_7) - n_8(V_{r_i} - n_9)\ \ell n\ T_{r_i}]\right] \qquad (6)$$

The V_{r_i} and T_{r_i} are reduced temperature and volume, (i.e., $T_{r_i} = T/T_{ii}^c$ and $V_{r_i} = V/V_{ii}^c$) each fluid requires a unique w_i which was estimated using pure fluid experimental data. A single set of the n_i's are used for all fluids. The n_i's were estimated using all of the pure fluid experimental data from this study. The Z_o^c/Z_i^c is the ratio of the compressibility factors ($Z^c = P_cV_c/RT_c$) at the critical point. The parameters n_j, n_j, w_i and Z_{ii} are given in Appendix B. All of these parameters were estimated using the experimental PVT data set from this laboratory and least squares estimation techniques.

The extension of the above to mixtures is now accomplished by the following application of the following combining rules:

$$h_{x,o} = \sum_i \sum_j x_i x_j\ h_{ij,o} \qquad (7)$$

$$f_{x,o}\ h_{x,o} = \sum_i \sum_j x_i x_j\ f_{ij,o}\ h_{ij,o} \qquad (8)$$

$$f_{ij,o} = \xi_{ij}\ (f_{ii,o}\ f_{jj,o})^{1/2} \qquad (9)$$

$$h_{ij,o} = \eta_{ij}\ (\tfrac{1}{2}\ h_{ii,o}^{1/3} + \tfrac{1}{2}\ h_{jj,o}^{1/3})^3 \qquad (10)$$

The ξ_{ij} and the η_{ij} are binary interaction parameters determined by least squares from the PVTx data for binary mixtures. These parameters are given in Appendix B.

This method works quite well as may be seen in the comparisons in Appendix A. It has indeed reproduced all of the present experimental data set to within $\pm$ 0.1% except for 14 out of a total of 285 experimental data points. Of these 14 points, 11 are judged to have an uncertainty greater than 0.1%. Figure 1 presents the deviations between the calculated and experimental densities for these 14 points. Appendix A contains comparisons of calculated and experimental densities for the entire data set. This is the best performance of the four models presented here. No pressure, temperature or composition restrictions have been placed on this model.

In the interim publications by McCarty [23] and Haynes, Hiza and McCarty [13] some doubt about the accuracy of the calculated densities was expressed because of the disagreement with a few binary and multicomponent systems containing methane and butane. This disagreement has since been resolved by additional measurements (Haynes [9], Haynes [10] and Miller and Hiza [25] on some of the systems which agree with the predictions of the model but disagree with the previous measurements. The net result of the new measurements is a very slight change in binary interaction coefficients of the methane-butane and nitrogen-butane system. These changes have no practical effect on LNG like mixtures where the concentrations of N_2, iC_4H_{10} and nC_4H_{10} are individually less than 5%. In other words either the models presented here or those in the interim publications may be used to predict the density of a LNG like mixture to within 0.1% of the true density.

3. A HARD SPHERE METHOD

The model of Rodosevich and Miller [33] is one of many modifications of the Longuet-Higgins and Widom [21] model, and was chosen to be included in this study

as a representative example of the application of the hard sphere equation of state concept to the correlation of PVTx data. The equation of state by Rodosevich and Miller [33] is

$$\frac{PV}{RT} = c\,\frac{1 + y + y^2}{(1 - y)^3} - \frac{a}{RTV} \tag{11}$$

where the y = b/4V and a, b, and c are adjustable parameters, P is pressure, V is specific volume, T is temperature and R is the gas constant. The equation is applied to mixtures by assuming the one-fluid theory and applying the following combining rules.

$$a_m = \sum_i \sum_j a_{ij}\, x_i x_j \tag{12}$$

$$b_m = \sum_i \sum_j b_{ij}\, x_i x_j \tag{13}$$

$$c_m = \sum_i \sum_j c_{ij}\, x_i x_j \tag{14}$$

The mixing rules are:

$$b_{ij} = \left[\frac{b_{ii}^{1/3} + b_{jj}^{1/3}}{2}\,(1 - j_{ij})\right]^3 \tag{15}$$

$$a_{ij} = (a_{ii}a_{jj})^{1/2}\left[\frac{b_{ij}^2}{b_{ii}b_{jj}}\right]^{1/2}(1 - k_{ij}) \tag{16}$$

$$c_{ij} = \frac{c_{ii} + c_{jj}}{2} \tag{17}$$

The parameters j_{ij} and k_{ij} are in this case the binary interaction parameters. The a's, b's, c's, j_{ij}'s and k_{ij}'s are given in Appendix B. The excess volume is now calculated using the equation of state and

$$V_E = \tilde{V}_m - \tilde{V}_i x_i \qquad (18)$$

where $\tilde{V}_m$ and the $\tilde{V}_i$ are calculated via the eqs 11 through 17 and then

$$V_m = V_i x_i + V_E \qquad (19)$$

where the V_E is from eq (18) and the V_i are from experimental data. The values of V_i in this case were calculated from the equations for the liquid density of the pure fluids given in Appendix C.

The above equations are those of Rodosevich and Miller [33] and Rodosevich [34] and only the j_{ij}'s and k_{ij}'s have been revised on the basis of the present new data set, and only binary systems data were used to estimate via least squares the j_{ij}'s and k_{ij}'s.

As the method is used here it is an excess volume method, and consequently when the temperature of the mixture approaches the critical temperature of one of the component fluids, the method fails. Since the critical temperature of nitrogen is about 126 K, this method should not be used for mixtures containing nitrogen at temperatures above 120 K. Eliminating the data points for mixtures which contain nitrogen at temperatures above 120 K reduces the set from 285 to 251 PVTx points. Figure 2 is a percentage deviation plot containing all of the data points from the set of 251 for which densities calculated by the hard sphere method differ from the experimental density by more than 0.1%. Two things are readily seen in comparing figs. 1 and 2; first, even though total number of points has been reduced in the comparison set, the number of points for which deviations exceed 0.1% in the hard sphere comparison, fig. 2, is far more than for the extended corresponding states comparison, fig. 1. Second, the hard sphere method becomes more uncertain for all mixtures, regardless of components as the temperature exceeds 115 K.

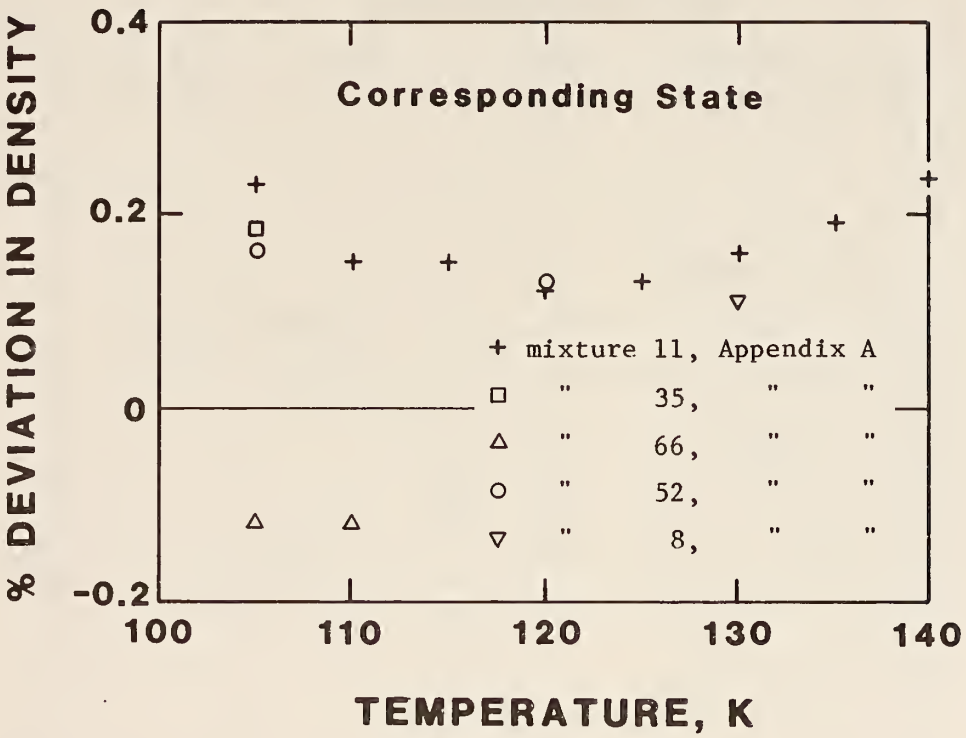

Figure 1. All deviations greater than 0.1% between experimental and calculated densities using the Extended Corresponding States method. The comparison set is all of the data points in Appendix A. (285 data points)

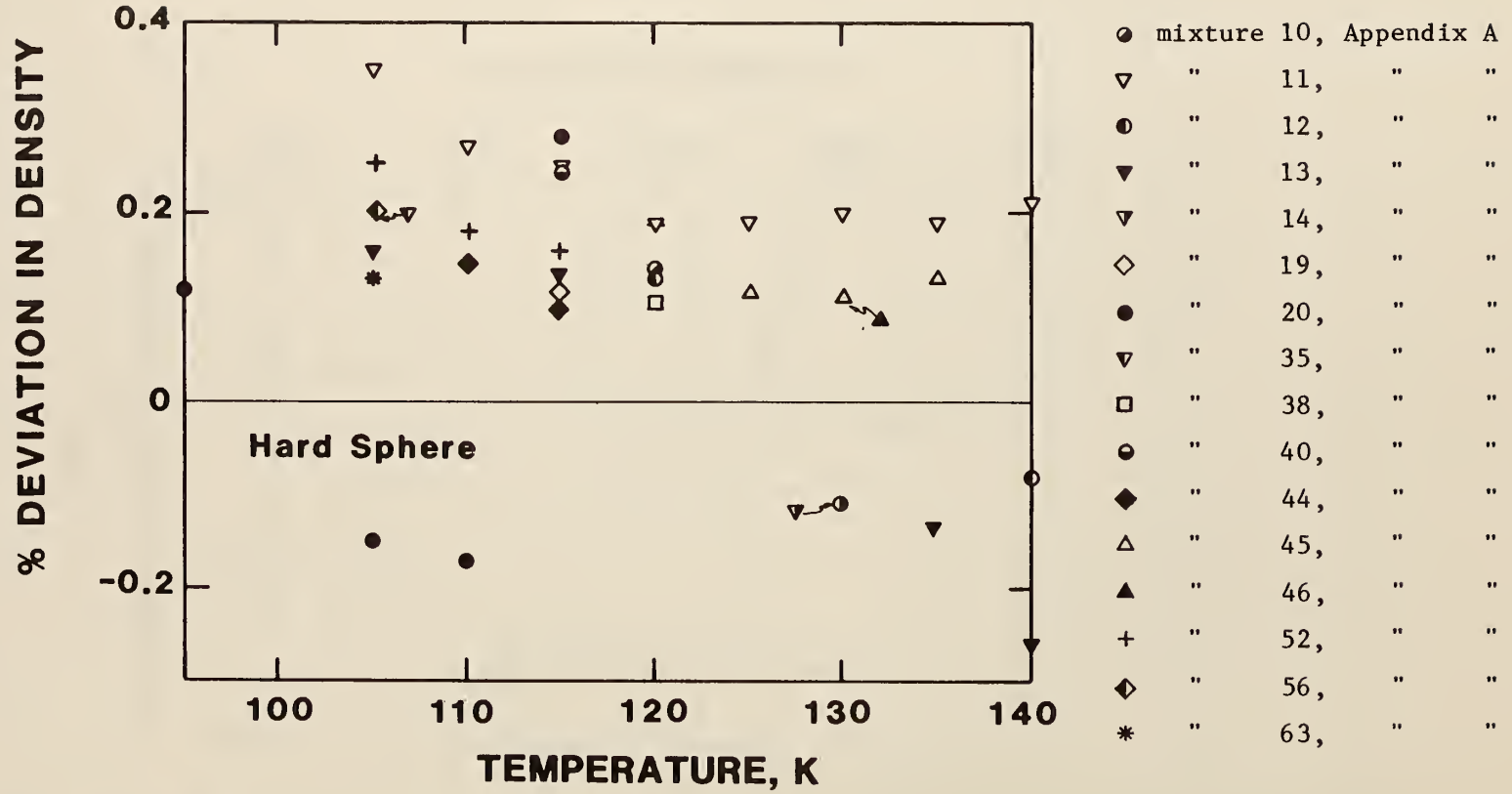

Figure 2. Deviations greater than 0.1% between experimental and calculated densities using the Hard Sphere model. The comparison set is all of the data in Appendix A except those data points for mixtures containing N_2 at temperatures above and including 120 K. (251 data points)

4. A REVISED KLOSEK AND MCKINLEY METHOD

The Klosek and McKinley method [18] is a totally empirical recipe for calculating the density of a LNG-like mixture given the temperature and composition. Pressure is not taken into account. However, this does not seem to be a serious omission. The procedure proposed by Klosek and McKinley [18] is as follows:

$$V_{mix} = \sum X_i V_i - kX_{CH_4} \tag{20}$$

where V_{mix} is the volume of the mixture, X_i and V_i are the mole fraction and volume of the i^{th} component, X_{CH_4} is the mole fraction of methane and k is a correction factor obtained from a table or graph. The V_i and k are obviously temperature dependent and in addition k is dependent upon the molecular weight of the mixture.

Using the present data set k was calculated for all of the experimental data points where methane was present in the mixture and excluding all data points where N_2 was present in greater than 5% concentration. Figure 3 shows a typical isotherm for k, with N_2 present (labeled k_2) and without N_2 present (labeled k_1). All of the isotherms available show similar behavior, i.e., all of the k's for mixtures containing nitrogen (of about 5%) fall on one line and all of those for mixtures without nitrogen fall on another. Since all of the mixtures with nitrogen have about the same amount of nitrogen present (about 4.5%), the method was modified by adding a term to take into account the nitrogen when it is present. The equation becomes

$$V_{mix} = \sum X_i V_i - [k_1 + (k_2 - k_1) X_{N_2}/.0425] X_{CH_4} \tag{21}$$

where everything is the same as in eq (20) except that k_1 is read from one

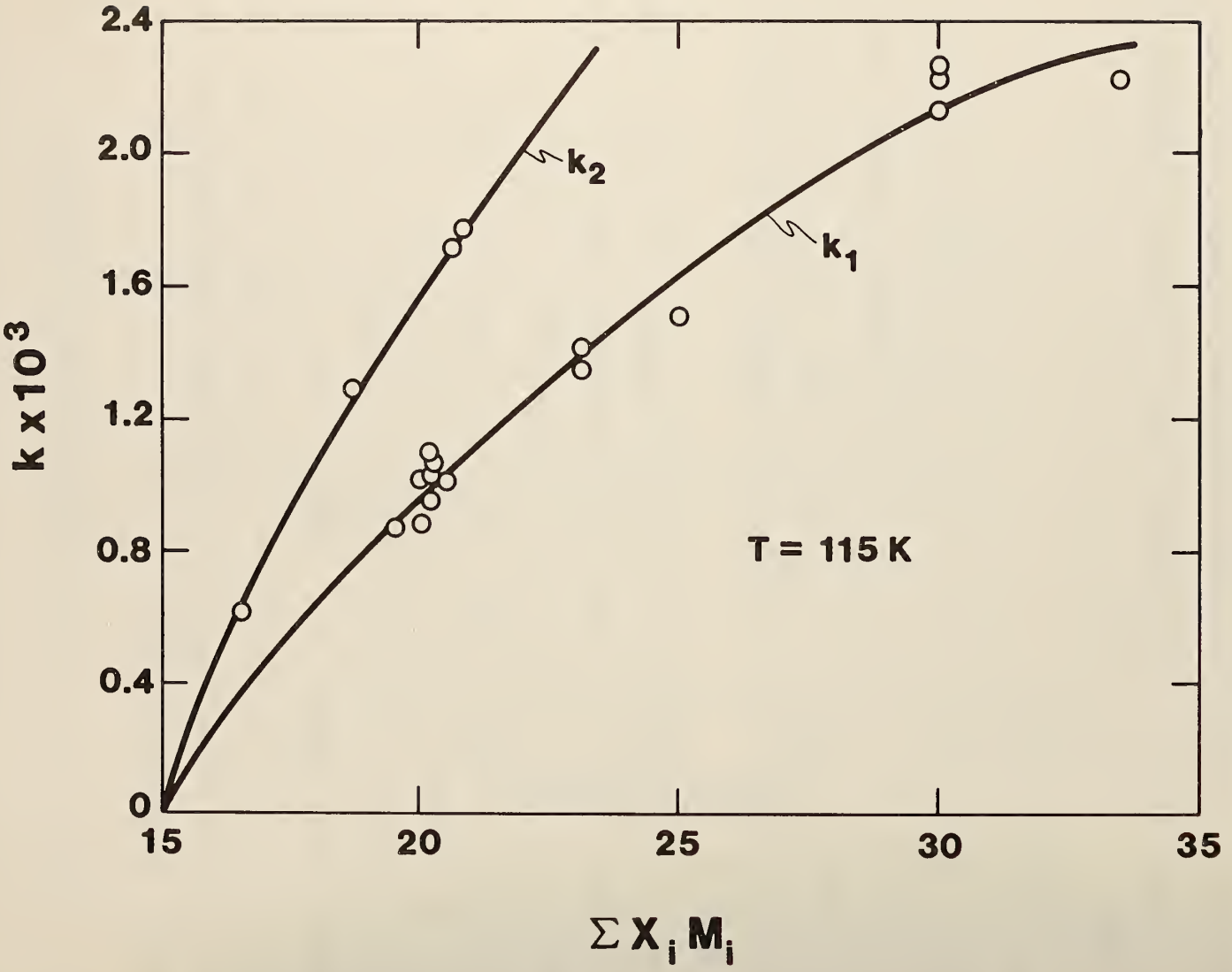

Figure 3. A plot of the correction factors (k_1 and k_2) for the 115 kelvin isotherms.

curve and k_2 is read from the other. Appendix D gives tables of values for the V_i, k_1 and k_2 which are spaced such that linear interpolation is adequate in both variables (i.e., temperature or molecular weight). The k factors in Appendix D have been obtained graphically from the multicomponent PVTx data of Hiza and Haynes [15] and Miller and Hiza [25] as well as densities calculated from the extended corresponding states method of section 2.

The limits of compositions of the revised Klosek and McKinley method are the most severe of any of the methods given here. This method should not be used for mixtures other than LNG like mixtures and for LNG like mixtures only when they contain at least 60% methane, less than 4% nitrogen, less than 4% each of iC_4H_{10} nC_4H_{10} and less than 2% total of iC_5H_{12} and nC_5H_{12}.

There are 40 experimental PVTx points from the original set of 285 which may be considered LNG like and fall within the composition limits outlined above. Figure 4 shows all of the deviations between calculated and experimental densities in this 40 point comparison set which exceeds the 0.1% criterion. The deviation trends for the revised Klosek and McKinley method (fig. 4) are very similar to those of the hard sphere method (fig. 2) and in fact all of the deviations in fig. 4 occur at temperatures at or above 115 K, therefore the method can only be considered as accurate as the others for LNG like mixtures at temperatures below 115 K.

5. THE CELL MODEL

The cell model considered here was originally proposed by Renon, et al. [32]. In a paper by the same three authors which appeared simultaneously (Eckert, et al. [7]), the cell model was applied to mixtures via Scott's [36] two-fluid theory and a three parameter corresponding states theory. Albright [2] further modified the method by modifying the mixing rules on the basis of a

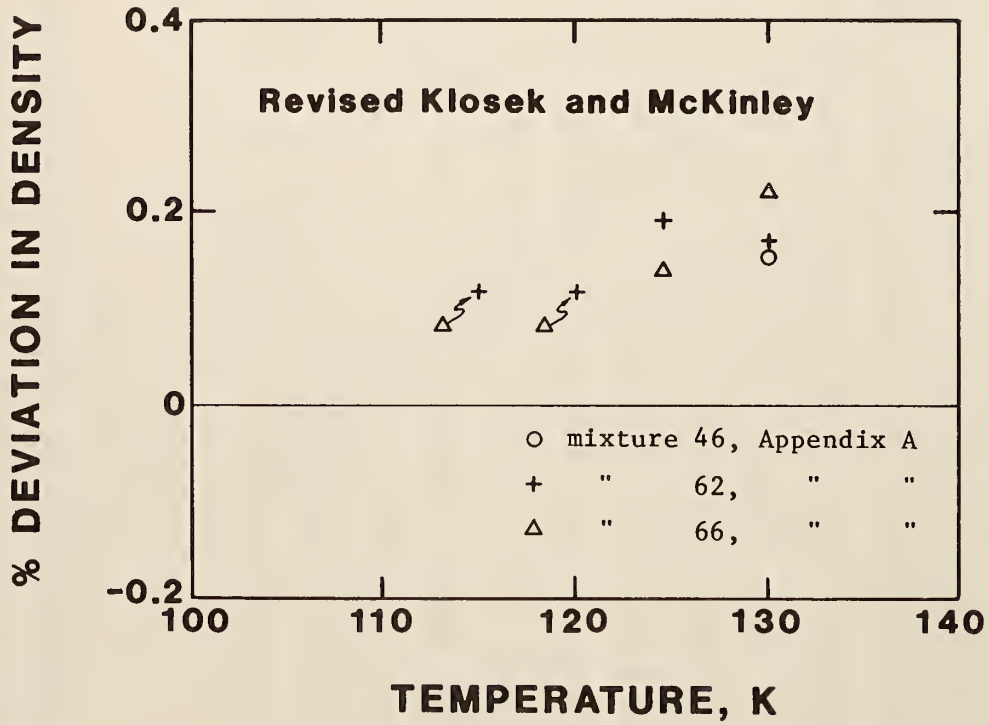

Figure 4. All deviations greater than 0.1% between experimental and calculated densities using the Revised Klosek and McKinley model. The comparison set is all multicomponent mixture data in Appendix A with > 60% CH_4, < 4% nC_4H_{10}, < 4% iC_4H_{10}, < 4% N_2 and < 2% of n + iC_5H_{12}. (40 data points)

proposal by Yuan [38] and by inserting a pressure dependence based on the experimental liquid ethane data by Pope [31].

The optimization of this method was carried out by M. Albright [1] at Phillips Petroleum Company in Bartlesville, Oklahoma and the details of this work will be published elsewhere. The model is included here because it was optimized to the same data set as the others and therefore the comparisons between experimental and calculated densities given here in fig. 5 together with figs. 1, 2 and 4 provide a common basis of comparison with the other three methods. A listing of the computer program is given in Appendix F.

The same data set as was used in the hard sphere method for comparison has been used here, i.e., all of the data points for mixtures containing nitrogen at temperatures 120 K and above have been taken out of the original 285 points leaving a total of 251 data points.

As in the case of the other methods fig. 5 shows all of the points for which the calculated and experimental densities differ by more than 0.1%.

6. USE OF THE METHODS

When the project started in 1972, the atomic weights of nitrogen, carbon and hydrogen were taken from the 1961 carbon 12 scale, IUPAC [16]. During the course of the investigation a revision, Atomic Weights of the Elements [3], to this scale appeared. The revision changed slightly the atomic weights of carbon and hydrogen, but since the changes were small (the maximum difference in any of the densities used here is 0.003%), and because changing the atomic weights would not change the relative results, the changes were not made. Therefore when using the tables and programs in the appendices, the molecular weights given in the tables and programs should be used to maintain consistency.

