Evaluation of Binary Excess Volume Data for C_6 Hydrocarbons. Benzene + Hexane

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The excess volume data for the benzene + hexane system are evaluated. The needs for new experimental data are defined.

Key words: benzene; excess volume; hexane; volume change of mixing.

Contents

List of Tables

List of Figures

1. Introduction

Of the C_6 hydrocarbon + C_6 hydrocarbon binary systems, three systems—benzene + cyclohexane, benzene + n-hexane, and cyclohexane + n-hexane—have special significance for correlators and experimentalists. Together they represent the molecular interactions between three important hydrocarbon types—aromatics, cycloalkanes, and alkanes. Also, these three binary systems have been widely used as test systems for vapor–liquid equilibrium, excess enthalpy, and excess volume measurement devices. As a result, relatively large amounts of VLE, H^E, and V^E data appear in the literature for each.

This paper presents the evaluation results for the V^E data for the benzene + hexane system. Parallel papers cover the VLE and H^E data. The benzene + cyclohexane system has been covered in three preceding papers[1–3] which also presented a detailed description of the evaluation procedures used for the individual properties. The cyclohexane + hexane system will be covered in three subsequent papers.

In each of these evaluation papers, an attempt is made to establish selected values of the property at mole fractions of 0.25, 0.50, and 0.75 over the temperature range covered by the literature data. The availability of selected values at three mole fractions will hopefully reduce the practice of comparing data sets only at the midpoint.

Whenever possible, recommended data sets are identified. If no data set can be recommended, the best data sets are identified.

Finally, based on the evaluation results, the needs for new experimental measurements are defined.

The procedures used to evaluate V^E data are not repeated in this and subsequent papers. The reader must obtain that information from the benzene + cyclohexane V^E data paper.[1]

2. Summary of Evaluation Results

Table 1 lists the evaluation results for the 31 sets of benzene + hexane V^E data evaluated. Each set of data is represented by a single line. The literature reference numbers are the Laboratory's Master Reference List (MRL) numbers which were assigned to the individual documents when they were retrieved. The literature citation for a given MRL number can be found in the Bibliography. The MRL number also appears on the tabulation for each set of data. Note that the data sets listed in Table 1 are ordered with respect to temperature. In most cases, an experimental pressure was not reported and has been assumed to be one atmosphere (101.325 kPa).

A previous paper[1] for the benzene + cyclohexane system has presented the evaluation methods and the significance of the reported results. There are five quality ratings ranging from A for very good data to E for very bad data. The quality rating assigned to a set of data represents a "summary" of the evaluation results.

Only two kinds of tests are applied to V^E data. The first is a scatter rating which not only reflects scatter in the experimental data points but also reflects how well the shapes of the experimental data plots agree with the basic characteristic shapes for the particular system. Seven scatter ratings are used: E = excellent, G = good, F = fair, U = unacceptable, M = marginal, S = smoothed, and N = none.

The second test is a comparison of the data set values to the "best" V^E versus T curves at three mole fractions: $x_1 = 0.25$, 0.50, and 0.75. The % deviation values in Table 1 show how far the individual data set points deviate from the best curves established by the evaluator. The test compares data sets at the same temperature and also at different temperatures.

The use of the V^E data sets for correlation purposes should be restricted to those with a quality rating of A, B, or C with a C set being used only if no A or B set is available.

Table 1. Summary list for excess volume data

LITERATURE REFERENCE	T, K	P, MPA	QUALITY RATING	SCATTER RATING	% DEVIATION FROM VE VS. T CURVE X(1)=0.25	X(1)=0.50	X(1)=0.75
BENZENE(1) + HEXANE(2)							
00141	273.150	0.1013	C	F	0.0	-0.3	0.0
03966	288.150	0.1013	D	F	4.4	2.4	-1.8
00171	293.150	0.1013	E	U	-	-	-
15650	293.150	0.1013	DE	M	-	-	-
21347	293.150	0.1013	E	M	-54.9	-	-37.9
00184	295.150	0.1013	E	F	6.2	5.7	2.1
00070	298.150	0.1013	E	M	-	21.3	30.1
00165	298.150	0.1013	E	F	0.0	11.3	19.3
00220	298.150	0.1013	E	U	-	14.9	-33.9
00917	298.150	0.1013	E	U	-18.4	-21.9	-77.8
01744	298.150	0.1013	C	F	1.1	-1.0	-1.5
01990	298.150	0.1013	E	M	8.1	13.7	4.1
03966	298.150	0.1013	D	F	4.5	3.2	0.6
08516	298.150	0.1013	D	F	-	0.5	5.3
10174	298.150	0.1013	C	F	-2.7	0.0	-1.3
10296	298.150	0.1013	E	F	-6.2	0.5	-1.9
18511	298.150	0.1013	DE	M	-	-	-
40227	298.150	0.1013	E	M	27.5	30.0	23.4
00070	303.150	0.1013	DE	M	-	-	-
00271	303.150	0.1013	E	U	-72.4	-51.6	-80.7
01773	303.150	0.1013	D	M	-3.2	-0.7	-3.3
03966	303.150	0.1013	D	M	0.2	2.6	-3.2
00070	308.150	0.1013	DE	M	-	-	-
01773	308.150	0.1013	E	M	-12.0	-3.0	-2.7
03966	308.150	0.1013	E	M	-2.5	8.0	1.7
00070	313.150	0.1013	DE	M	-	-	-
01773	313.150	0.1013	D	M	-3.4	-0.7	-4.9
15650	313.150	0.1013	DE	M	-	-	-
10295	323.150	0.1013	C	F	0.0	-0.5	0.0
40227	323.150	0.1013	E	M	18.5	26.1	-5.9
15650	333.150	0.1013	DE	M	-	-	-

3. Selected Point Values at 298.15 K

Tables 2, 3, and 4 show the V^E values at $x_1 = 0.25$, 0.50, and 0.75 for the benzene + hexane system. The data sets are listed in the order of increasing magnitude of the V^E values. The literature source for each set of data is identified by the MRL (Master Reference List) number in the first column; the MRL numbers are related to the document citations in the Bibliography. The scatter rating code (S = smoothed, E = excellent, G = good, F = fair, U = unacceptable, M = marginal, N = no scatter rating) appears in the S (for

Table 2. Magnitude listing of excess volume values at 298.15 K and x(1) = 0.25. The S and ET codes refer to the scatter rating and equipment type respectively.

MRL number	Codes S	ET	Authors	Year	Excess volume cc/mol
917	U	01	Ridgway, Butler	1967	0.2400
10296	F	04	Letcher	1975	0.2760
10174	F	01	Letcher	1972	0.2863
165	F	01	Mathieson, Thynne	1956	0.2942
1744	F	01	Harris, Dunlop	1970	0.2973
3966	F	04	Nigam, Mahl, Singh	1972	0.3073
1990	M	01	Heric, Brewer	1967	0.3180
40227	M	01	Teja, Rice	1976	0.3750

Table 3. Magnitude listing of excess volume values at 298.15 K and x(1) = 0.50. The S and ET codes refer to the scatter rating and equipment type respectively.

MRL number	Codes S	ET	Authors	Year	Excess volume cc/mol
917	U	01	Ridgway, Butler	1967	0.3122
1744	F	01	Harris, Dunlop	1970	0.3960
10174	F	01	Letcher	1972	0.4000
8516	F	01	Diaz Pena, Nunez Delgado	1974	0.4020
10296	F	04	Letcher	1975	0.4020
3966	F	04	Nigam, Mahl, Singh	1972	0.4127
165	F	01	Mathieson, Thynne	1956	0.4450
1990	M	01	Heric, Brewer	1967	0.4550
220	U	01	Williams, Ogg	1928	0.4594
70	M	01	Schmidt, Randall, Clever	1966	0.4852
40227	M	01	Teja, Rice	1976	0.5200

Table 4. Magnitude listing of excess volume values at 298.15 K and x(1) = 0.75. The S and ET codes refer to the scatter rating and equipment type respectively.