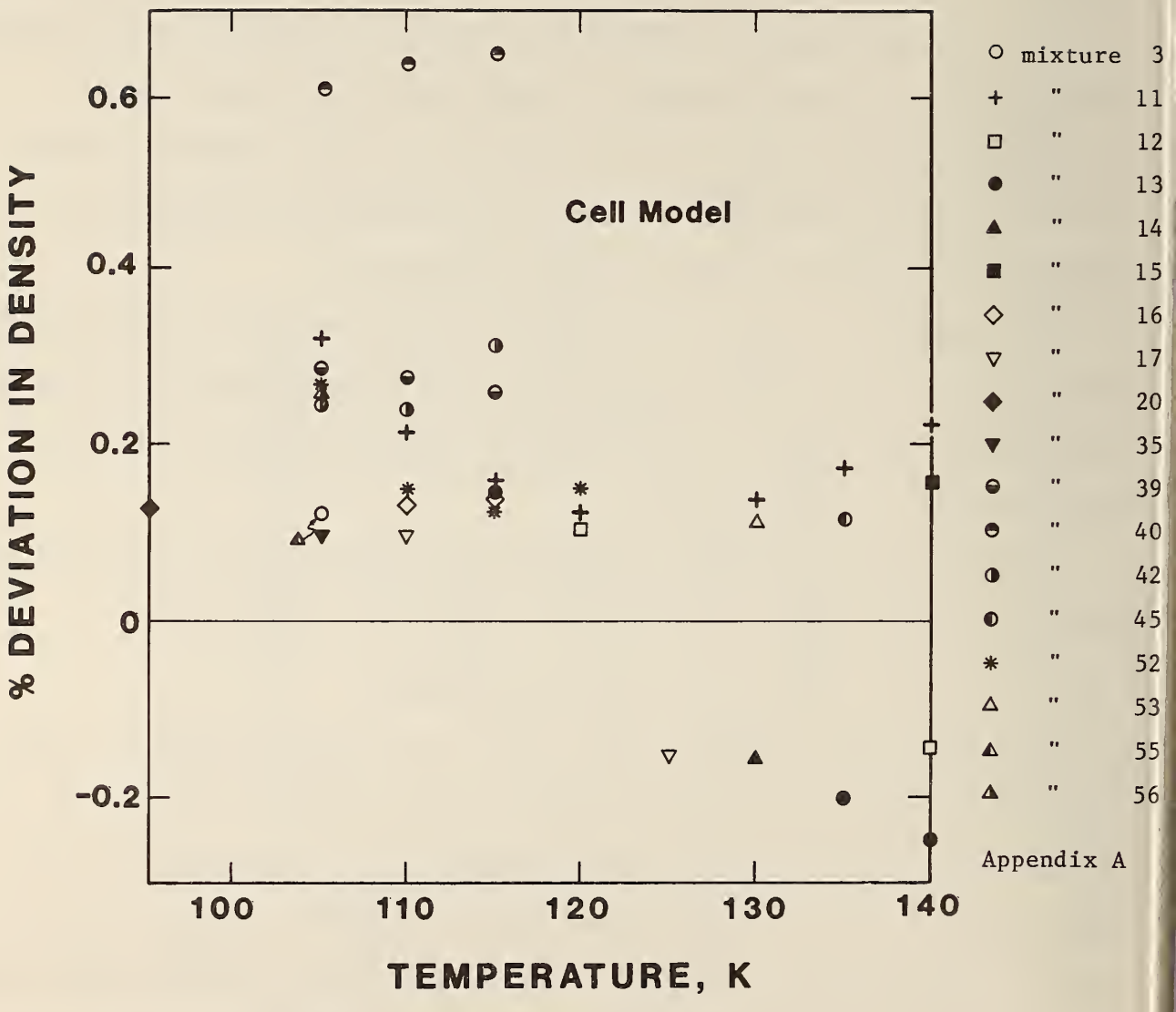

Figure 5. Deviations greater than 0.1% between experimental and calculated densities using the Cell model. The comparison set is all of the data in Appendix A except those data points for mixtures containing N_2 at temperatures above and including 120 K. (251 data points)

The critical parameters used here are from: CH_4, McCarty [22]; C_2H_6, Sliwinski [37]; C_3H_8, Das, et al. [4]; iC_4H_{10}, Das, et al. [5]; nC_4H_{10}, Das, et al. [6]; iC_5H_{12}, Kudchadker, et al. [19]; and N_2, Jacobsen, et al. [17].

Errors in the input variables will of course, cause errors in the density predicted by the models. In general, the error in density caused by an error in the input varibles is a function of those input variables, and must be treated on an individual basis. However, for LNG like mixtures certain general trends are found. An error in the pressure must be at least 50% before it will have any effect at all on the resulting density. An error in composition, unless it is of the order of several percent, will cause the same relative error in density as it will cause in the molecular weight of the mixture, i.e., if an error in composition causes a 0.1% error in the resulting molecular weight, it will also cause a 0.1% error in the predicted density.

The error in the calculated density due to an error in the input temperature is a function of the composition and the temperature. Table 1 gives resulting errors in density for a 1% error in temperature, for three hypothetical LNG like mixtures.

In general the errors in density caused by an error in temperature are the largest for mixtures containing a high concentration of the most volatile fluids, CH_4 and N_2, and correspondingly the errors decrease as the concentration of the heavier hydrocarbons increases in the mixture. These errors are not a function of which model is being used.

When using the extended corresponding states method, one should keep in mind that twelve significant figures are required by the methane equation of state. The hard sphere model also uses the methane equation from McCarty [22] and the nitrogen equation of Jacobsen, et al. [17] to calculate compressibilities and

TABLE 1. Errors in Density Caused by an Error in the Input Temperature of 1%.

Temperature	% Error in Density		
K	Mix A*	Mix B*	Mix C*
95	0.28	0.25	0.20
100	0.30	0.27	0.22
105	0.32	0.29	0.24
110	0.35	0.32	0.29
115	0.39	0.34	0.31

Mix A* = 0.95 CH_4, 0.05 N_2

Mix B* = 0.9 CH_4, 0.02 C_2H_6, 0.02 C_3H_8, 0.02 iC_4H_{10}, 0.02 nC_4H_{10}, 0.02 N_2

Mix C* = 0.6 CH_4, 0.3 C_2H_6, 0.02 C_3H_8, 0.02 iC_4H_{10}, 0.02 nC_4H_{10}, 0.02 N_2, 0.02 iC_5H_{12}.

*Arbitrary LNG like compositions assumed for the purpose of illustrating the effect of an error in the input temperature.

therefore requires twelve significant figures to insure the accuracy of the calculated density. The other two models require only eight significant figures to be carried along in the calculations.

7. CONCLUSIONS

On the basis of the performance of the four models given here and subject to the composition and temperature restrictions already noted, it is estimated that given the pressure, temperature and composition of LNG, any one of the four models may be used to predict the density to within 0.1% of the true value. As has already been mentioned (see section 1) the above accuracy statement is dependent entirely upon the accuracy of the experimental data in Haynes, et al. [11], Haynes, et al. [13], Hiza, et al. [14], Haynes [9], Hiza and Haynes [15], Miller and Hiza [25] and Haynes [10]. These data have been estimated by the authors to be accurate to within 0.1% of the true value with a precision of a few hundredths of a percent. The work on the models given here have provided no basis for questioning the claims of the experimenters, in fact the ability of the models to predict the densities of the multicomponent mixtures to within 0.1% of the measured values tends to support the accuracy claims of the experimenters.

Interim results of this study were reported by Haynes, et al. [13] and McCarty [23], both of which contain earlier versions of the mathematical models given here. These earlier versions are only slightly different than the final ones and for the purposes of calculating LNG densities either of the versions may be used. The reader is, however, cautioned to read the limitations of each model as defined in the earlier sections.

Computer programs for the four models are available at the Thermophysical Properties Division of the National Bureau of Standards in Boulder, Colorado.

8. REFERENCES

[1] Albright, M. A., Private communication (1976).

[2] Albright, M. A., A Model for the Precise Calculation of Liquefied Natural Gas Densities, NGPA Technical Publication TP-3 (1973).

[3] Atomic Weights of the Elements 1973, Pure and Applied Chemistry, Vol 37, 589-603 (1974).

[4] Das, T. R., Eubank, P. T., Advances in Cryogenic Engineering Vol 18, Plenum Press, New York (1973), p. 208.

[5] Das, T. R., Eubank, P. T., Advances in Cryogenic Engineering Vol 18, Plenum Press, New York (1973), p. 253.

[6] Das, T. R., Eubank, P. T., Advances in Cryogenic Engineering Vol 18, Plenum Press, New York (1973), p. 244.

[7] Eckert, C. A., Renon, H. and Prausnitz, J. M., Molecular Thermodynamics of Simple Liquids, IEC Fund 6, 58 (1967).

[8] Goodwin, R. D., The Thermophysical Properties of Methane, from 90 to 500 K at Pressures up to 700 Bars, Nat. Bur. Stand. (U.S.), Tech. Note 653 (1974).

[9] Haynes, W. M., Orthobaric Liquid Densities of Methane + Isobutane and Methane + Normal Butane Mixtures at Low Temperatures (to be published).

[10] Haynes, W. M., Measurements of Orthobaric Liquid Densities of Multicomponent Mixtures of LNG Components (N_2, CH_4, C_2H_6, C_3H_8, iC_4H_{10}, nC_4H_{10}, iC_5H_{12}, nC_5H_{12}) Between 110 and 130 K, to be published.

[11] Haynes, W. M., Hiza, M. J. and Frederick, N. V., A Magnetic Suspension Densimeter for Measurements on Fluids of Cryogenic Interest, Review of Scientific Instruments 47, No. 10, 1237-50 (Oct 1976).

[12] Haynes, W. M. and Hiza, M. J., Measurements of the Orthobaric Liquid Densities of Methane, Ethane, Propane, Isobutane, and Normal Butane, J. Chem. Thermodyn., 9, 179 (1977).

[13] Haynes, W. M., Hiza, M. J. and McCarty, R. D., Densities of LNG for Custody Transfer, (Proc. Fifth International Conference of LNG, Dusseldorf, Germany), J. W. White, W. McGrew, S. Farmer, D. Hansen, H. Jacobshagen, editors; Institute of Gas Technology, Chicago, Ill., Vol. 2, Paper II, Section III (1977).

[14] Hiza, M. J., Haynes, W. M. and Parrish, W. R., Orthobaric Liquid Densities and Excess Volumes for Binary Mixtures of Low Molecular Weight Alkanes and Nitrogen Between 105 and 140 K, J. Chem. Thermodyn. 9, 873 (1977).

[15] Hiza, M. J. and Haynes, W. M., Orthobaric Liquid Densities and Excess Volumes for Multicomponent Mixtures of Low Molar Mass Alkanes and Nitrogen Between 105 and 125 K, J. Chem. Thermodyn. 12, 1 (1980).

[16] International Union of Pure and Applied Chemistry, 1961 Carbon 12 Scale.

[17] Jacobsen, R. T., Stewart, R. B., McCarty, R. D. and Hanley, H. J. M., Thermophysical Properties of Nitrogen from the Fusion Line to 3500 R (1944 K) for Pressures to 150,000 psia (10342 x 10^5 N/m^2), Nat. Bur. Stand. (U.S.), Technical Note No. 648 (Dec 1973).

[18] Klosek, J. and McKinley, C., Densities of Liquefied Natural Gas and of Low Molecular Weight Hydrocarbons, Proc. First Int. Conf. on LNG, IGT, Chicago (1968).

[19] Kudchadker, A. P., Alani, G. H. and Zwolinski, B. J., The Critical Constants of Organic Substances, Chem. Rev. 68, 659-735 (1968).

[20] Leach, J. W., Molecular Structure Correction for Applying the Theory of Corresponding States to Non-Spherical Pure Fluids and Mixtures, Ph.D. Thesis, Rice University (1967).

[21] Longuet-Higgins, H. C. and Widom, B., A Rigid Sphere Model for Melting of Argon, Mol. Phys. 8, 549 (1964).

[22] McCarty, R. D., A Modified Benedict-Webb-Rubin Equation of State for Methane Using Recent Experimental Data, Cryogenics 14, No. 5, 276-80 (May 1974).

[23] McCarty, R. D., A Comparison of Mathematical Models for the Prediction of LNG Densities, Nat. Bur. Stand. (U.S.), Interagency Report NBSIR 77-867 (Oct 1977).

[24] McCarty, R. D., Optimization and Comparisons of Mathematical Models for the Prediction of Liquefied Natural Gas Densities, J. Chem. Thermodyn. (to be published, 1980).

[25] Miller, R. C. and Hiza, M. J., Experimental Molar Volumes for Some LNG Related Saturated Liquid Mixtures, Fluid Phase Equilibria 2, 49-57 (1978).

[26] Mollerup, J. and Rowlinson, J. S., The Prediction of Densities of Liquefied Natural Gas and of Lower Molecular Weight Hydrocarbons, Chem. Engng. Sci. 29 (1973).

[27] Mollerup, J., Correlated and Predicted Thermodynamic Properties of LNG and Related Mixtures in the Normal and Critical Region, Adv. Cryog. Eng. 20, 172 (K. D. Timmerhaus, editor, Plenum Press, New York, 1975).

[28] Mollerup, J., Thermodynamic Properties of Natural Gas, Petroleum Gas, and Related Mixtures Enthalpy Predictions, Adv. Cryog. Eng. 23, 550 (K. D. Timmerhaus, editor, Plenum Press, New York, 1978).

[29] Orrit, J. and Olives, J. F., Density of Liquefied Natural Gas and Its Components, Distributed at the 4th International Conference on Liquefied Natural Gas, Algeria (1974).

[30] Orrit, J. E. and Laupretre, T. M., Density of Liquefied Natural Gas Components, Adv. Cryog. Eng. 23, 573 (K. D. Timmerhaus, editor, Plenum Press, New York, 1978).

[31] Pope, G. A., Calculation of Argon, Methane and Ethane Virial Coefficients at Low Reduced Temperature, Ph.D. Thesis, Rice University (1971).

[32] Renon, H., Eckert, C. A. and Prausnitz, J. M., Molecular Thermodynamics of Simple Liquids, IEC Fund. 6, 52 (1967).

[33] Rodosevich, J. B. and Miller, R. C., Calculation of LNG Excess Volumes by a Modified Hard-Sphere Model, Adv. Cryog. Eng. 19, 339 (1974).

[34] Rodosevich, J. B., Measurements and Prediction of Liquefied Natural Gas Densities, M.Sc. Thesis, Univ. Wyoming (1973).

[35] Rowlinson, J. S. and Watson, I. D., The Prediction of the Thermodynamic Properties of Fluids and Fluid Mixtures - I. The Principle of Corresponding States and Its Extensions, Chem. Engng. Sci. 24, 1565 (1969).

[36] Scott, R. L., J. Chem. Phys. 25, 193 (1956).

[37] Sliwinski, P. Z., Die Lorentz-Lorenz Funktion Von Dampfformigem und Fluessiyem Athan, Propan and Butan, Phys. Chem. (Frankfurt) 63, 263 (1969).

[38] Yuan, I. C., Correlation and Prediction of Excess Properties for Selected Binaries in the C_6-C_7 Hydrocarbon Boiling Range of Conformal Solution Theory, D.Sc. Dissertation, Washington University (1971).

Appendix A. Experimental Data

The following is a list of all of the experimental PVTx mixtures data which were measured during the course of this project. The data are from Miller and Hiza [25]; Haynes and Hiza [12]; Haynes, et al. [11]; Hiza, et al. [14]; Hiza and Haynes [15] and Haynes [9,10).

All of the data are for the orthobaric liquid except for the data of Miller and Hiza [25] which are for the single phase liquid phase, very close to the orthobaric conditions. The units of the data are bars, moles per liter and kelvin. The columns labeled RKM, HS, CELL and CS correspond to percentage derivations between experimental and predicted densities by the Revised Klosek and McKinley, hard sphere, cell and extended corresponding states models respectively. The derivations are always calculated using experimental-calculated densities.

MIXTURE NO 1 (Miller and Hiza [25])

0.85147 CH_4 + 0.14853 C_2H_6

P	D	T	MW	RKM	HS	CELL	CS
.977	25.7499	110.08	18.1265	.13	- .02	.02	.01

MIXTURE NO 2 (Hiza, et al. [14])

0.68006 CH_4 + 0.31994 C_2H_6

.416	25.1027	105.00	20.5309	.14	.05	.10	.07
.645	24.7802	110.00	20.5309	.07	- .01	.02	.01
.961	24.4612	115.00	20.5309	.05	- .04	-.03	-.03
1.380	24.1402	120.00	20.5309	.06	- .06	-.06	-.05
1.930	23.8212	125.00	20.5309	.08	- .05	-.04	-.05
2.620	23.5007	130.00	20.5309	.12	- .01	.00	-.02

MIXTURE NO 3 (Hiza, et al. [14])

0.49325 CH_4 + 0.50675 C_2H_6

.325	23.9619	105.00	23.1513	.14	.07	.11	.07
.503	23.6937	110.00	23.1513	.09	.03	.03	.03
.749	23.4328	115.00	23.1513	.11	.03	.01	.03
1.080	23.1559	120.00	23.1513	.07	- .02	-.06	-.03
1.500	22.8933	125.00	23.1513	.08	.00	-.05	.00
2.050	22.6290	130.00	23.1513	.14	.04	-.02	.03
2.720	22.3581	135.00	23.1513	.19	.06	.01	.05
3.550	22.0765	140.00	23.1513	.25	.05	.02	.04

MIXTURE NO 4 (Hiza, et al. [14])

0.35457 CH_4 + 0.64543 C_2H_6

.256	23.1033	105.00	25.0965	- .04	- .07	-.04	-.07
.397	22.8777	110.00	25.0965	- .01	- .04	-.05	-.05
.580	22.6478	115.00	25.0965	.03	- .02	-.06	-.03
.826	22.4035	120.00	25.0965	.00	- .06	-.11	-.07
1.146	22.1872	125.00	25.0965	.10	.04	-.03	.03
1.550	21.9441	130.00	25.0965	.10	.03	-.04	.02

MIXTURE NO 5 (Hiza, et al. [14])

0.85796 CH_4 + 0.14204 C_3H_8

P	D	T	MW	RKM	HS	CELL	CS
.517	24.9622	105.00	20.0279	.01	.10	.10	.07
.817	24.6332	110.00	20.0279	- .02	.08	.07	.06
1.199	24.2942	115.00	20.0279	- .04	.04	.03	.03
1.785	23.9492	120.00	20.0279	- .07	.00	.00	.00
2.415	23.5942	125.00	20.0279	- .09	- .06	-.04	-.05
3.290	23.2461	130.00	20.0279	- .11	- .06	-.01	-.04

MIXTURE NO 6 (Hiza, et al. [14])

0.74920 CH_4 + 0.25080 C_3H_8

.478	23.4767	105.00	23.0790	.01	.02	.03	-.02
.738	23.2064	110.00	23.0790	- .03	.00	-.01	-.03
1.099	22.9364	115.00	23.0790	- .01	.00	-.02	-.02
1.582	22.6665	120.00	23.0790	.02	.01	-.01	.01
2.216	22.3818	125.00	23.0790	- .03	- .04	-.04	-.01
3.029	22.1019	130.00	23.0790	.02	- .05	-.02	.01

MIXTURE NO 7 (Hiza, et al. [14])

0.49637 CH_4 + 0.50363 C_3H_8

.384	20.4909	105.00	30.1720	- .05	.01	.04	-.03
.591	20.3046	110.00	30.1720	- .08	.01	.01	-.02
.874	20.1180	115.00	30.1720	- .06	.01	-.02	-.02
1.250	19.9311	120.00	30.1720	.00	.01	-.03	.00
1.730	19.7471	125.00	30.1720	.04	.03	-.01	.04
2.320	19.5546	130.00	30.1720	- .01	.01	-.03	.05

MIXTURE NO 8 (Hiza, et al. [14])

0.29538 CH_4 + 0.70462 C_3H_8

.271	18.5132	105.00	35.8106	- .19	- .08	-.05	-.12
.409	18.3624	110.00	35.8106	- .24	- .09	-.10	-.12

MIXTURE NO 9 (Haynes [10])

0.92788 CH_4 + 0.07212 nC_4H_{10}

P	D	T	MW	RKM	HS	CELL	CS
1.820	24.2615	120.00	19.0779	.00	.04	-.04	-.04
2.547	23.8868	125.00	19.0779	.00	.00	-.07	-.05
3.470	23.5047	130.00	19.0779	- .01	- .03	-.09	-.07
4.616	23.1225	135.00	19.0779	.04	- .03	-.06	-.05
6.023	22.7284	140.00	19.0779	.16	- .04	-.03	-.03

MIXTURE NO 10 (Haynes [10])

0.92780 CH_4 + 0.07220 nC_4H_{10}

1.270	24.6285	115.00	19.0813	.01	.08	.01	-.01
1.824	24.2783	120.00	19.0813	.07	.11	.03	.04
2.549	23.8999	125.00	19.0813	.06	.07	-.01	.01

MIXTURE NO 11 (Hiza, et al. [14])

0.91674 CH_4 + 0.08326 nC_4H_{10}

.521	25.1536	105.00	19.5467	.21	.35	.33	.22
.810	24.7960	110.00	19.5467	.12	.26	.21	.15
1.216	24.4512	115.00	19.5467	.14	.24	.18	.15
1.753	24.0889	120.00	19.5467	.10	.19	.11	.11
2.472	23.7370	125.00	19.5467	.15	.18	.10	.13
3.374	23.3789	130.00	19.5467	.17	.19	.12	.16
4.509	23.0110	135.00	19.5467	.21	.19	.15	.18
5.887	22.6391	140.00	19.5467	.35	.20	.22	.23

MIXTURE NO 12 (Haynes [10])

0.77982 CH_4 + 0.22018 nC_4H_{10}

1.702	21.6066	120.00	25.3805	.12	.14	.11	.06
2.369	21.3549	125.00	25.3805	.11	.08	.02	.04
3.228	21.1020	130.00	25.3805	.09	.00	-.07	.03
4.291	20.8555	135.00	25.3805	.12	- .03	-.10	.07
5.576	20.5978	140.00	25.3805	.20	- .13	-.15	.07

MIXTURE NO 13 (Haynes [10])

0.77762 CH_4 + 0.22238 nC_4H_{10}

P	D	L	T	MW	RKM	HS	CEL	CS
1.179	21.8054		115.00	25.4011	.07	.15	.16	.02
1.699	21.5601		120.00	25.4011	.07	.10	.07	.02
2.372	21.3164		125.00	25.4011	.10	.06	.01	.03
3.230	21.0605		130.00	25.4011	.06	- .03	-.10	.00
4.291	20.8011		135.00	25.4011	.02	- .14	-.20	-.03
5.579	20.5448		140.00	25.4011	.09	- .23	-.26	-.03

MIXTURE NO 14 (Hiza, et al. [14])

0.58828 CH_4 + 0.41172 nC_4H_{10}

3.183	18.3058	130.00	33.3687	- .32	- .11	-.15	-.08
2.342	18.4853	125.00	33.3687	- .17	.00	.00	-.03
2.281	18.4772	125.00	33.3687	- .21	- .05	-.04	-.08
1.636	18.6495	120.00	33.3687	- .18	.02	.07	-.06

MIXTURE NO 15 (Haynes [10])

0.92044 CH_4 + 0.07956 iC_4H_{10}

1.254	24.3633	115.00	19.3910	- .21	.05	.06	-.01
1.805	24.0029	120.00	19.3910	- .24	.02	.03	-.03
2.521	23.6403	125.00	19.3910	- .22	.01	.04	-.03
3.434	23.2752	130.00	19.3910	- .21	.03	.08	.00
4.567	22.8920	135.00	19.3910	- .22	.00	.09	-.02
5.950	22.5037	140.00	19.3910	- .14	- .02	.14	-.02

MIXTURE NO 16 (Haynes [10])

0.78329 CH_4 + 0.21671 iC_4H_{10}

.782	21.9144	110.00	25.1625	- .33	.10	.15	.01
1.164	21.6652	115.00	25.1625	- .33	.05	.07	-.01
1.671	21.4136	120.00	25.1625	- .35	.01	.00	-.02
2.329	21.1668	125.00	25.1625	- .34	.00	-.03	.00
3.170	20.9125	130.00	25.1625	- .35	- .04	-.07	.00
4.208	20.6629	135.00	25.1625	- .33	- .06	-.06	.04
5.474	20.4082	140.00	25.1625	- .23	- .09	-.04	.07

MIXTURE NO 17 (Hiza, et al. [14])

0.48687 CH_4 + 0.51313 iC_4H_{10}

P	D	T	MW	RKM	HS	CELL	CS
.629	17.3575	110.00	37.6362	- .62	.05	.11	.02
.938	17.2076	115.00	37.6362	- .70	.00	-.01	-.02
1.361	17.0639	120.00	37.6362	- .64	- .02	-.08	-.02
1.852	16.9156	125.00	37.6362	- .69	- .07	-.17	-.04

MIXTURE NO 18 (Hiza, et al. [14])

0.95248 CH_4 + 0.04752 N_2

1.380	26.8476	105.00	16.6119	- .05	- .04	-.04	-.02
1.990	26.4052	110.00	16.6119	.06	.00	.00	.04
2.634	25.9374	115.00	16.6119	.20	.01	.02	.06
3.500	25.4522	120.00	16.6119	.26	*	.03	.07
4.600	24.9496	125.00	16.6119	.54	*	.04	.07
5.830	24.4210	130.00	16.6119	.69	-5.69	.03	.05
7.300	23.8600	135.00	16.6119	.77	-3.99	-.01	-.03
9.200	23.2809	140.00	16.6119	1.09	-2.69	-.02	-.07

MIXTURE NO 19 (Hiza, et al. [14])

0.69651 CH_4 + 0.30349 N_2

3.450	26.8735	100.00	19.6759	- .39	.06	.09	.00
4.661	26.3393	105.00	19.6759	- .56	.02	.07	.04
6.181	25.7686	110.00	19.6759	- .92	- .05	-.02	.00
8.010	25.1790	115.00	19.6759	-1.28	.13	-.08	-.03
10.112	24.5737	120.00	19.6759	-1.56	1.88	-.07	-.04

MIXTURE NO 20 (Hiza, et al. [14])

0.50758 CH_4 + 0.49242 N_2

3.303	27.0801	95.00	21.9375	-2.16	.11	.14	.06
4.468	26.4588	100.00	21.9375	-1.92	- .01	.05	.04
6.383	25.8106	105.00	21.9375	-1.56	- .15	-.05	-.01
8.461	25.1387	110.00	21.9375	-1.29	- .17	-.09	-.03
10.740	24.4431	115.00	21.9375	1.12	.28	-.04	-.01
13.983	23.7096	120.00	21.9375	-1.79	3.41	.07	-.01
17.529	22.9315	125.00	21.9375	-1.14	*	.26	.00
21.076	22.1005	130.00	21.9375	- .72	-2.84	.05	.10

* The hard sphere solution for the density of N_2 failed.

MIXTURE NO 21 (Hiza, et al. [14])

0.67287 C_2H_6 + 0.32713 C_3H_8

P	D	T	MW	RKM	HS	CELL	CS
	18.6192	125.00	34.6588	.00	.02	-.02	.00
	18.4648	130.00	34.6588	.00	.02	.00	.01
	18.3059	135.00	34.6588	.00	.00	.00	.00
	18.1509	140.00	34.6588	.00	.01	.04	.02

MIXTURE NO 22 (Hiza, et al. [14])

0.50105 C_2H_6 + 0.49895 C_3H_8

18.3618	105.00	37.0689		.02	.04	.02
18.2169	110.00	37.0689		.01	.00	.00
18.0726	115.00	37.0689		.00	-.03	-.01
17.9282	120.00	37.0689		- .01	-.05	-.01
17.7880	125.00	37.0689		.00	-.03	.00
17.6412	130.00	37.0689		- .02	-.04	-.02
17.4988	135.00	37.0689		- .01	-.01	-.01
17.3526	140.00	37.0689		- .03	.00	-.01

MIXTURE NO 23 (Hiza, et al. [14])

0.67117 C_2H_6 + 0.32883 nC_4H_{10}

17.5047	110.00	39.2952		.00	.02	.00
17.3706	115.00	39.2952		.01	.00	.00
17.1031	125.00	39.2952		.02	.00	.02
16.9626	130.00	39.2952		- .01	-.03	-.02
16.8285	135.00	39.2952		.00	-.01	-.01
16.6947	140.00	39.2952		.00	.03	.01

MIXTURE NO 24 (Hiza, et al. [14])

0.65343 C_2H_6 + 0.34657 nC_4H_{10}

17.2184	115.00	39.7929		.00	.00	.00
17.0824	120.00	39.7929		- .02	-.03	-.02

MIXTURE NO 25 (Hiza, et al. [14])

0.72436 C_2H_6 + 0.27564 iC_4H_{10}

17.9779	105.00	37.8030		.01	.01	.01
17.8401	110.00	37.8030		.01	.00	.01
17.4235	125.00	37.8030		.01	.00	.01
17.2825	130.00	37.8030		.00	.01	.01

MIXTURE NO 26 (Hiza, et al. [14])

0.68939 C_2H_6 + 0.31061 iC_4H_{10}

P	D	T	MW	RKM	HS	CELL	CS
	17.3716	115.00	38.7840		.00	-.03	.00
	17.2344	120.00	38.7840		- .02	-.04	-.02

MIXTURE NO 27 (Hiza, et al. [14])

0.94067 C_2H_6 + 0.05933 N_2

3.850	21.4718	105.00	29.9481		- .02	-.01	-.03
4.630	21.2912	110.00	29.9481		.03	.02	.03
5.472	21.0845	115.00	29.9481		- .01	-.04	-.04
6.383	20.8998	120.00	29.9481		- .72	.01	.00

MIXTURE NO 28 (Hiza, et al. [14])

0.60949 C_3H_8 + 0.39051 nC_4H_{10}

	14.6487	115.00	49.5749		- .01	-.02	-.01
	14.5521	120.00	49.5749		.02	.00	.02

MIXTURE NO 29 (Hiza, et al. [14])

0.60650 C_3H_8 + 0.39350 nC_4H_{10}

	14.1343	140.00	49.6169		.00	-.03	.00
	14.0333	145.00	49.6169		.00	.00	.00
	13.9346	150.00	49.6169		.02	.05	.02

MIXTURE NO 30 (Hiza, et al. [14])

0.58692 C_3H_8 + 0.41308 nC_4H_{10}

	14.6839	110.00	49.8915		.01	.04	.01
	14.1748	135.00	49.8915		- .03	-.06	-.03
	14.0786	140.00	49.8915		.00	-.02	.00

MIXTURE NO 31 (Hiza, et al., [14])

0.49030 C_3H_8 + 0.50970 iC_4H_{10}

	14.3080	105.00	51.2468		- .01	-.04	-.01
	14.2136	110.00	51.2468		- .01	-.05	.00
	14.1219	115.00	51.2468		.02	-.03	.02
	14.0257	120.00	51.2468		.01	-.04	.01
	13.9300	125.00	51.2468		.00	-.04	.00
	13.8342	130.00	51.2468		- .01	-.04	-.01

MIXTURE NO 32 (Hiza, et al. [14])

0.50326 C_3H_8 + 0.49674 iC_4H_{10}

P	D	T	MW	RKM	HS	CELL	CS
	13.9718	125.00	51.0650		.01	-.03	.01
	13.8737	130.00	51.0650		- .01	-.04	-.01

MIXTURE NO 33 (Hiza, et al. [14])

0.97986 C_3H_8 + 0.02014 N_2

3.567	16.2131	110.00	43.7733		- .01	-.04	-.01
4.712	16.0931	115.00	43.7733		- .02	-.05	-.02

MIXTURE NO 34 (Hiza, et al. [14])

0.96206 C_3H_8 + 0.03794 N_2

4.955	16.4638	105.00	43.4870		.02	-.03	.01
6.708	16.3410	110.00	43.4870		.00	-.04	.00

MIXTURE NO 35 (Hiza, et al. [14])

0.93260 C_3H_8 + 0.06740 N_2

8.795	16.7084	105.00	43.0132		.20	.11	.18
6.313	16.8055	100.00	43.0132		.04	-.07	.01

MIXTURE NO 36 (Hiza, et al. [14])

0.52961 nC_4H_{10} + 0.47039 iC_4H_{10}

	12.6943	125.00	58.1243		- .01	-.03	-.01
	12.6133	130.00	58.1243		.01	-.01	.01
	12.5271	135.00	58.1243		- .01	-.04	-.01
	12.4447	140.00	58.1243		.00	-.02	.00

MIXTURE NO 37 (Hiza and Haynes [15])

0.80284 CH_4 + 0.09902 C_2H_6 + 0.09814 C_3H_8

.476	24.9975	105.00	20.1852	.02	.04	.06	.02
.747	24.6696	110.00	20.1852	- .03	.00	.00	-.02
1.119	24.3515	115.00	20.1852	.01	.02	.02	.01
1.616	24.0335	120.00	20.1852	.07	.07	.07	.06

MIXTURE NO 38 (Hiza and Haynes [15])

0.34242 CH_4 + 0.31372 C_2H_6 + 0.34386 C_3H_8

P	D	T	MW	RKM	HS	CELL	CS
.427	20.5342	110.00	30.0903	- .03	.02	.00	-.03
.636	20.3497	115.00	30.0903	- .01	.03	-.02	-.02
.638	20.3567	115.00	30.0903	.03	.06	.01	.01
.914	20.1786	120.00	30.0903	.12	.10	.04	.06
.914	20.1803	120.00	30.0903	.12	.11	.05	.07
1.280	19.9858	125.00	30.0903	.10	.08	.01	.04

MIXTURE NO 39 (Hiza and Haynes [15])

0.67040 CH_4 + 0.16610 C_2H_6 + 0.16350 N_2

3.688	25.8587	105.00	20.3301	.57	.01	.28	.00
4.813	25.4598	110.00	20.3301	.64	- .03	.27	-.01
6.181	25.0466	115.00	20.3301	.79	.02	.25	-.05
7.731	24.6359	120.00	20.3301	.70	.42	.31	-.04

MIXTURE NO 40 (Hiza and Haynes [15])

0.34140 CH_4 + 0.32060 C_2H_6 + 0.33800 N_2

7.190	24.9084	105.00	24.5861	2.06	.01	.61	.07
9.494	24.5350	110.00	24.5861	2.55	- .01	.64	.04
12.372	24.1433	115.00	24.5861	3.72	.24	.65	-.06

MIXTURE NO 41 (Miller and Hiza [25])

0.86786 CH_4 + 0.08181 C_3H_8 + 0.05033 iC_4H_{10}

.466	24.9239	100.00	20.4561	- .17	.04	.11	-.03
.769	24.4200	108.00	20.4561	- .17	.01	.05	-.04
1.327	23.9676	115.00	20.4561	- .18	- .01	.02	-.05

MIXTURE NO 42 (Hiza and Haynes [15])

0.79770 CH_4 + 0.10280 C_3H_8 + 0.09950 N_2

2.817	25.3950	105.00	20.1181	.17	.08	.24	-.01
3.620	25.0153	110.00	20.1181	.10	.02	.23	-.05
4.549	24.6465	115.00	20.1181	.08	.08	.33	.01
5.610	24.2667	120.00	20.1181	.02	*	.44	.04

* The hard sphere solution for the density of N_2 failed.

MIXTURE NO 43 (Haynes [9])

0.89071 CH_4 + 0.04998 nC_4H_{10} + 0.05931 N_2

P	D	T	MW	RKM	HS	CELL	CS
2.400	25.3450	110.00	18.8562	- .23	.00	.01	.03
3.145	24.9440	115.00	18.8562	- .25	- .04	-.02	.01
4.082	24.5383	120.00	18.8562	- .38	*	-.02	.00
5.196	24.1141	125.00	18.8562	- .32	*	-.04	-.04

MIXTURE NO 44 (Miller and Hiza [25])

0.85317 CH_4 + 0.05077 C_2H_6 + 0.04855 C_3H_8 + 0.04751 nC_4H_{10}

1.038	24.6056	110.04	20.1165	.02	.15	.10	.06
1.464	24.2712	115.00	20.1165	.00	.11	.04	.02

MIXTURE NO 45 (Haynes [9])

0.85133 CH_4 + 0.05759 C_2H_6 + 0.04808 C_3H_8 + 0.04300 nC_4H_{10}

1.180	24.3243	115.00	20.0092	- .04	.07	.00	-.01
1.700	23.9965	120.00	20.0092	.00	.10	.02	.03
2.374	23.6586	125.00	20.0092	.05	.11	.04	.05
3.232	23.3108	130.00	20.0092	.04	.11	.05	.05
4.301	22.9634	135.00	20.0092	.08	.13	.11	.09

MIXTURE NO 46 (Haynes [9])

0.84566 CH_4 + 0.07924 C_2H_6 + 0.05060 C_3H_8 + 0.02450 nC_4H_{10}

1.167	24.5569	115.00	19.6051	.00	.04	.00	-.01
1.683	24.2126	120.00	19.6051	.02	.04	-.01	-.01
2.350	23.8698	125.00	19.6051	.08	.07	.03	.03
3.201	23.5204	130.00	19.6051	.12	.11	.09	.07

MIXTURE NO 47 (Haynes [9])

0.86040 CH_4 + 0.04600 C_2H_6 + 0.04790 C_3H_8 + 0.04570 iC_4H_{10}

1.186	24.2654	115.00	19.9552	- .14	.01	.04	-.03
1.710	23.9371	120.00	19.9552	- .10	.04	.07	.01
2.387	23.5860	125.00	19.9552	- .10	.01	.05	-.01
3.248	23.2331	130.00	19.9552	- .13	- .01	.06	-.02
4.320	22.8637	135.00	19.9552	- .18	- .07	.05	-.07

* The hard sphere solution for the density of N_2 failed.

MIXTURE NO 48 (Haynes [9])

0.85378 CH_4 + 0.05178 C_2H_6 + 0.04703 C_3H_8 + 0.04741 iC_4H_{10}

P	D	T	MW	RKM	HS	CELL	CS
1.190	24.2100	115.00	20.0838	- .14	.01	.04	-.03
1.706	23.8779	120.00	20.0838	- .14	.01	.04	-.02
2.379	23.5324	125.00	20.0838	- .13	- .02	.02	-.04
3.238	23.1834	130.00	20.0838	- .15	- .04	.03	-.05

MIXTURE NO 49 (Miller and Hiza [25])

0.85378 CH_4 + 0.05178 C_2H_6 + 0.04703 C_3H_8 + 0.04741 iC_4H_{10}

.972	24.5434	110.02	20.0838	- .14	.03	.06	-.01
1.429	24.2154	115.01	20.0838	- .12	.02	.05	-.01

MIXTURE NO 50 (Hiza and Haynes [15])

0.85260 CH_4 + 0.04830 C_2H_6 + 0.05070 C_3H_8 + 0.04840 N_2

1.581	25.8910	105.00	18.7223	.01	- .04	.03	-.07
2.138	25.5081	110.00	18.7223	- .09	- .07	.03	-.07
2.827	25.1224	115.00	18.7223	- .05	- .06	.05	-.06
3.650	24.7283	120.00	18.7223	- .08	*	.08	-.06

MIXTURE NO 51 (Haynes [9])

0.85892 CH_4 + 0.11532 C_2H_6 + 0.01341 C_3H_8 + 0.00705 nC_4H_{10} + 0.00530 iC_4H_{10}

1.185	25.0957	115.00	18.5565	.05	- .02	-.01	-.03
1.706	24.7131	120.00	18.5565	.03	- .06	-.05	-.07
2.372	24.3294	125.00	18.5565	.02	- .07	-.05	-.09
3.225	23.9490	130.00	18.5565	.05	- .02	.01	-.06

MIXTURE NO 52 (Hiza and Haynes [15])

0.85442 CH_4 + 0.05042 C_2H_6 + 0.04038 C_3H_8 + 0.02901 nC_4H_{10} + 0.02577 iC_4H_{10}

.515	24.8775	105.00	20.1885	.09	.25	.25	.16
.818	24.5382	110.00	20.1885	.00	.17	.15	.09
1.190	24.2083	115.00	20.1885	.00	.15	.12	.08
1.695	23.8859	120.00	20.1885	.05	.19	.15	.13

MIXTURE NO 53 (Haynes [9])

0.84558 CH_4 + 0.08153 C_2H_6 + 0.04778 C_3H_8 + 0.01252 nC_4H_{10} + 0.01259 iC_4H_{10}

1.166	24.5586	115.00	19.5838	.01	.05	.04	.01
1.680	24.2180	120.00	19.5838	.04	.07	.05	.03
2.348	23.8688	125.00	19.5838	.08	.08	.08	.04
3.188	23.5154	130.00	19.5838	.10	.10	.12	.07

* The hard sphere solution for the density of N_2 failed.

MIXTURE NO 54 (Haynes [9])

0.81249 CH_4 + 0.08484 C_2H_6 + 0.04931 C_3H_8 + 0.02708 nC_4H_{10} + 0.02628 N_2

P	D	T	MW	RKM	HS	CELL	CS
2.214	24.4562	115.00	20.0706	.01	.01	.03	-.02
2.874	24.1119	120.00	20.0706	.00	*	.04	-.01
3.768	23.7507	125.00	20.0706	.02	*	.02	-.05
4.793	23.3954	130.00	20.0706	- .02	-3.67	.07	-.03

MIXTURE NO 55 (Hiza and Haynes [15])

0.79090 CH_4 + 0.05600 C_2H_6 + 0.05000 C_3H_8 + 0.04770 nC_4H_{10} + 0.05540 N_2

1.933	24.8080	105.00	20.9017	.04	.10	.12	.05
2.530	24.4664	110.00	20.9017	.02	.02	.03	-.02

MIXTURE NO 56 (Miller and Hiza [25])

0.79054 CH_4 + 0.05597 C_2H_6 + 0.04996 C_3H_8 + 0.04762 nC_4H_{10} + 0.05591 N_2

2.158	24.8354	105.03	20.9029	.15	.21	.23	.15

MIXTURE NO 57 (Haynes [9])

0.80940 CH_4 + 0.04542 C_2H_6 + 0.05050 C_3H_8 + 0.04667 iC_4H_{10} + 0.04801 N_2

3.005	24.1487	115.00	20.6355	.02	-.02	-.03	-.04
3.863	23.8075	120.00	20.6355	- .14	*	.00	-.03
4.874	23.4518	125.00	20.6355	- .12	*	.01	-.05
6.125	23.0893	130.00	20.6355	- .18	-5.06	.05	-.08

MIXTURE NO 58 (Hiza and Haynes [15])

0.80600 CH_4 + 0.04680 C_2H_6 + 0.04820 C_3H_8 + 0.05000 iC_4H_{10} + 0.04900 N_2

2.039	24.7803	105.00	20.7423	- .03	.08	.03	.01
2.550	24.4327	110.00	20.7423	- .09	- .02	-.06	-.06
3.135	24.1039	115.00	20.7423	.06	.00	-.02	-.02
3.790	23.7707	120.00	20.7423	- .10	*	-.01	-.02

MIXTURE NO 59 (Miller and Hiza [25])

0.80545 CH_4 + 0.04671 C_2H_6 + 0.04817 C_3H_8 + 0.04998 iC_4H_{10} + 0.04969 N_2

2.273	24.7831	105.06	20.7476	.00	.10	.05	.03

* The hard sphere solution for the density of N_2 failed.

MIXTURE NO 60 (Haynes [9])

0.90613 CH_4 + 0.06026 C_2H_6 + 0.02154 C_3H_8 + 0.00306 nC_4H_{10} + 0.00300 iC_4H_{10} + 0.00601 N_2

P	D	T	MW	RKM	HS	CELL	CS
1.478	25.3834	115.00	17.8195	.03	- .01	.01	-.01
2.043	24.9894	120.00	17.8195	.08	.00	.05	.02
2.785	24.5702	125.00	17.8195	.04	.00	.03	-.02
3.722	24.1578	130.00	17.8195	.06	-1.08	.11	.02

MIXTURE NO 61 (Haynes [9])

0.88225 CH_4 + 0.07259 C_2H_6 + 0.02561 C_3H_8 + 0.00492 nC_4H_{10} + 0.00490 iC_4H_{10} + 0.00973 N_2

1.639	25.2023	115.00	18.3094	.09	.04	.07	.04
2.247	24.8047	120.00	18.3094	.06	.00	.04	.00
3.022	24.4022	125.00	18.3094	.04	.00	.03	-.03

MIXTURE NO 62 (Haynes [9])

0.85934 CH_4 + 0.08477 C_2H_6 + 0.02980 C_3H_8 + 0.00707 nC_4H_{10} + 0.00519 iC_4H_{10} + 0.01383 N_2

1.812	25.0384	115.00	18.7496	.12	.06	.09	.06
2.441	24.6661	120.00	18.7496	.12	*	.11	.07
3.223	24.2880	125.00	18.7496	.19	.00	.16	.09
4.222	23.8981	130.00	18.7496	.17	-2.12	.20	.10

MIXTURE NO 63 (Hiza and Haynes [15])

0.81300 CH_4 + 0.04750 C_2H_6 + 0.04870 C_3H_8 + 0.02420 nC_4H_{10} + 0.02410 iC_4H_{10} + 0.04250 N_2

1.834	24.8496	105.00	20.6168	.03	.12	.10	.05
2.807	24.5159	110.00	20.6168	.02	.08	.06	.03
3.384	24.1783	115.00	20.6168	.09	.06	.05	.03
6.039	23.8577	120.00	20.6168	.04	*	.11	.08

MIXTURE NO 64 (Haynes [9])

0.85341 CH_4 + 0.07898 C_2H_6 + 0.04729 C_3H_8 + 0.00992 nC_4H_{10} + 0.00854 iC_4H_{10} + 0.00089 nC_5H_{12} + 0.00097 iC_5H_{12}

.787	25.0063	110.00	19.3588	.01	.02	.02	-.01
1.172	24.6566	115.00	19.3588	.00	- .01	-.01	-.03
1.686	24.3079	120.00	19.3588	.02	.00	.00	-.02
2.351	23.9525	125.00	19.3588	.05	.01	.03	-.01
3.210	23.5883	130.00	19.3588	.06	.02	.06	.00

* The hard sphere solution for the density of N_2 failed.