MRL number	Codes S	ET	Authors	Year	Excess volume cc/mol
917	U	01	Ridgway, Butler	1967	0.0700
220	U	01	Williams, Ogg	1928	0.2090
10296	F	04	Letcher	1975	0.3100
1744	F	01	Harris, Dunlop	1970	0.3114
10174	F	01	Letcher	1972	0.3120
3966	F	04	Nigam, Mahl, Singh	1972	0.3179
1990	M	01	Heric, Brewer	1967	0.3290
8516	F	01	Diaz Pena, Nunez Delgado	1974	0.3329
165	F	01	Mathieson, Thynne	1956	0.3770
40227	M	01	Teja, Rice	1976	0.3900
70	M	01	Schmidt, Randall, Clever	1966	0.4110

Table 5. Codes and symbols denoting equipment type

Code	Symbol	Equipment type
01	○	Pycnometer
02	△	Mechanical oscillator densimeter
03	+	Magnetic float densimeter
04	×	Batch dilatometer
05	◇	Dilution dilatometer

scatter) column. The ET column lists the equipment type codes which are defined in Table 5.

The values from Tables 2, 3, and 4 are shown graphically in Figs. 1–3. The literature source for each point is indicated by the MRL numbers on the abscissa. The scatter rating assigned to each set of data is shown just above the data set's point. The symbol used for the point denotes the equipment type as defined in Table 5.

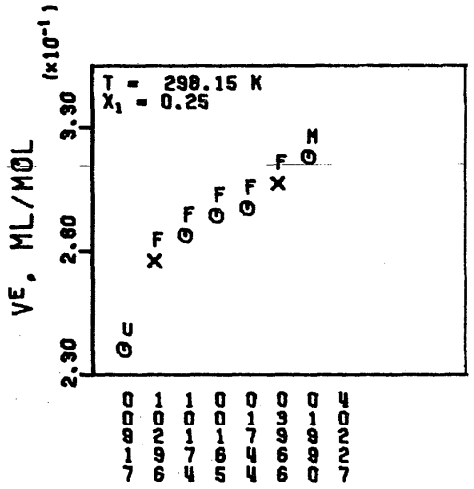

FIGURE 1. Magnitude comparison plot for V^E values at $x_1 = 0.25$ and 298.15 K.

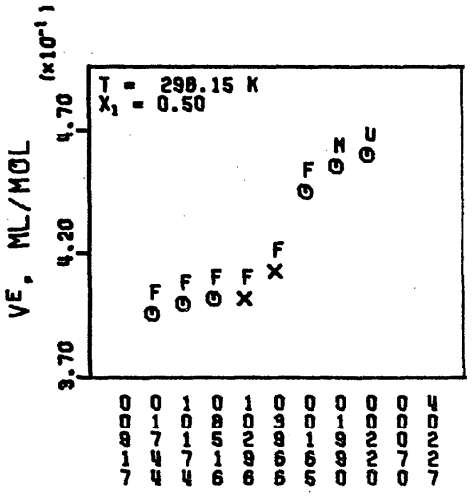

FIGURE 2. Magnitude comparison plot for V^E values at $x_1 = 0.50$ and 298.15 K.

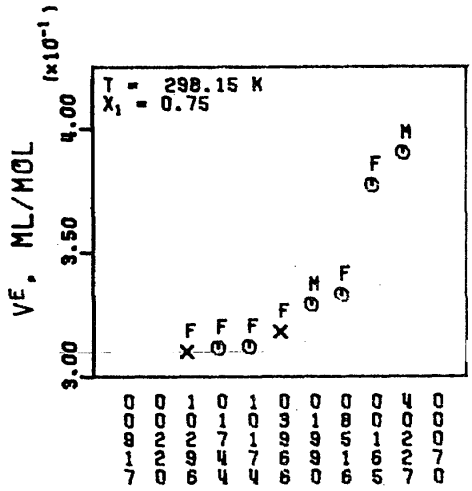

FIGURE 3. Magnitude comparison plot for V^E values at $x_1 = 0.75$ and 298.15 K.