MIXTURE NO 65 (Haynes [9])

0.75442 CH_4 + 0.15401 C_2H_6 + 0.06950 C_3H_8 + 0.01057 nC_4H_{10} + 0.00978 iC_4H_{10} + 0.00083 nC_5H_{12} + 0.00089 iC_5H_{12}

P	D	T	MW	RKM	HS	CELL	CS
.723	24.2529	110.00	21.1060	- .02	.00	-.01	-.04
1.081	23.9619	115.00	21.1060	.04	.04	.03	.01
1.549	23.6535	120.00	21.1060	.07	.04	.02	.01
2.153	23.3351	125.00	21.1060	.04	.01	.00	-.01

MIXTURE NO 66 (Haynes [9])

0.75713 CH_4 + 0.13585 C_2H_6 + 0.06742 C_3H_8 + 0.01326 nC_4H_{10} + 0.01336 iC_4H_{10} + 0.00859 N_2 + 0.00216 nC_5H_{12} + 0.00223 iC_5H_{12}

1.155	24.1809	110.00	21.3094	.09	.07	.08	.04
1.595	23.8731	115.00	21.3094	.12	.05	.05	.02
2.155	23.5709	120.00	21.3094	.12	.00	.08	.05
2.873	23.2644	125.00	21.3094	.15	.00	.12	.08
3.744	22.9514	130.00	21.3094	.22	-1.42	.16	.11

MIXTURE NO 67 (Haynes [9])

0.74275 CH_4 + 0.16505 C_2H_6 + 0.06547 C_3H_8 + 0.00893 nC_4H_{10} + 0.00843 iC_4H_{10} + 0.00801 N_2 + 0.00067 nC_5H_{12} + 0.00069 iC_5H_{12}

1.158	24.3141	110.00	21.0976	.02	- .02	.00	-.05
1.584	24.0160	115.00	21.0976	.09	.01	.02	-.01
2.093	23.6937	120.00	21.0976	.03	.00	-.03	-.06
2.853	23.3804	125.00	21.0976	.05	.00	-.01	-.05

MIXTURE NO 68 (Haynes [9])

0.90068 CH_4 + 0.06537 C_2H_6 + 0.02200 C_3H_8 + 0.00284 nC_4H_{10} + 0.00291 iC_4H_{10} + 0.00599 N_2 + 0.00011 nC_5H_{12} + 0.00010 iC_5H_{12}

1.456	25.3600	115.00	17.9026	.05	.01	.03	.01
2.024	24.9656	120.00	17.9026	.08	.00	.06	.03
2.762	24.5450	125.00	17.9026	.03	.00	.03	-.03
3.698	24.1289	130.00	17.9026	.02	-1.13	.07	-.02

Appendix B. Computer Program and Equation Parameters for the Extended Corresponding States Model

The program listings in Appendix F include the extended corresponding states method described in section 2. To use the program in its present form one must make one of the two possible calls to SUBROUTINE PDMIX(P,D,T,X) (lines LNG 1 through LNG 9). The two possible calls are:

```
CALL PDMIX(P,D,T,X)
```

or

```
CALL PMIX(P,D,T,X)
```

When the call to PDMIX(P,D,T,X) is made the input variables are: P (pressure in bars); T (temperature in kelvin); and X which is a matrix of the mole fraction of the components of the mixture in the following order:

X(1) = mole fraction of methane

X(2) = mole fraction of ethane

X(3) = mole fraction of propane

X(4) = mole fraction of normal butane

X(5) = mole fraction of isobutane

X(6) = mole fraction of nitrogen

X(7) = mole fraction of normal pentane

X(8) = mole fraction of isopentane

No other components are allowed and if one or more of the above components are absent, a zero should be inserted in the appropriate matrix element. The program then calculates a density and it is returned in the argument list as D (density in moles/liter).

When a call to PMIX(P,D,T,X) is made all of the above are the same except that the roles of P and D are interchanged, i.e., D is an input variable and P is calculated by the program.

The range of the program is 90 to 150 kelvin for the saturated liquid phase of any of the pure components of CH_4, C_2H_6, C_3H_8, nC_4H_{10}, iC_4H_{10}, N_2, nC_4H_{12} or iC_4H_{12} or any mixture of those fluids. The program will extrapolate to higher pressures (higher than saturation pressure) but the user is reminded that such a calculation is an extrapolation and should be used with caution.

Other subprograms required:

SUBROUTINE MIX DATA, line LNG 95, Appendix F

SUBROUTINE DATA CH4, line LNG 162, Appendix F

FUNCTION FINDM, line LNG 217, Appendix F

FUNCTION SATL, line LNG 236, Appendix F

SUBROUTINE PROPS, line LNG 248, Appendix F

The equation of state from which Z_0 and G_0 by eqs (1) and (2) may be derived is:

$$P = \rho RT + \rho^2(N_1T + N_2T^{1/2} + N_3 + N_4/T + N_5/T^2)$$

$$+ \rho^3(N_6T + N_7 + N_8/T + N_9/T^2)$$

$$+ \rho^4(N_{10}T + N_{11} + N_{12}/T) + \rho^5(N_{13})$$

$$+ \rho^6(N_{14}/T + N_{15}/T^2) + \rho^7(N_{16}/T)$$

$$+ \rho^8(N_{17}/T + N_{18}/T^2) + \rho^9(N_{19}/T^2)$$

$$+ \rho^3(N_{20}/T^2 + N_{21}/T^3)\ \exp(-\gamma\rho^2) \qquad \text{(B-1)}$$

$$+ \rho^5(N_{22}/T^2 + N_{23}/T^4)\ \exp(-\gamma\rho^2)$$

$$+ \rho^7(N_{24}/T^2 + N_{25}/T^3)\ \exp(-\gamma\rho^2)$$

$$+ \rho^9(N_{26}/T^2 + N_{27}/T^4)\ \exp(-\gamma\rho^2)$$

$$+ \rho^{11}(N_{28}/T^2 + N_{29}/T^3)\ \exp(-\gamma\rho^2)$$

$$+ \rho^{13}(N_{30}/T^2 + N_{31}/T^3 + N_{32}/T^4)\ \exp(-\gamma\rho^2)$$

The computer subroutine PROPS(PP,DD,TT), lines "LNG 248 through LNG 371 in Appendix F" are the FORTRAN statements for eq (B-1) and the derivative $(\partial P/\partial\rho)_T$.

The parameters for eq (B-1) as applied to methane are given in table 2 and in the FORTRAN SUBROUTINE DATA CH4, lines LNG 162 through LNG 216, Appendix F.

The parameters for eqs (5) and (6), (9) and (10) are given in table 3 and 4 and in the FORTRAN SUBROUTINE MIX DATA (IBASE), lines LNG 95 through LNG 161, Appendix F.

Table 2. Methane Coefficients N_i for Eq B-1.

$R = 0.08205616$

$\gamma = 0.0096$

$N_1 = -0.187027997685 \times 10^{-1}$

$N_2 = 0.103387108009 \times 10$

$N_3 = -0.155387625619 \times 10^{2}$

$N_4 = 0.772311478564 \times 10^{3}$

$N_5 = -0.377103300895 \times 10^{5}$

$N_6 = 0.846818843475 \times 10^{-3}$

$N_7 = -0.496415884529$

$N_8 = 0.869909352414 \times 10^{2}$

$N_9 = -0.322821592493 \times 10^{5}$

$N_{10} = -0.395843026318 \times 10^{-4}$

$N_{11} = 0.266772318035 \times 10^{-1}$

$N_{12} = -0.304010057839 \times 10$

$N_{13} = 0.191584507536 \times 10^{-3}$

$N_{14} = -0.195587933458 \times 10^{-3}$

$N_{15} = 0.607479967879 \times 10$

$N_{16} = -0.529609525984 \times 10^{-3}$

$N_{17} = 0.152264286004 \times 10^{-4}$

$N_{18} = -0.109952182842 \times 10^{-1}$

$N_{19} = 0.191395549929 \times 10^{-3}$

$N_{20} = 0.386470003746 \times 10^{5}$

$N_{21} = -0.157930582612 \times 10^{7}$

$N_{22} = 0.195270144401 \times 10^{3}$

$N_{23} = 0.165996081629 \times 10^{7}$

$N_{24} = 0.603051146711$

$N_{25} = 0.376485162808 \times 10^{2}$

$N_{26} = 0.125593680622 \times 10^{-2}$

$N_{27} = -0.343570032513 \times 10^{2}$

$N_{28} = -0.540945094139 \times 10^{-5}$

$N_{29} = 0.185622284663 \times 10^{-2}$

$N_{30} = 0.770786979245 \times 10^{-8}$

$N_{31} = -0.286868318650 \times 10^{-5}$

$N_{32} = 0.372376961647 \times 10^{-4}$

Table 3. Coefficients for Eqs 5 and 6.

n_1 = -0.109495 n_4 = -4.14192

n_2 = 0.919454 n_5 = 0.444850

n_3 = -4.01525

n_6 = 0.356808 n_8 = 0.893323

n_7 = 1.02619 n_9 = 0.761533

	w	P_c^* (bar)	T_c (K)	V_c (cm^3/mol)	M	Fluid No.
CH_4	0.0109	45.956967	190.555	98.522	16.04303	1
C_2H_6	0.110427	48.60314	305.5	146.2	30.07012	2
C_3H_8	0.154837	42.445123	370.	200.	44.09721	3
nC_4H_{10}	0.176372	38.295398	425.	251.62	58.1243	4
iC_4H_{10}	0.150115	36.88998	408.1	263.	58.1243	5
N_2	0.0291791	33.542557	126.2	89.827	28.0134	6
nC_5H_{12}	0.234320	33.812152	469.6	304.	72.15139	7
iC_5H_{12}	0.288886	31.988302	460.39	306.	72.15139	8

*Note: The large number of significant figures given for critical pressure is necessary to reproduce the Z_c ($Z_c = P_cV_c/RT_c$) in the least squares fit of the data. P_c's have been converted to bar from atmospheres.

Table 4. Binary Interaction Coefficients for Eqs 9 and 10.
Fluid numbers are given in table 3.

ij

i	1	2	3	4	5	6	7	8
1	1	1.00514	1.01922	1.04243	1.05048	1.01009	1.06	1.06
2		1	1.00599	1.01616	1.02369	1.02127	1.02	1.02
3			1	1.00172	1.01140	1.04606	1.01	1.01
4				1	0.997114	1.20955	1.0	1.0
5					1	1.13889	1.0	1.0
6						1	1.0	1.0
7							1	1.0
8								1

ij

i j	1	2	3	4	5	6	7	8
1	1	1.01127	0.988608	0.983130	0.986978	0.953430	0.98	0.98
2		1	0.999961	0.972223	0.998886	0.939622	0.99	0.99
3			1	0.985547	1.03099	0.912209	0.99	0.99
4				1	0.976416	0.849200	0.99	0.99
5					1	0.857310	0.99	0.99
6						1	0.99	0.99
7							1	1.0
8								1

Table 5. Values for Checking Calculations Using Corresponding States Equations.*

$0.6975\ CH_4 + 0.156\ C_2H_6 + 0.092\ C_3H_8 + 0.029\ nC_4H_{10} + 0.014\ iC_4H_{10} + 0.0115\ N_2$

Temperature K	Density in moles/liter	Pressure, bar
95	24.333	1
100	24.067	1.1
105	23.796	1.2

* Included for check purposes only; these values are calculated from the corresponding states model and are not experimental data.

Appendix C. Computer Program and Equation Parameters for the Hard Sphere Model

The program listing that follows is for the hard sphere model described in section 3. To use the program in its present form one must make the following reference to the computer program:

DEN = RODEN(P,T,X)

where DEN is density in moles/liter, P is pressure in bars, T is temperature in kelvin, and X is a matrix of the mole fractions of the components of the mixture in the following order:

X(1) = mole fraction of methane

X(2) = mole fraction of ethane

X(3) = mole fraction of propane

X(4) = mole fraction of normal butane

X(5) = mole fraction of isobutane

X(6) = mole fraction of nitrogen

X(7) = mole fraction of normal pentane

X(8) = mole fraction of isopentane

Note: the inclusion of the pentanes is due to Rodosevich and Miller [33] and no optimization of parameters has been included in this work for mixtures with pentane as a component.

The range of the program is 90 to 150 kelvin for the saturated liquid phase of mixtures of CH_4, C_2H_6, C_3H_8, nC_4H_{10}, iC_4H_{10}, N_2, nC_5H_{12} and iC_5H_{12}. The program will calculate densities of any of the pure components but they will be from a different model (i.e., some from an equation of state (CH_4 and N_2) and some from the equations for saturated liquid densities. Therefore in its present form, extrapolation to higher pressures is possible but the reliability of the results is questionable.

Other subprograms required:

SUBROUTINE FM, lines LNG 435 to 529, Appendix F

SUBROUTINE ZERO, lines LNG 617 to 623, Appendix F

FUNCTION FIND V1, lines LNG 530 to 548, Appendix F

FUNCTION FIND G1, lines LNG 549 to 560, Appendix F

FUNCTION EXCESS, lines LNG 561 to 616, Appendix F

FUNCTION FIND M, lines LNG 217 to 234, Appendix F

SUBROUTINE PROPS, lines LNG 248 to 371, Appendix F

SUBROUTINE DATA CH4, lines LNG 162 to 216, Appendix F

SUBROUTINE DATA N2, lines LNG 372 to 415, Appendix F

The parameters for eq (11), section 3 are given in table 6, and in lines LNG 462 through LNG 466 in Appendix F.

The binary interaction parameters j_{ij} and k_{ij} in eqs (15) and (16), section 3 are given in table 7 and in lines LNG 447 through LNG 461 in Appendix F.

Table 6. Coefficients for Eq 11.

Fluid	a_i	S_i	c_i	Fluid No.
CH_4	2.755×10^5	3.676×10^{-8}	1.00	1
C_2H_6	7.773×10^5	4.158×10^{-8}	1.50	2
C_3H_8	14.165×10^5	4.644×10^{-8}	1.67	3
nC_4H_{10}	22.733×10^5	5.051×10^{-8}	1.83	4
iC_4H_{10}	21.279×10^5	5.056×10^{-8}	1.79	5
N_2	1.718×10^5	3.546×10^{-8}	1.03	6
nC_5H_{12}	30.550×10^5	5.389×10^{-8}	1.91	7
iC_5H_{12}	42.946×10^5	5.706×10^{-8}	2.11	8

$$b_i = (2)(3.14159)(6.025 \times 10^{23})S_i^3/3$$

Table 7. Binary Interaction Parameters for Eqs 15 and 16.

j_{ij}

i \ j	1	2	3	4	5	6	7	8
1	0	-0.388616×10^{-2}	-0.120932×10^{-1}	-0.231577×10^{-1}	-0.238349×10^{-1}	-0.997547×10^{-2}	-0.326×10^{-1}	0.458×10^{-1}
2		0	-0.2162×10^{-2}	-0.400910×10^{-2}	-0.812712×10^{-2}	-0.143976×10^{-1}	-0.3×10^{-2}	-0.4×10^{-2}
3			0	0.761571×10^{-3}	-0.383743×10^{-2}	-0.24014×10^{-1}	0.0	0.0
4				0	0.222150×10^{-2}	-0.576043×10^{-1}	0.0	0.0
5					0	-0.576043×10^{-1}	0.0	0.0
6						0	0.4×10^{-1}	0.5×10^{-1}
7							0.0	0.0
8								0.0

k_{ij}

i \ j	1	2	3	4	5	6	7	8
1	0	0.298830×10^{-2}	0.597378×10^{-1}	0.110893	0.100298	0.197290×10^{-1}	0.14	0.1745
2		0	0.14527×10^{-1}	0.67703×10^{-1}	0.346632×10^{-1}	0.529034×10^{-1}	0.2×10^{-1}	0.3×10^{-1}
3			0	0.249291×10^{-1}	-0.838212×10^{-2}	0.14719	0.0	0.0
4				0	0.199213×10^{-1}	0.154365	0.0	0.0
5					0	0.154365	0.0	0.0
6						0	0.15	0.18
7								0.0
8								

Appendix D. Computer Program and Parameters for the Revised Klosek and McKinley Model

The program listing and tables that follow are for the Revised Klosek and McKinley model described in section 4. The method may be used in two ways. First using the equation:

$$V_{mix} = \sum X_i V_i - [k_1 + (k_2 - k_1) X_{N_2}/0.0425] X_{CH_4} \qquad (D\text{-}1)$$

The V_i, k_1 and k_2 may be obtained from tables 8, 9 and 10 and the volume of the mixture calculated. For example given the mixture of 0.8130 CH_4 + 0.0475 C_2H_6 + 0.0487 C_3H_8 + 0.0242 nC_4H_{10} + 0.0241 iC_4H_{10} + 0.0425 N_2 and a temperature of 105 kelvin.

The $\sum X_i V_i$ and $X_i W_i$ are obtained from table 8.

$$\sum X_i V_i = (.8130)(.037113) + (.0475)(.047267) + (.0487)(.061766)$$
$$+ (.0242)(.076100) + (.0241)(.077538) + (.0425)(.042565)$$
$$= 0.0409453$$

Then the $\sum X_i W_i$ $= (.8130)(16.04303) + (.0475)(30.07012) + (.0487)(44.09721)$
$+ (.0242)(58.1243) + (.0241)(58.1243) + (.0425)(28.0143)$
$= 20.6168$ the molecular weight of the mixture

from table 9 $k_1 = .697 \times 10^{-3}$

from table 10 $k_2 = .849 \times 10^{-3}$

plugging all this into eq (D-1) gives

$$V_{mix} = .040255$$

$$1/V_{mix} = \rho_{mix} = 24.842 \text{ moles/liter}$$

This compares to the experimental value of 24.850 (Appendix A, mixture No. 63) to within 0.03%.

The same result may be obtained by using the computer program in the following way:

D = FMKM(T,X)

where T is temperature in kelvin and X is a matrix of mole fractions of the components in the following order:

X(1) = mole fraction of methane

X(2) = mole fraction of ethane

X(3) = mole fraction of propane

X(4) = mole fraction of normal butane

X(5) = mole fraction of isobutane

X(6) = mole fraction of nitrogen

X(7) = mole fraction of normal pentane

X(8) = mole fraction of isopentane

for the example: T = 105., X(1) = .8130, X(2) = .0475, X(3) = .0487, X(4) = .0242, X(5) = .0241 and X(6) = .0425.

Other subprograms required:

FUNCTION VIDEAL, lines LNG 709 through 732, Appendix F

FUNCTION SAT, lines LNG 733 through 762, Appendix F

Table 8. Volumes of Saturated Liquid of the Pure Components in Liters/mole.

T, K	CH_4	C_2H_6	C_3H_8	nC_4H_{10}	iC_4H_{10}	N_2	nC_5H_{12}	iC_5H_{12}
90.	.035441	.046081	.060461	.074708	.076084	.037543	.089173	.089243
92.	.035649	.046235	.060632	.074891	.076274	.038081	.089379	.089454
94.	.035861	.046390	.060804	.075075	.076466	.038650	.089586	.089666
96.	.036077	.046547	.060977	.075259	.076659	.039254	.089793	.089878
98.	.036298	.046704	.061151	.075445	.076853	.039897	.090000	.090091
100.	.036524	.046863	.061325	.075631	.077047	.040586	.090208	.090304
102.	.036755	.047023	.061501	.075818	.077243	.041327	.090416	.090518
104.	.036992	.047185	.061677	.076006	.077440	.042128	.090624	.090733
106.	.037234	.047348	.061855	.076194	.077637	.043002	.090833	.090948
108.	.037481	.047512	.062033	.076384	.077836	.043963	.091042	.091163
110.	.037735	.047678	.062212	.076574	.078035	.045031	.091252	.091379
112.	.037995	.047845	.062392	.076765	.078236	.046231	.091462	.091596
114.	.038262	.048014	.062574	.076957	.078438	.047602	.091673	.091814
116.	.038536	.048184	.062756	.077150	.078640	.049179	.091884	.092032
118.	.038817	.048356	.062939	.077344	.078844	.050885	.092095	.092251
120.	.039106	.048529	.063124	.077539	.079049	.052714	.092307	.092470
122.	.039404	.048704	.063309	.077734	.079255	.054679	.092520	.092690
124.	.039710	.048881	.063496	.077931	.079462	.056797	.092733	.092911
126.	.040025	.049059	.063684	.078128	.079671	.059085	.092947	.093133
128.	.040350	.049239	.063873	.078327	.079880	.061565	.093161	.093355
130.	.040685	.049421	.064063	.078526	.080091	.064263	.093376	.093578
molecular weight*	16.04303	30.07012	44.09721	58.1243	58.1243	28.0143	72.15139	72.15139

weight*

Table 9. Correction Factor $k_1 \times 10^3$.

T/W	16	17	18	19	20	21	22	23	24	25
90	-.005	.120	.220	.340	.430	.515	.595	.660	.725	.795
95	-.006	.135	.260	.380	.500	.590	.665	.740	.810	.885
100	-.007	.150	.300	.425	.575	.675	.755	.830	.910	.990
105	-.007	.165	.340	.475	.635	.735	.840	.920	1.045	1.120
110	-.008	.180	.375	.535	.725	.835	.950	1.055	1.155	1.245
115	-.009	.220	.440	.610	.810	.945	1.065	1.180	1.280	1.380
120	-.01	.250	.500	.695	.920	1.055	1.205	1.330	1.450	1.550
125	-.013	.295	.590	.795	1.035	1.210	1.385	1.525	1.640	1.750
130	-.015	.345	.700	.920	1.200	1.370	1.555	1.715	1.860	1.990
135	-.017	.400	.825	1.060	1.390	1.590	1.800	1.950	2.105	2.272

Table 10. Correction Factor $k_2 \times 10^3$.

T/W	16	17	18	19	20	21	22	23	24	25
90	-.004	.10	.22	.35	.50	.60	.69	.78	.86	.95
95	-.005	.12	.28	.43	.59	.71	.83	.94	1.05	1.14
100	-.007	.16	.34	.49	.64	.79	.94	1.08	1.17	1.27
105	-.01	.24	.42	.61	.75	.91	1.05	1.19	1.33	1.45
110	-.015	.32	.59	.77	.92	1.07	1.22	1.37	1.52	1.71
115	-.024	.41	.72	.95	1.15	1.22	1.3	1.45	1.65	2.00
120	-.032	.60	.91	1.23	1.43	1.63	1.85	2.08	2.30	2.45
125	-.043	.71	1.13	1.48	1.73	1.98	2.23	2.48	2.75	2.90
130	-.058	.95	1.46	1.92	2.20	2.42	2.68	3.00	3.32	3.52
135	-.075	1.30	2.00	2.40	2.60	3.00	3.40	3.77	3.99	4.23

Appendix E. Computer Program for the Cell Model

The program listings for the cell model start at line LNG 763 and continue on to the end of Appendix F. As is mentioned in section 4, no details of the model are given here only the program listing. To use the program in its present form one must make the following reference to the computer program:

```
CALL ECKNON(P,D,T,X)
```

where P is input pressure in bars, D is the output density in moles/liter, T is the input temperature in kelvins and X is a matrix of the mole fractions of the components of the mixture in the following order:

X(1) = mole fraction of methane

X(2) = mole fraction of ethane

X(3) = mole fraction of propane

X(4) = mole fraction of normal butane

X(5) = mole fraction of isobutane

X(6) = mole fraction of nitrogen

X(7) = mole fraction of normal pentane

X(8) = mole fraction of isopentane

Appendix F. Computer Programs

Listing of computer programs for all four models. See the sections on the individual models for a list of subprograms needed for each model.