The following V^E values were selected:

Mole fraction benzene	Selected value $cm^3\ mol^{-1}$
0.25	0.2942 ± 0.0015
0.50	0.4000 ± 0.0020
0.75	0.3160 ± 0.0016

The $\pm$ values denote a $\pm 0.5\%$ band around the selected values. The 0.2942 value at $x_1 = 0.25$ is a compromise between the MRL 10296, 10174, 165, 1744, and 3966 sets and just happens to coincide with the MRL 165 value. The MRL 10174 value was selected at $x_1 = 0.50$. The 0.3160 at $x_1 = 0.75$ is a compromise between the MRL 8516 and 3966 values on the high side and the MRL 10296, 1744, and 10174 values on the low side. None of the sets received better than a fair scatter rating; hence the need for compromise values.

Of the five data sets which received fair scatter ratings and which contributed to the choice of the selected values, three were obtained with pycnometers and two with batch dilatometers. The newer devices (mechanical oscillator densimeter, magnetic float densimeter, dilution dilatometer) have not yet been applied to the benzene + hexane system. All those sets which fell outside the ordinate ranges in Figs. 1–3 were taken with pycnometers.

4. Selected Point Values at Other Temperatures

The data sets listed in Table 1 range from 273.15 to 333.15 K in temperature. Besides 298.15, multiple measurements have been reported only at 293.15, 303.15, 308.15, 313.15, and 323.15 K. Of the three sets reported at 293.15, two received a marginal scatter rating and one an unacceptable rating. Three of the four sets at 303.15 received marginal ratings and the other an unacceptable rating. All three of the 308.15 sets and all three of the 313.15 sets received marginal ratings. One of the two 323.15 sets received a fair and the other a marginal scatter rating. Hence it was not feasible to identify selected values at any other temperature by the approach used in the previous section for the 298.15 data.

Table 6. Best curve values for the benzene(1) + hexane(2) system

		Values from best curves		
t_1, °C	T,K	x_1=0.25	x_1=0.50	x_1=0.75
0	273.15	0.3192	0.4358	0.3465
15	288.15	0.3040	0.4145	0.3279
20	293.15	0.2992	0.4078	0.3220
22	295.15	0.2970	0.4042	0.3196
25	298.15	0.2942	0.4000	0.3160
30	303.15	0.2894	0.3928	0.3103
35	308.15	0.2842	0.3856	0.3043
40	313.15	0.2794	0.3785	0.2985
50	323.15	0.2700	0.3647	0.2870
60	333.15	0.2598	0.3501	0.2751

Outside of 298.15, only three other sets received a fair scatter rating. None received a good or excellent rating. The extreme temperature sets (MRL 141 at 273.15 and MRL 10295 at 323.15 K) fell very close to a straight line through the 298.15 selected values at all three mole fractions. The MRL 3966 set at 288.15 fell above that straight line at $x_1 = 0.25$ and 0.50 but fell below it at $x_1 = 0.75$.

The best V^E versus T curves were straight lines through (or very close to) the selected 298.15 values and the MRL 141 and 10295 points at 273.15 and 323.15 K. Values read from those "best curves" at the temperature of the various sets of data are listed in Table 6. Because of the paucity of reliable data sets, those values must be considered as only tentative estimates which will undoubtedly change as additional good data accumulate.

Table 7. The best low-temperature set

SYSTEM. Benzene(1) + Hexane(2)

TEMPERATURE. 273.15 K PRESSURE. 0.1013 MPa

MOLAR VOLUMES (ml/mol). 1 = 86.787 2 = 127.283

QUALITY RATING. C SCATTER. Fair

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.25. 0.0 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.50. -0.3 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.75. 0.0 %

REFERENCE. Jackson, D. H., Young, S., Journal of the Chemical Society, London, 73, 922 (1898). (MRL 141)

x(1) mole fraction	EXCESS VOLUME ml/mol	x(1) mole fraction	EXCESS VOLUME ml/mol
0.0137	0.0329	0.3544	0.4031
0.0167	0.0167	0.3882	0.4101
0.1013	0.1505	0.4073	0.4337
0.1341	0.1956	0.4279	0.4400
0.1637	0.2321	0.4379	0.4205
0.1838	0.2344	0.4394	0.4205
0.2328	0.3067	0.4948	0.4338
0.2416	0.3028	0.5771	0.4180
0.2607	0.3352	0.6582	0.3868
0.3404	0.3896	0.7123	0.3827
0.7277	0.3662	0.8956	0.1729
0.7905	0.3107	0.9231	0.1442
0.8138	0.2929	0.9343	0.1202
0.8279	0.2723	0.9563	0.0902
0.8739	0.2084		