The programs are written in FORTRAN IV and are operational on a CDC 6600 computer.

```
      SUBROUTINE PDMIX(P,D,T,X)                                          LNG   1
C     FOR A CALL TO PDMIX, P,T AND X ARE INPUT.  P IS IN BAR,T IS IN     LNG   2
C     KELVIN AND D IS OUTPUT IN THE UNITS OF MOLES/LITER                 LNG   3
C     FOR A CALL TO P MIX, D,T AND X ARE INPUT AND P IS OUTPUT, THE      LNG   4
C     UNITS ARE THE SAME                                                 LNG   5
C     THE X MATRIX MUST CONTAIN THE MOL FRACTION OF THE ALLOWABLE FLUIDS LNG   6
C     IN THE FOLLOWING ORDER,1=C1,2=C2,3=C3,4=NC4,5=IC4,6=N2,7=NC5,8=IC5 LNG   7
C     PLACE A ZERO IN THE ELEMENTS OF X WHERE THAT PARTICULAR GAS IS NOT LNG   8
C     PRESENT                                                            LNG   9
      DIMENSION ZATA(10,10),ATA(10,10),TC(10),VC(10),ZC(10),AC(10),W(10) LNG  10
     1,PC(10),CF(9)                                                      LNG  11
      DIMENSION THETA(10,10),TH(10,10),PHI(10,10),PH(10,10),F(10,10),FH  LNG  12
     1(10,10),H(10,10),HH(10,10),VR(10,10),TR(10,10),X(10)               LNG  13
      COMMON/DATA M/ZATA,ATA,TC,VC,W,TCO,VCO,ACO,ZCO,RR,R,OMEGO,AC,ZC,N  LNG  14
     1,PC,CF                                                             LNG  15
      DATA(IE=0)                                                         LNG  16
      D=0.0 $ PI=P/1.01325                                               LNG  17
      GO TO 4                                                            LNG  18
      ENTRY PMIX                                                         LNG  19
      P=0.0                                                              LNG  20
      IP=1                                                               LNG  21
      GO TO 5                                                            LNG  22
    4 IP=0                                                               LNG  23
    5 CONTINUE                                                           LNG  24
      IF(IE.GT.0)GO TO 6                                                 LNG  25
      IE=1                                                               LNG  26
      CALL DATA CH4                                                      LNG  27
      IBASE=L=1                                                          LNG  28
      CALL MIX DATA(IBASE)                                               LNG  29
    6 CONTINUE                                                           LNG  30
      DO 1 I=1,N                                                         LNG  31
      F(I,I)=H(I,I)=1.                                                   LNG  32
      THETA(I,I)=1.                                                      LNG  33
    1 PHI(I,I)=1.                                                        LNG  34
      DO 30 J=1,30                                                       LNG  35
      HX=FXHX=0.0                                                        LNG  36
      DO 10 I=1,N                                                        LNG  37
      FH(I,I)=F(I,I)                                                     LNG  38
      HH(I,I)=H(I,I)                                                     LNG  39
      IF(X(I).LT..0001)GO T O 10                                         LNG  40
      F(I,I)=(TC(I)/TC(L))*THETA(I,I)                                    LNG  41
      H(I,I)=(VC(I)/VC(L))*PHI(I,I)                                      LNG  42
   10 CONTINUE                                                           LNG  43
      DO 11 IA=1,N                                                       LNG  44
      DO 11 IB=1,N                                                       LNG  45
      IF(X(IA).LT..0001)GO TO 11                                         LNG  46
      IF(X(IB).LT..0001)GO TO 11                                         LNG  47
      FAB=ZATA(IA,IB)*(F(IA,IA)* F(IB,IB))**.5                           LNG  48
      HAB=ATA(IA,IB)*(.5*H(IA,IA)**(1./3.)+.5*H(IB,IB)**(1./3.))**3      LNG  49
      HX=HX+X(IA)*X(IB)*HAB                                              LNG  50
      FXHX=FXHX+X(IA)*X(IB)*HAB*FAB                                      LNG  51
   11 CONTINUE                                                           LNG  52
      FX=FXHX/HX                                                         LNG  53
      PRO=PI*HX/FX                                                       LNG  54
      TRO=T/FX                                                           LNG  55
      DEN=D*HX                                                           LNG  56
      IF(IP.EQ.1)GO TO 8                                                 LNG  57
      DD=SATL(TRO)*1000.+1.                                              LNG  58
    9 DEN=FIND M(PRO,TRO,DD)                                             LNG  59
    8 CONTINUE                                                           LNG  60
      IF(DEN.LE.0.0) GO TO 33                                            LNG  61
      VRO=1000./DEN                                                      LNG  62
      DO 12 I=1,N                                                        LNG  63
```

```
      IF(X(I).LT..0001)GO TO 12                                          LNG  64
      VR(I,I)=VRO*PHI(I,I)/VCO                                           LNG  65
      TR(I,I)=TRO*THETA(I,I)/TCO                                         LNG  66
      IF(VR(I,I).GT.2.)VR(I,I)=2.                                        LNG  67
      IF(VR(I,I).LT..5)VR(I,I)=.5                                        LNG  68
      TH(I,I)=THETA(I,I)                                                 LNG  69
      THETA(I,I)=1.+(AC(I)-OMEGO)*(CF(1)-CF(2)*ALOG(TR(I,I))+(CF(3)-CF(4 LNG  70
     1)/TR(I,I))*(VR(I,I)-CF(5)))                                        LNG  71
      PH(I,I)=PHI(I,I)                                                   LNG  72
      PHI(I,I)=(1.+(AC(I)-OMEGO)*(CF(6)*(VR(I,I)-CF(7))-CF(8)*(VR(I,I)   LNG  73
     1-CF(9))*ALOG(TR(I,I))))*ZCO/ZC(I)                                  LNG  74
   12 CONTINUE                                                           LNG  75
      DO 13 I=1,N                                                        LNG  76
      IF(X(I).LT..0001)GO TO 13                                          LNG  77
      IF(ABS ((FH(I,I)-F(I,I))/F(I,I)).GT..001)GO TO 30                  LNG  78
      IF(ABS ((HH(I,I)-H(I,I))/H(I,I)).GT..001)GO TO 30                  LNG  79
      IF(ABS ((TH(I,I)-THETA(I,I))/THETA(I,I)).GT..001)GO TO 30          LNG  80
      IF(ABS ((PH(I,I)-PHI(I,I))/PHI(I,I)).GT..001)GO TO 30              LNG  81
   13 CONTINUE                                                           LNG  82
      GO TO 31                                                           LNG  83
   30 CONTINUE                                                           LNG  84
   33 PRINT 100,P,DEN,T                                                  LNG  85
  100 FORMAT(*  ITTERATION FAILED AT*,3F10.4)                            LNG  86
      STOP                                                               LNG  87
   31 D=DEN/HX                                                           LNG  88
      THC=THETA(6,6)                                                     LNG  89
      IF(IP.EQ.0)GO TO 32                                                LNG  90
      CALL PRESS(P,DEN,TRO)                                              LNG  91
      PI=1.01325*P*FX/HX                                                 LNG  92
   32 RETURN                                                             LNG  93
      END                                                                LNG  94
      SUBROUTINE MIX DATA(IBASE)                                         LNG  95
      DIMENSION ZATA(10,10),ATA(10,10),TC(10),VC(10),ZC(10),AC(10),W(10) LNG  96
     1,PC(10),CF(9)                                                      LNG  97
      COMMON/DATA M/ZATA,ATA,TC,VC,W,TCO,VCO,ACO,ZCO,RR,R,OMEGO,AC,ZC,N  LNG  98
     1,PC,CF                                                             LNG  99
      DATA(PC=45.356,47.96757,41.89008,37.79462,36.40758,33.10393)       LNG 100
      DATA(TC=190.555,305.5,370.,425.,408.1,126.2)                       LNG 101
      DATA(W=16.04303,30.07012,44.09721,58.1243,58.1243,28.0134)         LNG 102
      DATA(VC=98.522,146.2,200.,251.62,263.00,89.827)                    LNG 103
      DATA(AC=.0109,.110427,.154837,.176372,.150115,.0291791)            LNG 104
      DATA(R=82.05606),(RR=8.3144),(N=6)                                 LNG 105
      DATA(CF=-.109495,.919454,-4.01525,-4.14192,.444850,.356808,        LNG 106
     11.02619,.893323,.761533)                                           LNG 107
      DATA(ATA(1,2)=1.00514),(ATA(1,3)=1.01922),(ATA(1,4)=1.04243),      LNG 108
     1(ATA(1,5)=1.05048),(ATA(1,6)=1.01009),                             LNG 109
     A(ATA(2,3)=1.00599),(ATA(2,4)=1.01616),                             LNG 110
     2(ATA(2,5)=1.02369),(ATA(2,6)=1.02127),(ATA(3,4)=1.00172),          LNG 111
     3(ATA(3,5)=1.01140),(ATA(3,6)=1.04606),(ATA(4,5)=.997114),          LNG 112
     4(ATA(4,6)=1.20955),(ATA(5,6)=1.13889)                              LNG 113
      DATA(ZATA(1,2)=1.01127),(ZATA(1,3)=.988608),(ZATA(1,4)=.983130),   LNG 114
     1(ZATA(1,5)=.986978),(ZATA(1,6)=.953430),(ZATA(2,3)=.999961),       LNG 115
     2(ZATA(2,4)=.972223),(ZATA(2,5)=.998886),(ZATA(2,6)=.939622),       LNG 116
     3(ZATA(3,4)=.985547),(ZATA(3,5)=1.03099),(ZATA(3,6)=.912209),       LNG 117
     4(ZATA(4,5)=.976416),(ZATA(4,6)= .849200),(ZATA(5,6)=.857310)       LNG 118
      L=IBASE                                                            LNG 119
      N=8                                                                LNG 120
      N1=N-1                                                             LNG 121
      PC(7)=33.37                                                        LNG 122
      PC(8)=31.57                                                        LNG 123
      TC(7)=469.6                                                        LNG 124
      TC(8)=460.39                                                       LNG 125
      VC(7)=304.                                                         LNG 126
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      VC(8)=306.                                                        LNG 127
      W(7)=72.15139                                                     LNG 128
      W(8)=72.15139                                                     LNG 129
      AC(7)=.234320                                                     LNG 130
      AC(8)=.288886                                                     LNG 131
      ATA(1,7)=ATA(1,8)=1.06                                            LNG 132
      ATA(2,7)=ATA(2,8)=1.02                                            LNG 133
      ATA(3,7)=ATA(3,8)=1.01                                            LNG 134
      ATA(4,7)=ATA(4,8)=1.                                              LNG 135
      ATA(5,7)=ATA(5,8)=1.                                              LNG 136
      ATA(6,7)=ATA(6,8)=1.                                              LNG 137
      ATA(7,8)=1.                                                       LNG 138
      ZATA(1,7)=ZATA(1,8)=.98                                           LNG 139
      ZATA(2,7)=ZATA(2,8)=.99                                           LNG 140
      ZATA(3,7)=ZATA(3,8)=.99                                           LNG 141
      ZATA(4,7)=ZATA(4,8)=.99                                           LNG 142
      ZATA(5,7)=ZATA(5,8)=.99                                           LNG 143
      ZATA(6,7)=ZATA(6,8)=.99                                           LNG 144
      ZATA(7,8)=1.                                                      LNG 145
      DO 3 J=1,N1                                                       LNG 146
      J1=J+1                                                            LNG 147
      DO 3 K=J1,N                                                       LNG 148
      ZATA(K,J)=ZATA(J,K)                                               LNG 149
    3 ATA(K,J)=ATA(J,K)                                                 LNG 150
      DO 4 I=1,N                                                        LNG 151
    4 ATA(I,I)=ZATA(I,I)=1.                                             LNG 152
      PCO=PC(L)                                                         LNG 153
      VCO=VC(L)                                                         LNG 154
      TCO=TC(L)                                                         LNG 155
      OMEGO=AC(L)                                                       LNG 156
      ZCO=PCO*VCO/TCO/R                                                 LNG 157
      DO 5 I=1,N                                                        LNG 158
    5 ZC(I)=PC(I)*VC(I)/TC(I)/R                                         LNG 159
      RETURN                                                            LNG 160
      END                                                               LNG 161
      SUBROUTINE DATA CH4                                               LNG 162
C     INITIALIZES THE EQUATION OF STATE CONSTANTS TO METHANE            LNG 163
      DIMENSION G(32),VP(9),GI(11)                                      LNG 164
      COMMON/DATA/G,R,GAMMA,VP,DTP                                      LNG 165
      DIMENSION A(10)                                                   LNG 166
      COMMON/SATC/A                                                     LNG 167
      R=.08205616                                                       LNG 168
      GAMMA=-.0096                                                      LNG 169
      A(1)=190.555                                                      LNG 170
      A(2)=10.23                                                        LNG 171
      A(3)=18.404156472                                                 LNG 172
      A(4)=7.3498921512                                                 LNG 173
      A(5)=-1.4313160833                                                LNG 174
      A(6)=A(7)=0.0                                                     LNG 175
      G( 1)=-.187027997685E-01                                          LNG 176
      G( 2)= .103387108009E+01                                          LNG 177
      G( 3)=-.155387625619E+02                                          LNG 178
      G( 4)= .772311478564E+03                                          LNG 179
      G( 5)=-.377103300895E+05                                          LNG 180
      G( 6)= .846818843475E-03                                          LNG 181
      G( 7)=-.496415884529E+00                                          LNG 182
      G( 8)= .869909352414E+02                                          LNG 183
      G( 9)=-.322821592493E+05                                          LNG 184
      G(10)=-.395843026318E-04                                          LNG 185
      G(11)= .266772318035E-01                                          LNG 186
      G(12)=-.304010057839E+01                                          LNG 187
      G(13)= .191584507536E-03                                          LNG 188
      G(14)=-.195587933458E-03                                          LNG 189
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      G(15)= .607479967879E+01                                          LNG 190
      G(16)=-.529609525984E-03                                          LNG 191
      G(17)= .152264286004E-04                                          LNG 192
      G(18)=-.109952182842E-01                                          LNG 193
      G(19)= .191395549929E-03                                          LNG 194
      G(20)= .386470003746E+05                                          LNG 195
      G(21)=-.157930582612E+07                                          LNG 196
      G(22)= .195270144401E+03                                          LNG 197
      G(23)= .165996081629E+07                                          LNG 198
      G(24)= .603051146711E+00                                          LNG 199
      G(25)= .376485162808E+02                                          LNG 200
      G(26)= .125593680622E-02                                          LNG 201
      G(27)=-.343570032513E+02                                          LNG 202
      G(28)=-.540945094139E-05                                          LNG 203
      G(29)= .185622284663E-02                                          LNG 204
      G(30)= .770786979245E-08                                          LNG 205
      G(31)=-.286868318650E-05                                          LNG 206
      G(32)= .372376961647E-04                                          LNG 207
      VP(1)=4.77748580                                                  LNG 208
      VP(2)=1.76065363                                                  LNG 209
      VP(3)=-.56788894                                                  LNG 210
      VP(4)=1.32786231                                                  LNG 211
      VP(5)=1.5                                                         LNG 212
      VP(6)=.1159                                                       LNG 213
      VP(7)=90.68                                                       LNG 214
      VP(8)=190.555                                                     LNG 215
      END                                                               LNG 216
      FUNCTION FIND M(P,T,D)                                            LNG 217
C     SOLVES THE EQUATION OF STATE OF METHANE FOR DENSITY GIVEN P AND T  LNG 218
      DD=D                                                              LNG 219
      TT=T                                                              LNG 220
      DO 10 I=1,50                                                      LNG 221
      CALL PRESS(PP,DD,TT)                                              LNG 222
      P2=PP                                                             LNG 223
      IF(ABS (P-P2)-1.E-7*P)20,20,1                                     LNG 224
    1 CALL DPDD(PP,DD,TT)                                               LNG 225
      DP=PP                                                             LNG 226
      CORR=(P2-P)/DP                                                    LNG 227
      IF(ABS (CORR)-1.E-7*DD)20,20,10                                   LNG 228
   10 DD=DD-CORR                                                        LNG 229
      FIND M=0                                                          LNG 230
      RETURN                                                            LNG 231
   20 FIND M=DD                                                         LNG 232
      RETURN                                                            LNG 233
      END                                                               LNG 234
      FUNCTION SATL(T)                                                  LNG 235
C     CALCULATES THE SATURATED LIQUID DENSITY OF METHANE                LNG 236
      DIMENSION A(10)                                                   LNG 237
      COMMON/SATC/A                                                     LNG 238
      IF(T.GT.A(1))GO TO 1                                              LNG 239
      X=(1.-T/A(1))                                                     LNG 240
      SATL=A(2)+A(3)*X**(.35)+A(4)*X+A(5)*X**(4./3.)+A(6)*X**(5./3.     LNG 241
     1)+A(7)*X**2                                                       LNG 242
      SATL=SATL/1000.                                                   LNG 243
      RETURN                                                            LNG 244
    1 SATL=1.E20                                                        LNG 245
      RETURN                                                            LNG 246
      END                                                               LNG 247
      SUBROUTINE PROPS(PP,DD,TT)                                        LNG 248
C     EQUATION OF STATE FOR METHANE AND NITROGEN                        LNG 249
      DIMENSION X(33)                                                   LNG 250
      DIMENSION B(33),G(32)                                             LNG 251
      EQUIVALENCE (B,X)                                                 LNG 252
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      COMMON/DATA/G,R,GAMMA                                    LNG 253
      DATA(ID=1)                                               LNG 254
      DATA(IZ=1)                                               LNG 255
    1 CONTINUE                                                 LNG 256
      D=DD                                                     LNG 257
      P=PP                                                     LNG 258
      T=TT                                                     LNG 259
      GM=GAMMA                                                 LNG 260
      D2=D*D                                                   LNG 261
      D3=D2*D                                                  LNG 262
      D4=D3*D                                                  LNG 263
      D5=D4*D                                                  LNG 264
      D6=D5*D                                                  LNG 265
      D7=D6*D                                                  LNG 266
      D8=D7*D                                                  LNG 267
      D9=D8*D                                                  LNG 268
      D10=D9*D                                                 LNG 269
      D11=D10*D                                                LNG 270
      D12=D11*D                                                LNG 271
      D13=D12*D                                                LNG 272
      TS=SQRT (T)                                              LNG 273
      T2=T*T                                                   LNG 274
      T3=T2*T                                                  LNG 275
      T4=T3*T                                                  LNG 276
      T5=T4*T                                                  LNG 277
      F=EXP (GM*D2)                                            LNG 278
      GO TO (100,200),K                                        LNG 279
      ENTRY PRESS                                              LNG 280
      K=1                                                      LNG 281
      GO TO 1                                                  LNG 282
  100 CONTINUE                                                 LNG 283
      B( 1)=D2*T                                               LNG 284
      B( 2)=D2*TS                                              LNG 285
      B( 3)=D2                                                 LNG 286
      B( 4)=D2/T                                               LNG 287
      B( 5)=D2/T2                                              LNG 288
      B( 6)=D3*T                                               LNG 289
      B( 7)=D3                                                 LNG 290
      B( 8)=D3/T                                               LNG 291
      B( 9)=D3/T2                                              LNG 292
      B(10)=D4*T                                               LNG 293
      B(11)=D4                                                 LNG 294
      B(12)=D4/T                                               LNG 295
      B(13)=D5                                                 LNG 296
      B(14)=D6/T                                               LNG 297
      B(15)=D6/T2                                              LNG 298
      B(16)=D7/T                                               LNG 299
      B(17)=D8/T                                               LNG 300
      B(18)=D8/T2                                              LNG 301
      B(19)=D9/T2                                              LNG 302
      B(20)=D3*F/T2                                            LNG 303
      B(21)=D3*F/T3                                            LNG 304
      B(22)=D5*F/T2                                            LNG 305
      B(23)=D5*F/T4                                            LNG 306
      B(24)=D7*F/T2                                            LNG 307
      B(25)=D7*F/T3                                            LNG 308
      B(26)=D9*F/T2                                            LNG 309
      B(27)=D9*F/T4                                            LNG 310
      B(28)=D11*F/T2                                           LNG 311
      B(29)=D11*F/T3                                           LNG 312
      B(30)=D13*F/T2                                           LNG 313
      B(31)=D13*F/T3                                           LNG 314
      B(32)=D13*F/T4                                           LNG 315
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  102 P=0                                                              LNG 316
      DO 101 I=1,32                                                    LNG 317
  101 P=P+B(I)*G(I)                                                    LNG 318
      P=P+R*D*T                                                        LNG 319
      PP=P                                                             LNG 320
      RETURN                                                           LNG 321
      ENTRY DPDD                                                       LNG 322
      K=2                                                              LNG 323
      GO TO 1                                                          LNG 324
  200 CONTINUE                                                         LNG 325
      F1=2.00*F*GM*D                                                   LNG 326
      F21=3.000*F*D2 +F1*D3                                            LNG 327
      F22=5.000*F*D4 +F1*D5                                            LNG 328
      F23=7.000*F*D6 +F1*D7                                            LNG 329
      F24=9.000*F*D8 +F1*D9                                            LNG 330
      F25=11.00*F*D10+F1*D11                                           LNG 331
      F26=13.00*F*D12+F1*D13                                           LNG 332
      B( 1)=2.00*D*T                                                   LNG 333
      B( 2)=2.00*D*TS                                                  LNG 334
      B( 3)=2.00*D                                                     LNG 335
      B( 4)=2.00*D/T                                                   LNG 336
      B( 5)=2.00*D/T2                                                  LNG 337
      B( 6)=3.00*D2*T                                                  LNG 338
      B( 7)=3.00*D2                                                    LNG 339
      B( 8)=3.00*D2/T                                                  LNG 340
      B( 9)=3.00*D2/T2                                                 LNG 341
      B(10)=4.00*D3*T                                                  LNG 342
      B(11)=4.00*D3                                                    LNG 343
      B(12)=4.00*D3/T                                                  LNG 344
      B(13)=5.00*D4                                                    LNG 345
      B(14)=6.00*D5/T                                                  LNG 346
      B(15)=6.00*D5/T2                                                 LNG 347
      B(16)=7.00*D6/T                                                  LNG 348
      B(17)=8.00*D7/T                                                  LNG 349
      B(18)=8.00*D7/T2                                                 LNG 350
      B(19)=9.00*D8/T2                                                 LNG 351
      B(20)=F21/T2                                                     LNG 352
      B(21)=F21/T3                                                     LNG 353
      B(22)=F22/T2                                                     LNG 354
      B(23)=F22/T4                                                     LNG 355
      B(24)=F23/T2                                                     LNG 356
      B(25)=F23/T3                                                     LNG 357
      B(26)=F24/T2                                                     LNG 358
      B(27)=F24/T4                                                     LNG 359
      B(28)=F25/T2                                                     LNG 360
      B(29)=F25/T3                                                     LNG 361
      B(30)=F26/T2                                                     LNG 362
      B(31)=F26/T3                                                     LNG 363
      B(32)=F26/T4                                                     LNG 364
  202 P=0                                                              LNG 365
      DO 201 I=1,32                                                    LNG 366
  201 P=P+B(I)*G(I)                                                    LNG 367
      P=P+R*T                                                          LNG 368
      PP=P                                                             LNG 369
      RETURN                                                           LNG 370
      END                                                              LNG 371
      SUBROUTINE DATA N2                                               LNG 372
C     INITIALIZES THE EQUATION OF STATE CONSTANTS TO NITROGEN          LNG 373
      DIMENSION G(32),VP(9),GI(11)                                     LNG 374
      COMMON/DATA/G,R,GAMMA,VP,DTP                                     LNG 375
      R=8.20539E-2                                                     LNG 376
      GAMMA=-.0056                                                     LNG 377
      G( 1)=    0.136224769272827E-02                                  LNG 378
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      G( 2)=    0.107032469908591E 00                                    LNG 379
      G( 3)=   -0.243900721871413E 01                                    LNG 380
      G( 4)=    0.341007449376470E 02                                    LNG 381
      G( 5)=   -0.422374309466167E 04                                    LNG 382
      G( 6)=    0.105098600246494E-03                                    LNG 383
      G( 7)=   -0.112594826522081E-01                                    LNG 384
      G( 8)=    0.142600789270907E-03                                    LNG 385
      G( 9)=    0.184698501609007E 05                                    LNG 386
      G(10)=    0.811140082588776E-07                                    LNG 387
      G(11)=    0.233011645038006E-02                                    LNG 388
      G(12)=   -0.507752586350986E 00                                    LNG 389
      G(13)=    0.485027881931214E-04                                    LNG 390
      G(14)=   -0.113656764115364E-02                                    LNG 391
      G(15)=   -0.707430273540575E 00                                    LNG 392
      G(16)=    0.751706648852680E-04                                    LNG 393
      G(17)=   -0.111614119537424E-05                                    LNG 394
      G(18)=    0.368796562233495E-03                                    LNG 395
      G(19)=   -0.201317691347729E-05                                    LNG 396
      G(20)=   -0.169717444755949E 05                                    LNG 397
      G(21)=   -0.119719240044192E 06                                    LNG 398
      G(22)=   -0.975218272038281E 02                                    LNG 399
      G(23)=    0.554639713151823E 05                                    LNG 400
      G(24)=   -0.179920450443470E 00                                    LNG 401
      G(25)=   -0.256582926077184E 01                                    LNG 402
      G(26)=   -0.413707715090789E-03                                    LNG 403
      G(27)=   -0.256245415300293E 00                                    LNG 404
      G(28)=   -0.124222373740063E-06                                    LNG 405
      G(29)=    0.103556535840165E-04                                    LNG 406
      G(30)=   -0.538699166558303E-09                                    LNG 407
      G(31)=   -0.757415412839596E-08                                    LNG 408
      G(32)=    0.585367172069521E-07                                    LNG 409
      VP(1)=5.1113192094 $ VP(2)=6.482667539E-1                          LNG 410
      VP(3)=-1.5108730916E-1 $ VP(4)=7.4028493342E-1                     LNG 411
      VP(5)=1.5 $ VP(6)=.123 $ VP(7)=63.15 $ VP(8)=126.26                LNG 412
      VP(9)=0.0                                                          LNG 413
      DTP=31.0                                                           LNG 414
      RETURN  $  END                                                     LNG 415
      FUNCTION VPN(TT)                                                   LNG 416
C     CALCULATES THE VAPOR PRESSURE OF BOTH METHANE AND NITROGEN         LNG 417
      DIMENSION G(32),VP(9)                                              LNG 418
      COMMON/DATA/G,R,GAMMA,VP,DTP                                       LNG 419
      T=TT                                                               LNG 420
      X=(1.-VP(7)/T)/(1.-VP(7)/VP(8))                                    LNG 421
      VPN=VP(6)*EXP(VP(1)*X+VP(2)*X*X+VP(3)*X**3+VP(9)*X**4+VP(4)*X*     LNG 422
     1(1.-X)**VP(5))                                                     LNG 423
      RETURN                                                             LNG 424
      END                                                                LNG 425
      FUNCTION RODEN(P,T,X)                                              LNG 426
C     THE HARD SPHERE MODEL,SEE SUBROUTINE FM FOR THE ARGUMENT LIST      LNG 427
      DIMENSION X(10)                                                    LNG 428
      CALL FM(P,T,X,V,G)                                                 LNG 429
      D=28.                                                              LNG 430
      V=V+V IDEL(P,D,T,X)                                                LNG 431
      RODEN=1000./V                                                      LNG 432
      RETURN                                                             LNG 433
      END                                                                LNG 434
      SUBROUTINE FM(Q,T,X,V9,G9)                                         LNG 435
C     PREDICTION OF EXCESS PROPERTIES WITH LHW POTENTIAL, COMP 1-METH    LNG 436
C     ANE,COMP 2-ETHANE,COMP 3-PROPANE,COMP 4 N-BUTANE,COMP 5-I-BUT      LNG 437
C     ANE, COMP 6-NITROGEN, COMP 7-NORMAL PENTANE,COMP 8-ISOPENTANE      LNG 438
C     ADV CRYO ENGR. VOL. 19 (1973)-REPROGRAMED BY R. MCCARTY,2/22/74    LNG 439
C     ARGUMENTS ARE X-MOLE FRACTIONS,T-TEMPERATURE,Q-PRESSURE,V9-EXCESS  LNG 440
C     VOLUME,G9-EXCESS GIBBS ENERGY, FIRST THREE ARE INPUT,LAST TWO ARE  LNG 441
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C     OUTPUT,INPUT IS IN KELVINS AND BAR,                                    LNG 442
      DIMENSION A(8),B(8)      ,S(8),V(8),G(8),C(8,8),D(8,8),K(8,8),         LNG 443
     1J(8,8),E(8),Y(8),O(8),X(10)                                            LNG 444
      TYPE REAL J,K,N1                                                       LNG 445
      DATA(KEY=1)                                                            LNG 446
      DATA(J(1,2)=-.00388616),(J(1,3)=-.0120932),(J(1,4)=-.0231577),         LNG 447
     A(J(1,5)=-.0238349),(J(1,6)=-.00997547),(J(1,7)=-.0326),(J(1,8)=        LNG 448
     B-.0458),(J(2,4)=-.00400910),(J(2,5)=-.00812712),(J(2,6)=-.0143976)     LNG 449
     C,(J(2,7)=-.003),(J(2,8)=-.004),(J(3,4)=+.0007615710),(J(3,5)=-.003     LNG 450
     D83743),(J(3,6)=-.024014),(J(3,7)=0.0),(J(3,8)=0.0),(J(4,5)=.002221     LNG 451
     E50),(J(4,7)=.0),(J(4,8)=.0),(J(5,6)=-.0576043),(J(4,6)=-.0576043),     LNG 452
     F(J(5,7)=.0),(J(5,8)=0.0),(J(6,7)=-.04),(J(6,8)=-.05),(J(7,8)=.0)       LNG 453
     G ,(J(2,3)=-.002162)                                                    LNG 454
      DATA(K(1,2)=.00298830),(K(1,3)=.0597378),(K(1,4)=.110893),(            LNG 455
     AK(1,5)=.100298),(K(1,6)=.0197290),(K(1,7)=.14),(K(1,8)=.1745),         LNG 456
     B(K(2,4)=.0677703),(K(2,5)=.0346632),(K(2,6)=.0529034),(K(2,7)=         LNG 457
     C.02),(K(2,8)=.03),(K(3,4)=.0249291),(K(3,5)=-.00838212),(K(4,5)=.0     LNG 458
     D199213),(K(4,6)=.154365 ),(K(4,7)=.0),(K(4,8)=.0),(K(5,6)=.154365)     LNG 459
     E,(K(5,7)=.0),(K(5,8)=.0),(K(6,7)=.15),(K(6,8)=.18),(K(7,8)=0.0)        LNG 460
     F,(K(2,3)=.014527),(K(3,6)=.14719),(K(3,7)=0.0),(K(3,8)=0.0)            LNG 461
      DATA(S=3.676E-8,4.158E-8,4.644E-8,5.051E-8,5.056E-8,3.546E-8,          LNG 462
     15.389E-8,5.706E-8)                                                     LNG 463
      DATA(A=2.755E+5,7.773E+5,14.165E+5,22.733E+5,21.279E+5,1.718E+5,       LNG 464
     130.550E+5,42.946E+5)                                                   LNG 465
      DATA(O=1.,1.5,1.67,1.83,1.79,1.03,1.91,2.11)                           LNG 466
C     THESE ARE THE ACENTRICITY FACTORS (FOR MOLECULAR SHAPES,ETC.)  ***     LNG 467
      DATA(Y=35.,45.,60.,75.,75.,40.,90.,105.)                               LNG 468
      DATA(P1=3.14159),(N1=6.025E+23),(R=8.3143  )                           LNG 469
      IF(KEY.EQ.0)GO TO 1                                                    LNG 470
      KEY=0                                                                  LNG 471
      DO 2 I=1,8                                                             LNG 472
      J(I,I)=0.0                                                             LNG 473
      K(I,I)=0.0                                                             LNG 474
      DO 2 M=I,8                                                             LNG 475
      J(M,I)=J(I,M)                                                          LNG 476
    2 K(M,I)=K(I,M)                                                          LNG 477
    1 CONTINUE                                                               LNG 478
      P=Q*.1                                                                 LNG 479
      IW=8                                                                   LNG 480
      DO 10 I=1,IW                                                           LNG 481
   10 E(I)=O(I)                                                              LNG 482
      DO 15 I=1,IW                                                           LNG 483
   15 B(I)=(2./3.)*P1*N1*S(I)**3                                             LNG 484
      DO 20 I=1,IW                                                           LNG 485
      DO 20 M=1,IW                                                           LNG 486
      D(I,M) =(((B(I)**( 1./3. ) + B(M) ** ( 1./3.))/ 2. )*(1.-J(I,M) ))     LNG 487
     1 **3                                                                   LNG 488
   20 C( I , M ) = ( 1. - K( I, M ) ) * ( A(I)* A(M) ) ** ( 1. / 2. ) *      LNG 489
     1( D( I, M ) **2 / ( B(I) * B(M) ) ) ** ( 1./2. )                       LNG 490
      A2=0  $  B2=0                                                          LNG 491
      E2=0  $  V2=0                                                          LNG 492
      DO 25 I=1,IW                                                           LNG 493
      V2=V2+X(I)*Y(I)                                                        LNG 494
      DO 25 M=1,IW                                                           LNG 495
      E2=E2+X(I)*X(M)*(E(I)+E(M))/2.                                         LNG 496
      A2 = A2   + X(I) * X(M) * C( I, M )                                    LNG 497
   25 B2=B2+X(I)*X(M)*D(I,M)                                                 LNG 498
      V6=V2                                                                  LNG 499
      DO 30 I = 1, IW                                                        LNG 500
      A3 = A(I)                                                              LNG 501
      B3 = B(I)                                                              LNG 502
      E3 = E(I)                                                              LNG 503
      V2 = Y(I)                                                              LNG 504
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      V1 = FIND V1( A3, B3, E3, R, V2, P, T)                                LNG 505
      G1 = FIND G1( A3, B3, E3, R, V2, P, T)                                LNG 506
      V(I) = V1                                                             LNG 507
   30 G(I) = G1                                                             LNG 508
      A3=A2                                                                 LNG 509
      B3=B2                                                                 LNG 510
      E3=E2                                                                 LNG 511
      V2=V6                                                                 LNG 512
      V1=FIND V1(A3,B3,E3,R,V2,P,T)                                         LNG 513
C     G1=FIND G1(A3,B3,E3,R,V2,P,T)                                         LNG 514
      V7=V1                                                                 LNG 515
      G7=G1                                                                 LNG 516
C     EXCEAS VOLUME AND GIBS ENERGY                                         LNG 517
      V9=0                                                                  LNG 518
      G9=0                                                                  LNG 519
      DO 35 I=1,IW                                                          LNG 520
      V9=V9-X(I)*V(I)                                                       LNG 521
   35 G9=G9-X(I)*G(I)                                                       LNG 522
      W9 = -V9                                                              LNG 523
      H9 = -G9                                                              LNG 524
      V9=V9+V7                                                              LNG 525
      G9=G9+G7                                                              LNG 526
      VIWW=V(IWW)                                                           LNG 527
      RETURN                                                                LNG 528
      END                                                                   LNG 529
      FUNCTION FIND V1( A3, B3, E3,R, V2, P, T )                            LNG 530
C     SOLVES THE HARD SPHERE EQUATION OF STATE FOR VOLUME GIVEN P AND T     LNG 531
C     A2 IS THE CONSTANT A, B2 IS THE CONSTANT B, E3 IS THE ACENTRICITY     LNG 532
      INDEX = 0                                                             LNG 533
    1 V1 = V2                                                               LNG 534
      X1 = B3 / ( 4. * V1 )                                                 LNG 535
      F2 = (( 1. + X1 + X1**2) / ( 1. - X1)**3) * E3 -A3 / ( V1 * R * T)    LNG 536
     1- (P * V1) / ( R*T)                                                   LNG 537
      F3 = A3 / (R*T*V1**2) - P/ (R*T)                                      LNG 538
      F3 = F3 - ((( X1 + 2.* X1**2) * ( 1 - X1) + 3* ( 1. + X1 + X1**2)     LNG 539
     1* X1) / (( 1. - X1 )**4 * V1)) * E3                                   LNG 540
      V2 = V1 - F2/ F3                                                      LNG 541
      IF( ABS( (V2 - V1) / V2) .LT. .00001 )   GO TO 2                      LNG 542
      INDEX = INDEX + 1                                                     LNG 543
      IF( INDEX .LT. 250 ) GO TO 1                                          LNG 544
    2 V1 = V2                                                               LNG 545
      FIND V1 = V2                                                          LNG 546
      RETURN                                                                LNG 547
      END                                                                   LNG 548
      FUNCTION FIND G1( A3, B3, E3,R, V2, P, T )                            LNG 549
C     CALCULATES THE GIBBS FREE ENERGY FOR THE HARD SPHERE EOS              LNG 550
      V1 = V2                                                               LNG 551
      X1 = B3 / ( 4. * V1 )                                                 LNG 552
      G1 =ALOG( 1. / ( 1. - X1 )) +(3. *X1) / ( 1. - X1) + ( 3. * X1**2)    LNG 553
     1 / (2. * ( 1. - X1 )**2)                                              LNG 554
      G1 = G1 - A3 / ( E3 * R * T*V1 ) + ( P * V1) / ( E3 * R * T ) -1.0    LNG 555
     1 - ALOG( V1 )                                                         LNG 556
      G1 = R * T * E3 * G1                                                  LNG 557
      FIND G1 = G1                                                          LNG 558
      RETURN                                                                LNG 559
      END                                                                   LNG 560
      FUNCTION EXCESS(P,DD,T,X)                                             LNG 561
C     CALCULATES THE EXCESS OR IDEAL VOLUM DEPENDING ON THE ENTRY           LNG 562
      DIMENSION X(10),F(10)                                                 LNG 563
      KR=0                                                                  LNG 564
      GO TO 1                                                               LNG 565
      ENTRY V IDEL                                                          LNG 566
      KR=1                                                                  LNG 567
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    1 CONTINUE                                                          LNG 568
      CALL ZERO(F)                                                      LNG 569
      IF(X(1).LE..000001)GO TO 2                                        LNG 570
      CALL DATA CH4                                                     LNG 571
      IF(T.GT.190.555)GO TO 12                                          LNG 572
      PM=VPN(T)+.00001                                                  LNG 573
      DELP=P-PM                                                         LNG 574
      D=SAT(T,1)                                                        LNG 575
      CALL DPDD(DP,D,T)                                                 LNG 576
      DELD=DELP/DP                                                      LNG 577
      D=DELD+D                                                          LNG 578
      F(1)=X(1)*1000./D                                                 LNG 579
      GO TO 2                                                           LNG 580
   12 D=FIND M(P,T,DD)                                                  LNG 581
      IF(D.LE.0.0)D=1000.                                               LNG 582
      F(1)=X(1)*1000./D                                                 LNG 583
    2 IF(X(2).LE. .000001)GO TO 3                                       LNG 584
      F(2)=X(2)*1000./SAT(T,2)                                          LNG 585
    3 IF(X(3).LE..000001)GO TO 4                                        LNG 586
      F(3)=X(3)*1000./SAT(T,3)                                          LNG 587
    4 IF(X(4).LE..00001)GO TO 5                                         LNG 588
      F(4)=X(4)*1000./SAT(T,4)                                          LNG 589
    5 IF(X(5).LE..00001)GO TO 6                                         LNG 590
      F(5)=X(5)*1000./SAT(T,5)                                          LNG 591
    6 IF(X(7).LE..00001)GO TO 61                                        LNG 592
      F(7)=X(7)*1000./SAT(T,7)                                          LNG 593
   61 IF(X(8).LE..00001)GO TO 62                                        LNG 594
      F(8)=X(8)*1000./SAT(T,8)                                          LNG 595
   62 IF(X(6).LE..00001)GO TO 8                                         LNG 596
      CALL DATA N2                                                      LNG 597
      IF(T.GT.126.6)GO TO 7                                             LNG 598
      PN=VPN(T)+.000001                                                 LNG 599
      DELP=P-PN                                                         LNG 600
      D=SAT(T,6)                                                        LNG 601
      CALL DPDD(DP,D,T)                                                 LNG 602
      F(6)=X(6)*1000./(D+DELP/DP)                                       LNG 603
      GO TO 8                                                           LNG 604
    7 D=FIND M(P,T,DD)                                                  LNG 605
      IF(D.LE.0.0)D=1000.                                               LNG 606
      F(6)=X(6)*1000./D                                                 LNG 607
    8 V=1000./DD                                                        LNG 608
      VS=0                                                              LNG 609
      DO 21 I=1,8                                                       LNG 610
   21 VS=VS+F(I)                                                        LNG 611
      EXCESS=V-VS                                                       LNG 612
      IF(KR.GT.0)EXCESS=VS                                              LNG 613
      CALL DATA CH4                                                     LNG 614
      RETURN                                                            LNG 615
      END                                                               LNG 616
      SUBROUTINE ZERO(X)                                                LNG 617
C     INITIALIZES THE COMPONENT MATRIX TO 0                             LNG 618
      DIMENSION X(10)                                                   LNG 619
      DO 1 I=1,10                                                       LNG 620
    1 X(I)=0.0                                                          LNG 621
      RETURN                                                            LNG 622
      END                                                               LNG 623
      FUNCTION FMKM(T,X)                                                LNG 624
C     THE REVISED KLOSEK AND MCKINLEY METHOD,THE INPUT IS TEMPERATURE   LNG 625
C     AND THE COMPONENT MATRIX.  TEMPERATURE IS IN KELVIN,OUTPUT IS     LNG 626
C     DENSITY IN MOLES PER LITER.  THE ALLOWABLE COMPONENTS ARE C1,C2,C3 LNG 627
C     NC4,IC4,N2,NC5,IC5 IN THAT ORDER.  THIS METHOD SHOULD NOT BE USED LNG 628
C     FOR MIXTURES WITH LESS THAN 60% METHANE, OR FOR MIXTURES CONTAININ LNG 629
C     MORE THAN 4% NITROGEN OR MORE THAN 4% EACH OF NC4 OR IC4 OR MORE  LNG 630
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C     THAN 2% TOTAL OF NC5 AND IC5.                                      LNG 631
      DIMENSION TM(100),TN(100),X(10),Q(8)                               LNG 632
      DATA(Q=16.04303,30.07012,44.09721,58.1243,58.1243,28.0134,72.1513  LNG 633
     19,72.15139)                                                        LNG 634
      DATA((TM(I),I=1,10)=-.005,.12,.22,.34,.43,.515,.595,.66,.725,.795) LNG 635
      DATA((TM(I),I=11,20)=-.006,.135,.26,.38,.5,.59,.665,.74,.81,.885)  LNG 636
      DATA((TM(I),I=21,30)=-.007,.15,.3,.425,.575,.675,.755,.83,.91,.99) LNG 637
      DATA((TM(I),I=31,40)=-.007,.165,.34,.475,.635,.735,.84,.92,1.045,  LNG 638
     A1.12)                                                              LNG 639
      DATA((TM(I),I=41,50)=-.008,.19,.375,.535,.725,.835,.95,1.055,1.155 LNG 640
     A,1.245)                                                            LNG 641
      DATA((TM(I),I=51,60)=-.009,.22,.44,.61,.81,.945,1.065,1.18,1.28,   LNG 642
     A1.38)                                                              LNG 643
      DATA((TM(I),I=61,70)=-.01,.25,.5,.695,.92,1.055,1.205,1.33,1.45,   LNG 644
     A1.55)                                                              LNG 645
      DATA((TM(I),I=71,80)=-.013,.295,.59,.795,1.035,1.21,1.385,1.525,   LNG 646
     11.64,1.75)                                                         LNG 647
      DATA((TM(I),I=81,90)=-.015,.345,.7,.92,1.2,1.37,1.555,1.715,       LNG 648
     A1.86,1.99)                                                         LNG 649
      DATA((TM(I),I=91,100)=-.017,.4,.825,1.06,1.39,1.59,1.8,1.95,       LNG 650
     A2.105,2.272)                                                       LNG 651
      DATA((TN(I),I=1,10)=-.004,.1,.22,.35,.5,.6,.69,.78,.86,.95)        LNG 652
      DATA((TN(I),I=11,20)=-.005,.12,.28,.43,.59,.71,.83,.94,1.05,1.14)  LNG 653
      DATA((TN(I),I=21,30)=-.007,.16,.34,.49,.64,.79,.94,1.08,1.17,1.27) LNG 654
      DATA((TN(I),I=31,40)=-.01,.24,.42,.61,.75,.91,1.05,1.19,1.33,1.45) LNG 655
      DATA((TN(I),I=41,50)=-.015,.32,.59,.77,.92,1.07,1.22,1.37,1.52,    LNG 656
     11.71)                                                              LNG 657
      DATA((TN(I),I=51,60)=-.024,.41,.72,.95,1.15,1.22,1.3,1.45,1.65,2.) LNG 658
      DATA((TN(I),I=61,70)=-.032,.6,.91,1.23,1.43,1.63,1.85,2.08,2.3,    LNG 659
     12.45)                                                              LNG 660
      DATA((TN(I),I=71,80)=-.043,.71,1.13,1.48,1.73,1.98,2.23,2.48,2.75  LNG 661
     1,2.9)                                                              LNG 662
      DATA((TN(I),I=81,90)=-.058,.95,1.46,1.92,2.2,2.42,2.68,3.,         LNG 663
     A3.32,3.52)                                                         LNG 664
      DATA((TN(I),I=91,100)=-.075,1.3,2.,2.4,2.6,3.,3.4,3.77,            LNG 665
     A3.99,4.23)                                                         LNG 666
      IF(X(1).LT..00001)GO TO 20                                         LNG 667
      AW=0.0                                                             LNG 668
      DO 1 I=1,8                                                         LNG 669
    1 AW=AW+X(I)*Q(I)                                                    LNG 670
      VI=VIDEAL(T,X)                                                     LNG 671
      J=1                                                                LNG 672
      IF(T.GE.95.)J=11                                                   LNG 673
      IF(T.GE.100.)J=21                                                  LNG 674
      IF(T.GE.105.)J=31                                                  LNG 675
      IF(T.GE.110.)J=41                                                  LNG 676
      IF(T.GE.115.)J=51                                                  LNG 677
      IF(T.GE.120.)J=61                                                  LNG 678
      IF(T.GE.125.)J=71                                                  LNG 679
      IF(T.GE.130.)J=81                                                  LNG 680
      JJ=J+9                                                             LNG 681
      W=15.                                                              LNG 682
      DO 5 I=J,JJ                                                        LNG 683
      W=W+1.                                                             LNG 684
      IF(AW.GT.W)GO TO 5                                                 LNG 685
      GO TO 6                                                            LNG 686
    5 CONTINUE                                                           LNG 687
      I=JJ                                                               LNG 688
    6 DIF1=AW-W                                                          LNG 689
      J=I-1                                                              LNG 690
      FK=(TM(I)-TM(J))*DIF1+TM(I)                                        LNG 691
      FK1=(TM(I+10)-TM(J+10))*DIF1+TM(I+10)                              LNG 692
      IT=(T+.00001)/5.                                                   LNG 693
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      DIF2=T-IT*5                                                       LNG 694
      IF(T.GE.135.)DIF2=T-130.                                          LNG 695
      IF(T.LT.90.)DIF2=T-90.                                            LNG 696
      FK=FK+(FK1-FK)*DIF2/5.                                            LNG 697
      IF(X(6).LT..0001)GO TO 17                                         LNG 698
      FKN=(TN(I)-TN(J))*DIF1+TN(I)                                      LNG 699
      FK1=(TN(I+10)-TN(J+10))*DIF1+TN(I+10)                             LNG 700
      FKN=FKN+(FK1-FKN)*DIF2/5.                                         LNG 701
      FK=FK+(FKN-FK)*X(6)/.0425                                         LNG 702
   17 FK=FK/1000.                                                       LNG 703
      FMKM=1./(VI-FK*X(1))                                              LNG 704
      RETURN                                                            LNG 705
   20 FMKM=0.0                                                          LNG 706
      RETURN                                                            LNG 707
      END                                                               LNG 708
      FUNCTION V  IDEAL(T,X)                                            LNG 709
C     CALCULATES THE IDEAL VOLUME OF A MIXTURE FOR THE K AND M METHOD   LNG 710
      DIMENSION X(10)                                                   LNG 711
      V=0                                                               LNG 712
      J=0                                                               LNG 713
      IF(X(6).GT..0001.AND.T.GT.115.)J=1                                LNG 714
      DO 10 I=1,8                                                       LNG 715
      IF(X(I).LE..000001)GO TO 10                                       LNG 716
      IF(J.GT.0.AND.I.EQ.6)GO TO 10                                     LNG 717
      V=V+X(I)/SAT(T,I)                                                 LNG 718
   10 CONTINUE                                                          LNG 719
      IF(J.EQ.1)V=V+X(6)/SATN2(T)                                       LNG 720
      VIDEAL=V                                                          LNG 721
      RETURN                                                            LNG 722
      END                                                               LNG 723
      FUNCTION SATN2(T)                                                 LNG 724
C     CALCULATES A PSEUDO SATURATED LIQUID DENSITY FOR N2 ABOVE 115 K   LNG 725
      IF(T.LT.115.)GO TO 1                                              LNG 726
      DELT=(T-115.)                                                     LNG 727
      SATN2=SAT(115.,6)+DELT*(SAT(115.05,6)-SAT(114.95,6))/.1           LNG 728
      RETURN                                                            LNG 729
    1 SATN2=SAT(T,6)                                                    LNG 730
      RETURN                                                            LNG 731
      END                                                               LNG 732
      FUNCTION SAT(T,I)                                                 LNG 733
C     CALCUALTES THE PURE FLUID DENSITIES FOR THE K AND M METHOD        LNG 734
C     UNITS ARE DEG K AND MOLES/LITER                                   LNG 735
      DIMENSION A(7,8)                                                  LNG 736
      DATA((A(I),I=1,7)=190.555,10.16,18.65812322,6.712030737,          LNG 737
     1-.9472019702,0.0,0.0)                                             LNG 738
      DATA((A(I),I=8,14)=305.33,                                        LNG 739
     1      6.86,12.55205121,13.43284373,-19.00461066,11.07715985,0.0)  LNG 740
      DATA((A(I),I=15,21)=369.82,5.,8.684458671,18.04085714,-29.46261356 LNG 741
     1,16.43559114,0.0)                                                 LNG 742
      DATA((A(I),I=22,28)=425.16,3.92,                                  LNG 743
     1                        7.286062567,11.96307859,-19.87591962,     LNG 744
     211.60211932,0.0)                                                  LNG 745
      DATA((A(I),I=29,35)=408.13,3.8,                                   LNG 746
     1                        7.657535400,8.145251283,-13.10582462,     LNG 747
     28.145894091,0.0)                                                  LNG 748
      DATA((A(I),I=36,42)=126.2,11.21,19.39216835,26.01408462,-39.497587 LNG 749
     191,23.32977312,0.0)                                               LNG 750
      DATA((A(I),I=43,49)=469.6,3.285,-.0362004993,59.00202990,         LNG 751
     1-93.44193819,43.66780833,0.0)                                     LNG 752
      DATA((A(I),I=50,56)=460.39,3.271,2.946310456,35.50770979,         LNG 753
     1-57.41242993,28.15898339,0.0)                                     LNG 754
      IF(T.GT.A(1,I))GO TO 1                                            LNG 755
      X=(1.-T/A(1,I))                                                   LNG 756
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      SAT=(A(2,I)+A(3,I)*X**(.35)+A(4,I)*X+A(5,I)*X**(4./3.)                LNG 757
     1+A(6,I)*X**(5./3.))                                                   LNG 758
      RETURN                                                                LNG 759
    1 SAT=1.E+20                                                            LNG 760
      RETURN                                                                LNG 761
      END                                                                   LNG 762
      SUBROUTINE ECKNON(PIN,DOUT,TIN,Q)                                     LNG 763
C     MAIN: ECKERT-RENON DENSITY MODEL                                      LNG 764
C                                                                           LNG 765
C                                                                           LNG 766
C        THIS PROGRAM IS DESIGNED FOR THE CALCULATION OF                    LNG 767
C             LNG LIQUID DENSITIES ONLY AND WILL NOT                        LNG 768
C             COMPUTE DENSITIES ABOVE 160 K.                                LNG 769
C                                                                           LNG 770
C                                                                           LNG 771
C        THIS PROGRAM IS CALIBRATED SOLELY AGAINST DATA                     LNG 772
C             MEASURED BY                                                   LNG 773
C                  UNITED STATES DEPARTMENT OF COMMERCE                     LNG 774
C                  NATIONAL BUREAU OF STANDARDS                             LNG 775
C                  CRYOGENICS LABORATORY                                    LNG 776
C                  BOULDER, COLORADO                                        LNG 777
C                                                                           LNG 778
C                                                                           LNG 779
C                                                                           LNG 780
C                                                                           LNG 781
C         >>>>>>>>>>>>>>NOTICE<<<<<<<<<<<<<<<<<<<<<<<<<<<<                 LNG 782
C        NO WARRANTY IS MADE OR IMPLIED AS TO THE ACCURACY                  LNG 783
C        OF DENSITIES CALCULATED BY USE OF THIS MODEL.                      LNG 784
C                                                                           LNG 785
C        USERS ARE AT THEIR OWN RISK IN THE USE OF THIS PROGRAM.            LNG 786
C                                                                           LNG 787
C                                                                           LNG 788
C                                                                           LNG 789
C     COMPONENT ID NUMBERS ARE                                              LNG 790
C        1 METHANE                                                          LNG 791
C        2 ETHANE                                                           LNG 792
C        3 PROPANE                                                          LNG 793
C        4 N-BUTANE                                                         LNG 794
C        5 I-BUTANE                                                         LNG 795
C        6 NITROGEN                                                         LNG 796
C        7 N-PENTANE                                                        LNG 797
C        8 I-PENTANE                                                        LNG 798
C                                                                           LNG 799
C                                                                           LNG 800
C                                                                           LNG 801
C                                                                           LNG 802
C     INPUT VARIABLES ARE                                                   LNG 803
C       PIN=PRESSURE IN BAR                                                 LNG 804
C       TIN=TEMPERATURE IN K                                                LNG 805
C      Q=MATRIX OF MOLE FPRACTIONS OF COMPONENTS                            LNG 806
C     OUTPUT VARIABLE IS                                                    LNG 807
C      DOUT=DENSITY IN MOLES/LITER                                          LNG 808
      DIMENSION VHOLD(12),VPHOLD(12),Q(8)                                   LNG 809
      TYPE INTEGER CNT1,CNT2,SWITCH                                         LNG 810
      TYPE REAL MWM,INCR,KIJ,MW,MOL,LAM,LAMB                                LNG 811
      COMMON /RUN/ID(12),X(12),NAM(2,12),C12(12,12),R(12),Z(12),S(12),VS    LNG 812
     1(12),TS(12),US(12),SG(12),EP(12),LAM(12),NC,MW(12),AIJ(12,12),PST(    LNG 813
     212),TCT(12)                                                           LNG 814
      COMMON /DAT/TBI(12),VBI(12),TBIM(12),VBIM(12),C(12),SGIM(12),SIM(1    LNG 815
     12),EPIM(12),PIM(12),SIN(12),VSIM(12),USIM(12),TSIM(12),DVBIM(12),P    LNG 816
     2D(12),PV(12),RHO(12),VBIMP(12),TTM(12),MWM,CNT1,CNT2,TOLD(12),INCR    LNG 817
     3(12),SWITCH,JPC,JPCS,JMIX,DENS,VEX,HEX,GEX,TMP,VMP,TP                 LNG 818
      COMMON/PAR/KIJ( 8, 8),AJI( 8, 8)                                      LNG 819
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      COMMON/UNITS/ITC,IPC,T,TMAX,DT,P,PMAX,DP,PS                    LNG 820
      SWITCH = 0                                                     LNG 821
    1 CALL ZERO1                                                     LNG 822
C     THE NEXT VALUES ARE SET TO PREVENT UNDER/OVER FLOWS            LNG 823
      TMAX = 100.0                                                   LNG 824
      DT = 100.0                                                     LNG 825
      PMAX = 100.0                                                   LNG 826
                                                                     LNG 827
      DP = 100.0                                                     LNG 828
      PS = 100.0                                                     LNG 829
      CALL INPUT(Q)                                                  LNG 830
      CODE=0                                                         LNG 831
      ITC=0                                                          LNG 832
      IPC=0                                                          LNG 833
      P=PIN/1.01325                                                  LNG 834
      T=TIN                                                          LNG 835
      IF(T.GT.(160.)) GO TO 19                                       LNG 836
    8 CALL PZERO(CTMX)                                               LNG 837
      MWM = 0                                                        LNG 838
      DO 9 I = 1,NC                                                  LNG 839
      MWM = MWM + (X(I)*MW(I))                                       LNG 840
    9 CONTINUE                                                       LNG 841
   10 DO 11 I = 1,NC                                                 LNG 842
      TBI(I) = T / TS(I)                                             LNG 843
      CALL VOLUME(TBI(I),VBI(I),JPUR,NC)                             LNG 844
      TBIM(I) = T / TSIM(I)                                          LNG 845
      CALL VOLUME(TBIM(I),VBIM(I),JMIX,NC)                           LNG 846
   11 CONTINUE                                                       LNG 847
      VM =  0.0                                                      LNG 848
      DO  12  I = 1,NC                                               LNG 849
      VM =  VM  + (X(I)*VBIM(I)*VSIM(I))                             LNG 850
   12 CONTINUE                                                       LNG 851
      TMP = ( T * 1.8) - (459.67)                                    LNG 852
      DO 13 I = 1,NC                                                 LNG 853
      VHOLD(I) = VBI(I)                                              LNG 854
      VPHOLD(I) = VBIM(I)                                            LNG 855
      PP = (P*TS(I))/US(I)                                           LNG 856
      VBIMP(I) = VBI(I)                                              LNG 857
      CALL PRES(PP,VBIMP(I),TBI(I),NC)                               LNG 858
      VBI(I) = VBIMP(I)                                              LNG 859
      PP = (P * TSIM(I)) / USIM(I)                                   LNG 860
      VBIMP(I) = VBIM(I)                                             LNG 861
      CALL PRES(PP,VBIMP(I),TBIM(I),NC)                              LNG 862
      VBIM(I) = VBIMP(I)                                             LNG 863
   13 CONTINUE                                                       LNG 864
      VMP = 0.0                                                      LNG 865
      DO 14 I = 1,NC                                                 LNG 866
      VMP = VMP + (X(I)*VBIMP(I)*VSIM(I))                            LNG 867
      PD(I) = VBIMP(I) * VSIM(I)                                     LNG 868
   14 CONTINUE                                                       LNG 869
      DENS=MWM/VMP                                                   LNG 870
      DOUT=DENS*1000./MWM                                            LNG 871
      RETURN                                                         LNG 872
   19 DOUT=0.0                                                       LNG 873
      RETURN                                                         LNG 874
      END                                                            LNG 875
      SUBROUTINE ZERO1                                               LNG 876
      COMMON /RUN/A(361)                                             LNG 877
      COMMON /DAT/B(224)                                             LNG 878
      DO 1 I = 1,361                                                 LNG 879
    1 A(I) = 0.0                                                     LNG 880
      DO 2 I = 1,224                                                 LNG 881
    2 B(I) = 0.0                                                     LNG 882
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      RETURN                                                            LNG 883
      END                                                               LNG 884
      SUBROUTINE INPUT(Q)                                               LNG 885
      DIMENSION NNO( 8),SIGM( 8),EPSI( 8),LAMB( 8),NAME(2, 8),SNO( 8),MO LNG 886
     1L( 8),CT( 8),TCH( 8),VCH( 8),ECH( 8),AR( 8),AZ( 8),Q(8)           LNG 887
      TYPE INTEGER CNT1,CNT2,SWITCH                                     LNG 888
      TYPE REAL MWM,INCR,KIJ,MW,MOL,LAM,LAMB                            LNG 889
      COMMON /RUN/ID(12),X(12),NAM(2,12),C12(12,12),R(12),Z(12),S(12),VS LNG 890
     1(12),TS(12),US(12),SG(12),EP(12),LAM(12),NC,MW(12),AIJ(12,12),PST( LNG 891
     212),TCT(12)                                                       LNG 892
      COMMON /DAT/TBI(12),VBI(12),TBIM(12),VBIM(12),C(12),SGIM(12),SIM(1 LNG 893
     12),EPIM(12),PIM(12),SIN(12),VSIM(12),USIM(12),TSIM(12),DVBIM(12),P LNG 894
     2D(12),PV(12),RHO(12),VBIMP(12),TTM(12),MWM,CNT1,CNT2,TOLD(12),INCR LNG 895
     3(12),SWITCH,JPC,JPCS,JMIX,DENS,VEX,HEX,GEX,TMP,VMP,TP             LNG 896
      COMMON/PAR/KIJ( 8, 8),AJI( 8, 8)                                  LNG 897
      DATA SIGM/                                                        LNG 898
     *0.991000,1.029000,1.155000,1.278000,1.388752,1.392995,            LNG 899
     *1.47162,1.47217/                                                  LNG 900
      DATA EPSI/                                                        LNG 901
     *0.640000,0.909000,1.69800,2.237000,2.705262,2.545907,             LNG 902
     *3.76195,3.68336/                                                  LNG 903
      DATA LAMB/                                                        LNG 904
     *1.053604,0.986325,1.227117,1.408519,1.473346,1.461676,            LNG 905
     *1.698790,1.695012/                                                LNG 906
      DATA  MOL/                                                        LNG 907
     *28.01600,16.04200,30.06800,44.09400,58.12000,58.12000,            LNG 908
     *72.146000,72.146000/                                              LNG 909
      DATA  SNO/                                                        LNG 910
     *10.00000,10.00000,10.00000,10.00000,10.00000,10.00000,            LNG 911
     *10.00000,10.00000/                                                LNG 912
      DATA CT/                                                          LNG 913
     *126.0600,190.5600,305.4300,369.8200,425.1600,408.0300,            LNG 914
     *469.65,460.39/                                                    LNG 915
      DATA  TCH/                                                        LNG 916
     *112.7699,170.9645,256.8311,294.7255,340.7867,322.6109,            LNG 917
     *411.0100,403.3200/                                                LNG 918
      DATA  VCH/                                                        LNG 919
     *26.01846,29.04010,40.88188,54.60301,68.72075,69.44272,            LNG 920
     *84.90197,84.99830/                                                LNG 921
      DATA  ECH/                                                        LNG 922
     *1393.000,1977.000,3695.000,4867.000,5886.652,5539.895,            LNG 923
     *8186.000,8015.000/                                                LNG 924
      DATA   AR/                                                        LNG 925
     *1.000000,1.000000,1.000000,1.000000,1.000000,1.000000,            LNG 926
     *1.000000,1.000000/                                                LNG 927
      DATA   AZ/                                                        LNG 928
     *10.00000,10.00000,10.00000,10.00000,10.00000,10.00000,            LNG 929
     *10.00000,10.00000/                                                LNG 930
      DO 4 I=2,6                                                        LNG 931
    4 X(I)=Q(I-1)                                                       LNG 932
      X(1)=Q(6)                                                         LNG 933
      X(7)=Q(7)                                                         LNG 934
      X(8)=Q(8)                                                         LNG 935
      NC=0                                                              LNG 936
      DO 1 I=1,8                                                        LNG 937
      IF(X(I).LE.0.0)GO TO 1                                            LNG 938
      NC=NC+1                                                           LNG 939
      X(NC)=X(I)                                                        LNG 940
      ID(NC)=I                                                          LNG 941
    1 CONTINUE                                                          LNG 942
      DO 2 I = 1,NC                                                     LNG 943
      J = ID(I)                                                         LNG 944
      R(I) = AR(J)                                                      LNG 945
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      Z(I) = AZ(J)                                                          LNG 946
      S(I) = SNO(J)                                                         LNG 947
      TS(I)= TCH(J)                                                         LNG 948
      US(I)= ECH(J)                                                         LNG 949
      SG(I) = SIGM(J)                                                       LNG 950
      TCT(I) = CT(J)                                                        LNG 951
      EP(I) = EPSI(J)                                                       LNG 952
      LAM(I) = LAMB(J)                                                      LNG 953
      VS(I) = VCH(J)                                                        LNG 954
      MW(I) = MOL(J)                                                        LNG 955
    2 CONTINUE                                                              LNG 956
      DO 3 I = 1,NC                                                         LNG 957
      DO 3 J = 1,NC                                                         LNG 958
      M = ID(I)                                                             LNG 959
      L = ID(J)                                                             LNG 960
      C12(I,J) = KIJ(M,L)                                                   LNG 961
      C12(J,I) = KIJ(L,M)                                                   LNG 962
      AIJ(I,J) = AJI(M,L)                                                   LNG 963
      AIJ(J,I) = AJI(L,M)                                                   LNG 964
    3 CONTINUE                                                              LNG 965
      RETURN                                                                LNG 966
      END                                                                   LNG 967
      END                                                                   LNG 968
      SUBROUTINE PZERO(TRT)                                                 LNG 969
      TYPE INTEGER CNT1,CNT2,SWITCH                                         LNG 970
      TYPE REAL MWM,INCR,KIJ,MW,MOL,LAM,LAMB                                LNG 971
      COMMON /RUN/ID(12),X(12),NAM(2,12),C12(12,12),R(12),Z(12),S(12),VS    LNG 972
     1(12),TS(12),US(12),SG(12),EP(12),LAM(12),NC,MW(12),AIJ(12,12),PST(    LNG 973
     212),TCT(12)                                                           LNG 974
      COMMON /DAT/TBI(12),VBI(12),TBIM(12),VBIM(12),C(12),SGIM(12),SIM(1    LNG 975
     12),EPIM(12),PIM(12),SIN(12),VSIM(12),USIM(12),TSIM(12),DVBIM(12),P    LNG 976
     2D(12),PV(12),RHO(12),VBIMP(12),TTM(12),MWM,CNT1,CNT2,TOLD(12),INCR    LNG 977
     3(12),SWITCH,JPC,JPCS,JMIX,DENS,VEX,HEX,GEX,TMP,VMP,TP                 LNG 978
      COMMON/PAR/KIJ( 8, 8),AJI( 8, 8)                                      LNG 979
      COMMON /UNITS/ITC,IPC,T,TMAX,DT,P,PMAX,DP,PS                          LNG 980
      IF(NC.EQ.1) TRT = T/TCT(1)                                            LNG 981
      IF(NC.GT.1) CALL TCM(TRT)                                             LNG 982
      IF(NC.GT.1) TRT = T/TRT                                               LNG 983
      IF(NC.EQ.1) CALL PURE(TRT,TRT,R(1),RXX)                               LNG 984
      DO 1 I = 1,NC                                                         LNG 985
      TCXX = T/TCT(I)                                                       LNG 986
      IF(NC.GT.1) CALL PURE(TCXX,TRT,R(I),RTR)                              LNG 987
      JPC = 0                                                               LNG 988
      IF(TRT.GT.(.87)) JPC = 2                                              LNG 989
      IF(TRT.GT.(1.0)) JPC = 1                                              LNG 990
      IF(NC.EQ.1) RTR = RXX                                                 LNG 991
C     THERE ARE SEVERAL OTHER FORMS OF THESE EQUATIONS                      LNG 992
C     REFER TO ORIGINAL ARTICLES FOR VALUES TO USE                          LNG 993
      S(I) = RTR  * Z(I) - 2. * RTR  + 2.                                   LNG 994
      SIM(I) = S(I)                                                         LNG 995
                                                                            LNG 996
      SG(I) = (VS(I)/RTR ) **(1./3.)                                        LNG 997
      EP(I) = US(I) / S(I)                                                  LNG 998
      C(I) = LAM(I) * (2176./((185.6)*(1.98726)))                           LNG 999
      TTM(I) = TS(I)                                                        LNG1000
      TOLD(I) = TS(I)                                                       LNG1001
      INCR(I) = (20.0)                                                      LNG1002
    1 CONTINUE                                                              LNG1003
      JPUR = 0                                                              LNG1004
      CNT1 = 1                                                              LNG1005
      CNT2 = 1                                                              LNG1006
      JMIX = 0                                                              LNG1007
    2 DEN = 0                                                               LNG1008
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      DO 3 I = 1,NC                                                    LNG1009
      DEN = DEN + (X(I) * SIM(I))                                      LNG1010
    3 CONTINUE                                                         LNG1011
      DO 4 I = 1,NC                                                    LNG1012
      PIM(I) = (X(I) * SIM(I)) / DEN                                   LNG1013
    4 CONTINUE                                                         LNG1014
      DO 6 I = 1,NC                                                    LNG1015
      SGIM(I) = 0.0                                                    LNG1016
      DO 6 J = 1,NC                                                    LNG1017
      M = I                                                            LNG1018
      N = J                                                            LNG1019
      IF(I.GT.J) M = J                                                 LNG1020
      IF(I.GT.J) N = I                                                 LNG1021
      IF(I.EQ.J) GO TO 5                                               LNG1022
      TRUDX = (AIJ(N,M)/(T/TTM(I)))                                    LNG1023
      IF(TRUDX.LT.(-180.0)) GO TO 5                                    LNG1024
      SGIM(I) = SGIM(I) +((PIM(J) *(((SG(I)**(1./3.) + SG(J)**(1./3.))  LNG1025
     1/(2.0)) **3))* (AIJ(M,N) * EXP(AIJ(N,M)/(T/TTM(I)))))            LNG1026
      GO TO 6                                                          LNG1027
    5 SGIM(I) = SGIM(I) +(PIM(J) *(((SG(I)**(1./3.) + SG(J)**(1./3.))  LNG1028
     1/(2.0)))**3 )                                                    LNG1029
    6 CONTINUE                                                         LNG1030
      DEN = 0.0                                                        LNG1031
      DO 7 I = 1,NC                                                    LNG1032
      DEN = DEN + (PIM(I)* SGIM(I)* SGIM(I))                           LNG1033
    7 CONTINUE                                                         LNG1034
      DO 8 I = 1,NC                                                    LNG1035
      SIN(I) =(S(I) * (SGIM(I)**(2.))) / DEN                           LNG1036
    8 CONTINUE                                                         LNG1037
                                                                       LNG1038
      DO 9 I = 1,NC                                                    LNG1039
      TEST = (1.0) - (SIM(I) / SIN(I))                                 LNG1040
      TEST = ABS (TEST)                                                LNG1041
      IF(TEST.GT.(0.00001)) GO TO 10                                   LNG1042
    9 CONTINUE                                                         LNG1043
      GO TO 12                                                         LNG1044
   10 DO 11 I = 1,NC                                                   LNG1045
      SIM(I) =(SIM(I) + SIN(I)) /(2.0)                                 LNG1046
   11 CONTINUE                                                         LNG1047
      CNT1 = CNT1 + 1                                                  LNG1048
      IF(CNT1.GT.250) GO TO 12                                         LNG1049
      GO TO 2                                                          LNG1050
   12 DO 14 I = 1,NC                                                   LNG1051
      SIM(I) = SIN(I)                                                  LNG1052
      EPIM(I) = 0.0                                                    LNG1053
      DO 14 J = 1,NC                                                   LNG1054
      K = I                                                            LNG1055
      L = J                                                            LNG1056
      IF(J.LT.I) K = J                                                 LNG1057
      IF(J.LT.I) L = I                                                 LNG1058
      IF(I.EQ.J) GO TO 13                                              LNG1059
      TRUDX = C12(L,K) / (T/TTM(I))                                    LNG1060
      IF(TRUDX.LT.(-180.0)) GO TO 13                                   LNG1061
      EPIM(I) = EPIM(I) +((PIM(J) * SQRT(EP(I)*EP(J)))*                LNG1062
     1(C12(K,L) * EXP((C12(L,K) /(T/TTM(I))))))                        LNG1063
      GO TO 14                                                         LNG1064
   13 EPIM(I) = EPIM(I) + (PIM(J) * SQRT(EP(I) * EP(J)))               LNG1065
   14 CONTINUE                                                         LNG1066
C     ASSUME NUMBER OF MOLS OF MIXTURE = 1.0                           LNG1067
      DO 15 I = 1,NC                                                   LNG1068
      VSIM(I) = R(I) * (SGIM(I)**(3.0))                                LNG1069
      USIM(I) = SIM(I) * EPIM(I)                                       LNG1070
      TSIM(I) = USIM(I) / ((1.98726) * C(I))                           LNG1071
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15 CONTINUE                                                          LNG1072
   DO 16 I = 1,NC                                                    LNG1073
   TEST = (1.0) - (TSIM(I)/TTM(I))                                   LNG1074
   TEST = ABS(TEST)                                                  LNG1075
   IF(TEST.GT.(0.00001)) GO TO 17                                    LNG1076
16 CONTINUE                                                          LNG1077
   RETURN                                                            LNG1078
17 DO 20 I = 1,NC                                                    LNG1079
                                                                     LNG1080
   IF(TSIM(I) - TTM(I))18,20,19                                      LNG1081
18 TNEW = TTM(I) - INCR(I)                                           LNG1082
   IF(TNEW.EQ.TOLD(I)) INCR(I) = INCR(I)/ (2.0)                      LNG1083
   IF(TNEW.EQ.TOLD(I)) GO TO 18                                      LNG1084
   TOLD(I) = TTM(I)                                                  LNG1085
   TTM(I) = TNEW                                                     LNG1086
   GO TO 20                                                          LNG1087
19 TNEW = TTM(I) + INCR(I)                                           LNG1088
   IF(TNEW.EQ.TOLD(I)) INCR(I) = INCR(I) / (2.0)                     LNG1089
   IF(TNEW.EQ.TOLD(I)) GO TO 19                                      LNG1090
   TOLD(I) = TTM(I)                                                  LNG1091
   TTM(I) = TNEW                                                     LNG1092
20 CONTINUE                                                          LNG1093
   CNT2 = CNT2 + 1                                                   LNG1094
   IF(CNT2.GT.250) RETURN                                            LNG1095
   GO TO 2                                                           LNG1096
   END                                                               LNG1097
   SUBROUTINE PURE(TZ,T,Z,R)                                         LNG1098
   DIMENSION A(15)                                                   LNG1099
   IF(TZ.LT.(.8653525)) R = Z                                        LNG1100
   IF(TZ.LT.(.8653525)) GO TO 2                                      LNG1101
   IF(T.LT.(.8653525)) R = Z                                         LNG1102
   IF(T.LT.(.8653525)) GO TO 2                                       LNG1103
   IF(T.GT.(1.0)) TA = 1.0                                           LNG1104
   IF(T.LE.(1.0)) TA = T                                             LNG1105
   DATA A/.9184780,-.1530647,-.1090050,.8073883,1.441803,            LNG1106
  1-10.85944,-6.041687,51.26758,.9062500,-108.9805,28.88672,         LNG1107
  2106.1406,-45.78125,-38.43750,20.56250/                            LNG1108
   R = 0.0                                                           LNG1109
   TT = (TA - (.9267292)) / (.7267517E-01)                           LNG1110
   DO 1 K = 1,14                                                     LNG1111
   R = R + A(16-K)                                                   LNG1112
   R = R * TT                                                        LNG1113
 1 CONTINUE                                                          LNG1114
   R = R + A(1)                                                      LNG1115
 2 RETURN                                                            LNG1116
   END                                                               LNG1117
   SUBROUTINE VOLUME(T,V,J,NC)                                       LNG1118
   TR = T                                                            LNG1119
   IF(TR.GT.(1.00)) GO TO 4                                          LNG1120
   V = 0.5                                                           LNG1121
                                                                     LNG1122
 1 VT = (1.0) + (0.1*TR*V**(4.0))                                    LNG1123
   ERR = (1.0) - (V/VT)                                              LNG1124
   TEST = ABS(ERR)                                                   LNG1125
   IF(TEST.LE.(0.00001)) GO TO 2                                     LNG1126
   V = VT                                                            LNG1127
   GO TO 1                                                           LNG1128
 2 V = V**(3.0)                                                      LNG1129
 3 RETURN                                                            LNG1130
 4 V = ((((((10.06600 *TR)-24.79837)*TR)+23.260722)*TR)-6.686880)    LNG1131
   GO TO 3                                                           LNG1132
   END                                                               LNG1133
   SUBROUTINE BETA(P,DV,T,V,KK)                                      LNG1134
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      TYPE REAL AF,BF,CF,DF,EF,AG,BG,CG,DG,P,DV,T,V,F,F1,G,G1,DEN,EG     LNG1135
      TYPE REAL C,B1,B2,B3                                              LNG1136
      DIMENSION C(11)                                                   LNG1137
      DATA AF/0.11566564E+02/,BF/-0.53510144E+01/,CF/-0.74598207E-01/,  LNG1138
     1DF/0.67068653E+00/,EF/-0.11939887E+00/,AG/0.62721813E+01/,        LNG1139
     2BG/0.47698365E+00/,CG/-0.16023080E+01/,DG/0.49837746E+00/,        LNG1140
     3EG/-0.42639183E-01/                                               LNG1141
      F = AF +(BF*V)+(CF*V*V)+ (DF*V*V*V)+(EF*V*V*V*V)                  LNG1142
      IF(F.LT.(0.0)) GO TO 1                                            LNG1143
      F1 = (BF+(CF*V*2.E+00)+(DF*V*V*3.E+00)+(EF*V*V*V*4.E+00))*        LNG1144
     1T * EXP(F)                                                        LNG1145
      G = AG+(BG*V)+(CG*V*V)+(DG*V*V*V)+(EG*V*V*V*V)                    LNG1146
      IF(G.LT.(0.00)) GO TO 1                                           LNG1147
      G1 = (BG+(CG*V*2.E+00)+(DG*V*V*3.E+00+(EG*V*V*V*4.E+00))) * EXP(G LNG1148
     1)                                                                 LNG1149
      G1 = -G1                                                          LNG1150
      DEN = F1 + G1                                                     LNG1151
      DV = (1.E+00) / DEN                                               LNG1152
      IF(DV.GT.(0.0).OR.DV.LT.(-1.0E+00)) GO TO 1                       LNG1153
      GO TO 2                                                           LNG1154
      DATA C/-.118659822E+02,-.5509099E+00,.4172102E+01,-.8996686E+00,  LNG1155
     1-.1500376E+01,-.1958324E+00,-.2788988E+01,.5195283E+00,           LNG1156
     2.3734878E+01,.1361904E+00,.4671948E-01/                           LNG1157
    1 B1 = C(1) + C(2)*T + C(3)*V                                       LNG1158
      B2 = C(4)*((V+C(5))**(2 ))*EXP((C(6))*((T+C(7))**(2 )))           LNG1159
      B3 = C(11) * (V+C(8))**(C(9))/(T**(C(10)))                        LNG1160
      DV = B1 + B2 + B3                                                 LNG1161
      DV = EXP(DV)                                                      LNG1162
      DV = - DV                                                         LNG1163
    2 RETURN                                                            LNG1164
      END                                                               LNG1165
      SUBROUTINE PRES(A,B,C,NC)                                         LNG1166
      TYPE REAL H,P,V,T,K1,K2,K3,K4,VT,PT                               LNG1167
      P = A                                                             LNG1168
      V = B                                                             LNG1169
      T = C                                                             LNG1170
      H = 1.E-02                                                        LNG1171
      IF(H.GT.P) H = P                                                  LNG1172
      ASSIGN 3 TO KK                                                    LNG1173
    1 CALL BETA(P,K1,T,V,NC)                                            LNG1174
      K1 = H * K1                                                       LNG1175
      VT = V + ((0.5E+00)  *K1)                                         LNG1176
      PT = P + ((0.5E+00)*H)                                            LNG1177
      CALL BETA(PT,K2,T,VT,NC)                                          LNG1178
      K2 = K2 * H                                                       LNG1179
      VT = V + ((0.5E+00)  *K2)                                         LNG1180
      CALL BETA(PT,K3,T,VT,NC)                                          LNG1181
      K3 = K3 * H                                                       LNG1182
      PT = P + H                                                        LNG1183
      VT = V + (  K3)                                                   LNG1184
      CALL BETA(PT,K4,T,VT,NC)                                          LNG1185
      K4 = K4 * H                                                       LNG1186
      V = V + (((K1 + (2.E+00*K2) + (2.E+00*K3) + K4) / (6.E+00))    )  LNG1187
      P = P - H                                                         LNG1188
      IF(P.EQ.(0.0)) GO TO 4                                            LNG1189
      IF(P.LT.(0.0)) GO TO 2                                            LNG1190
      GO TO 1                                                           LNG1191
    2 GO TO KK , (3,4)                                                  LNG1192
    3 P = P + H                                                         LNG1193
      H = P                                                             LNG1194
      ASSIGN 4 TO KK                                                    LNG1195
      GO TO 1                                                           LNG1196
    4 A = P                                                             LNG1197
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```
      B = V                                                              LNG1198
      C = T                                                              LNG1199
      RETURN                                                             LNG1200
      END                                                                LNG1201
      SUBROUTINE TCM(TMC)                                                LNG1202
      TYPE REAL NUM                                                      LNG1203
      TYPE INTEGER CNT1,CNT2,SWITCH                                      LNG1204
      TYPE REAL MWM,INCR,KIJ,MW,MOL,LAM,LAMB                             LNG1205
      DIMENSION VCI(6),TH(12)                                            LNG1206
      COMMON /RUN/ID(12),X(12),NAM(2,12),C12(12,12),R(12),Z(12),S(12),VS LNG1207
     1(12),TS(12),US(12),SG(12),EP(12),LAM(12),NC,MW(12),AIJ(12,12),PST( LNG1208
     212),TCT(12)                                                        LNG1209
      COMMON /DAT/TBI(12),VBI(12),TBIM(12),VBIM(12),C(12),SGIM(12),SIM(1 LNG1210
     12),EPIM(12),PIM(12),SIN(12),VSIM(12),USIM(12),TSIM(12),DVBIM(12),P LNG1211
     2D(12),PV(12),RHO(12),VBIMP(12),TTM(12),MWM,CNT1,CNT2,TOLD(12),INCR LNG1212
     3(12),SWITCH,JPC,JPCS,JMIX,DENS,VEX,HEX,GEX,TMP,VMP,TP              LNG1213
      COMMON/PAR/KIJ( 8, 8),AJI( 8, 8)                                   LNG1214
      COMMON/UNITS/ITC,IPC,T,TMAX,DT,P,PMAX,DP,PS                        LNG1215
      DATA VCI/ 1.44,1.59,2.27,3.18,4.03,4.21/                           LNG1216
      DO 1 I = 1,NC                                                      LNG1217
      J = ID(I)                                                          LNG1218
      V = VCI(J)**(2./3.)                                                LNG1219
      TH(I) = X(I) * V                                                   LNG1220
    1 CONTINUE                                                           LNG1221
      TTZ = 0.0                                                          LNG1222
      DO 2 I = 1,NC                                                      LNG1223
      TTZ = TTZ + TH(I)                                                  LNG1224
    2 CONTINUE                                                           LNG1225
      DO 3 I = 1,NC                                                      LNG1226
      TH(I) = TH(I) / TTZ                                                LNG1227
    3 CONTINUE                                                           LNG1228
      SUM1 = 0.0                                                         LNG1229
      SUM2 = 0.0                                                         LNG1230
      K = NC - 1                                                         LNG1231
      DO 5 I = 1,K                                                       LNG1232
      L = I + 1                                                          LNG1233
      DO 4 J = L,NC                                                      LNG1234
      TTZ = (TCT(I)-TCT(J))/(TCT(I)+TCT(J))                              LNG1235
      TTZ= ABS(TTZ)                                                      LNG1236
      T12 = (((((((TTZ*(-3.038))+(5.443))*TTZ)+(-1.343))*TTZ)            LNG1237
     1 + (0.287))*TTZ) - (.0076)                                         LNG1238
      T12 = T12*(TCT(I) + TCT(J)) * (0.9)                                LNG1239
      SUM2 = SUM2 + ((2.)*TH(I)*TH(J)*T12)                               LNG1240
    4 CONTINUE                                                           LNG1241
      SUM1 = SUM1 + (TH(I)*TCT(I)*(1.8))                                 LNG1242
    5 CONTINUE                                                           LNG1243
      TMC = SUM1 + SUM2 + (TH(NC)*TCT(NC)*(1.8))                         LNG1244
      V = 0.0                                                            LNG1245
      DO 6 I = 1,NC                                                      LNG1246
                                                                         LNG1247
      J = ID(I)                                                          LNG1248
      TH(I) = X(I) * VCI(J)                                              LNG1249
      V = V + TH(I)                                                      LNG1250
    6 CONTINUE                                                           LNG1251
      DO 7 I = 1,NC                                                      LNG1252
      TH(I) = TH(I) / V                                                  LNG1253
    7 CONTINUE                                                           LNG1254
      TMM = 0.0                                                          LNG1255
      DO 9 I = 1,NC                                                      LNG1256
      DO 8 J = 1,NC                                                      LNG1257
      M = ID(I)                                                          LNG1258
      N = ID(J)                                                          LNG1259
      NUM = (VCI(M)**(1./3.)) * (VCI(N)**(1./3.))                        LNG1260
```

```
      NUM = SQRT(NUM)                                                   LNG1261
      DEN = (0.5)*((VCI(M)**(1./3.))+(VCI(N)**(1./3.)))                 LNG1262
      NUM = NUM / DEN                                                   LNG1263
      NUM = NUM**(3.)                                                   LNG1264
      AKIJ = (1.0) - NUM                                                LNG1265
      TCIJ = (1.0 - AKIJ) * SQRT(TCT(I)*TCT(J)*1.8*1.8)                 LNG1266
      TMM = TMM + TH(I)*TH(J)*TCIJ                                      LNG1267
    8 CONTINUE                                                          LNG1268
    9 CONTINUE                                                          LNG1269
      TCMP = TMM + (10.0)                                               LNG1270
      ICT = 0                                                           LNG1271
      TPO = TMM  + (10.0)                                               LNG1272
   10 TR = (T*1.8)/TCMP                                                 LNG1273
      NUM = (2901.01) -((5738.92)*TR) +((2849.85)*TR*TR)                LNG1274
     1 + ((1.74127)/ (1.01 - TR))                                       LNG1275
      NUM = NUM * (TR - (1.0))                                          LNG1276
      IF(NUM.LT.(-180.)) DD = 0                                         LNG1277
      IF(NUM.LT.(-180.)) GO TO 11                                       LNG1278
      DD = EXP(NUM)                                                     LNG1279
C        THE STATEMENT ABOVE MAY RESULT IN AN UNDERFLOW SENSE LIGHT     LNG1280
C          ON SOME OPERATING SYSTEMS WHEN THE NUMBER NUM IS A LARGE     LNG1281
C          NEGATIVE NUMBER. THE LARGE NEGATIVE VALUE IS PROPER AND      LNG1282
C          THE CORRECT ANSWER FOR DD IS ZERO.                           LNG1283
   11 TCMP = TMM + ((TMC-TMM)*DD)                                       LNG1284
      TEST = (1.0) - (TCMP/TPO)                                         LNG1285
      TEST = ABS(TEST)                                                  LNG1286
      IF(TEST.LT.(.0001)) GO TO 12                                      LNG1287
      ICT = ICT + 1                                                     LNG1288
                                                                        LNG1289
      IF(ICT.GT.250) GO TO 12                                           LNG1290
      TPO = TCMP                                                        LNG1291
      GO TO 10                                                          LNG1292
   12 TMC = TCMP/(1.8)                                                  LNG1293
      RETURN                                                            LNG1294
      END                                                               LNG1295
      SUBROUTINE BLOCK                                                  LNG1296
      TYPE REAL KIJ,AJI                                                 LNG1297
      DIMENSION KIJ(8,8),AJI(8,8)                                       LNG1298
      COMMON /PAR/ KIJ,AJI                                              LNG1299
      DATA KIJ/                                                         LNG1300
     *0.000000,.293E-07,.911E-06,.0030498,0.001000,0.001000,0.001000,   LNG1301
     *0.001000,1.088608,0.000000,.188E-10,.463E-06,.462E-08,.120E-03,   LNG1302
     *0.001000,0.001000,1.098880,1.078910,0.000000,.517E-06,.535E-06,   LNG1303
     *.188E-10,0.001000,0.001000,.8578232,1.146026,1.009559,0.000000,   LNG1304
     *.546E-06,.136E-05,0.001000,0.001000,0.995000,1.287762,.9868486,   LNG1305
     *.9690485,0.000000,.541E-06,0.001000,0.001000,1.178993,1.266327,   LNG1306
     *1.016231,.9976706,.9435714,0.000000,0.001000,0.001000,0.950000,   LNG1307
     *1.000000,1.000000,1.000000,1.000000,1.000000,0.000000,0.001000,   LNG1308
     *0.995000,1.000000,1.000000,1.000000,1.000000,1.000000,1.000000,   LNG1309
     *0.000000/                                                         LNG1310
      DATA AJI/                                                         LNG1311
     *0.000000,.161E-04,.257E-06,.0003462,0.001000,0.001000,0.001000,   LNG1312
     *0.001000,1.014967,0.000000,.188E-10,.197E-06,.184E-05,.335E-06,   LNG1313
     *0.001000,0.001000,1.008061,1.004105,0.000000,.142E-06,.103E-04,   LNG1314
     *.188E-10,0.001000,0.001000,.9863463,1.011399,1.001862,0.000000,   LNG1315
     *.105E-04,.119E-05,0.001000,0.001000,0.995000,1.024894,1.003289,   LNG1316
     *.9987221,0.000000,.119E-05,0.001000,0.001000,.9566020,1.025269,   LNG1317
     *1.005949,1.001225,.9968984,0.000000,0.001000,0.001000,0.995000,   LNG1318
     *1.000000,1.000000,1.000000,1.000000,1.000000,0.000000,0.001000,   LNG1319
     *0.995000,1.000000,1.000000,1.000000,1.000000,1.000000,1.000000,   LNG1320
     *0.000000/                                                         LNG1321
      END                                                               LNG1322
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10. SUPPLEMENTARY NOTES

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)

Four mathematical models of the equation of state for LNG like mixtures are presented. The four models include an extended corresponding states model, a cell model, a hard sphere model and a revised Klosek and McKinley model. Each of the models has been optimized to the same experimental data set which included data for pure nitrogen, methane, ethane, propane, iso and normal butane, iso and normal pentane and mixtures thereof. For LNG like mixtures (mixtures of the orthobaric liquid state at temperatures of 120 K or less and containing at least 60% methane, less than 4% nitrogen, less than 4% each of iso and normal butane and less than 2% total of iso and normal pentane), all of the models are estimated to predict densities to within 0.1% of the true value. The revised Klosek and McKinley model is valid only for mixtures within the range of temperature and composition specified above while the other three models are valid for a broader range of pressure, temperature and composition. The experimental PVTx data set used in the optimization together with comparisons are given and listings of computer programs for each of the models are included.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)

Cell model; comparisons; computer programs; corresponding states; equation of state; hard sphere; LNG; mixtures; PVTx data; revised Kosek and McKinley.

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