5. Best Data Sets

None of the benzene + hexane V^E data sets received a scatter rating better than fair and hence none received a quality rating higher than C. (The quality ratings and the relations of those ratings to the various evaluation tests have been defined in the paper evaluating the V^E data for the benzene + cyclohexane system.[1]) Consequently, none of the available data sets can be designated as recommended data sets. All that can be done is to identify those sets which are probably more reliable than the other sets.

As described in Sec. 3, the selected values at 298.15 represent reasonable "averages" of the five data sets with fair scatter ratings. Once the compromise value had been selected, three of those five data sets were reduced to a D quality rating by their percent deviations from the best curve, leav-

Table 8. One of the two best sets at 298.15

SYSTEM. Benzene(1) + Hexane(2)

TEMPERATURE. 298.15 K PRESSURE. 0.1013 MPa

MOLAR VOLUMES (ml/mol). 1 = 89.407 2 = 131.610

QUALITY RATING. C SCATTER. Fair

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.25. 1.1 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.50. -1.0 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.75. -1.5 %

REFERENCE. Harris, K. R., Dunlop, P. J., Journal of Chemical Thermodynamics, 2, 813 (1970). (MRL 1744)

x(1) mole fraction	EXCESS VOLUME ml/mol	x(1) mole fraction	EXCESS VOLUME ml/mol
0.0	0.0	0.6057	0.3853
0.1030	0.1420	0.7036	0.3451
0.2044	0.2593	0.7940	0.2763
0.2902	0.3278	0.8990	0.1595
0.4069	0.3859	1.0000	0.0
0.4958	0.3962		

Table 9. The other of the two best sets at 298.15

SYSTEM. Benzene(1) + Hexane(2)

TEMPERATURE. 298.15 K PRESSURE. 0.1013 MPa

MOLAR VOLUMES (ml/mol). 1 = 89.408 2 = 131.528

QUALITY RATING. C SCATTER. Fair

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.25. -2.7 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.50. 0.0 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.75. -1.3 %

REFERENCE. Letcher, T. M., Journal of Chemical Thermodynamics, 4, 159 (1972). (MRL 10174)

x(1) mole fraction	EXCESS VOLUME ml/mol	x(1) mole fraction	EXCESS VOLUME ml/mol
0.0662	0.0770	0.6441	0.3730
0.1781	0.2210	0.6675	0.3710
0.2845	0.3180	0.6972	0.3490
0.4317	0.3850	0.7972	0.2710
0.5434	0.3950	0.8973	0.1680
0.5613	0.3990	0.9621	0.0670

Table 10. The best high-temperature set

SYSTEM. Benzene(1) + Hexane(2)

TEMPERATURE. 323.15 K PRESSURE. 0.1013 MPa

MOLAR VOLUMES (ml/mol). 1 = 89.407 2 = 131.610

QUALITY RATING. C SCATTER. Fair

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.25. 0.0 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.50. -0.5 %

DEVIATION FROM VE VS. T CURVE AT x(1) = 0.75. 0.0 %

REFERENCE. Diaz Pena, M., Nunez Delgado, J., Journal of Chemical Thermodynamics, 7, 201 (1975). (MRL 10295)

x(1) mole fraction	EXCESS VOLUME ml/mol	x(1) mole fraction	EXCESS VOLUME ml/mol
0.1980	0.2300	0.5260	0.3650
0.3170	0.3090	0.5530	0.3590
0.3300	0.3170	0.5820	0.3600
0.4250	0.3550	0.5960	0.3540
0.4700	0.3620	0.6890	0.3190
0.4810	0.3540	0.8160	0.236
0.4970	0.3630		

ing only the MRL 1744 and 10174 sets with C quality ratings at 298.15 K. There is no decisive reason for favoring one of those two sets over the other.

The other two C sets in Table 1 are the MRL 141 (at

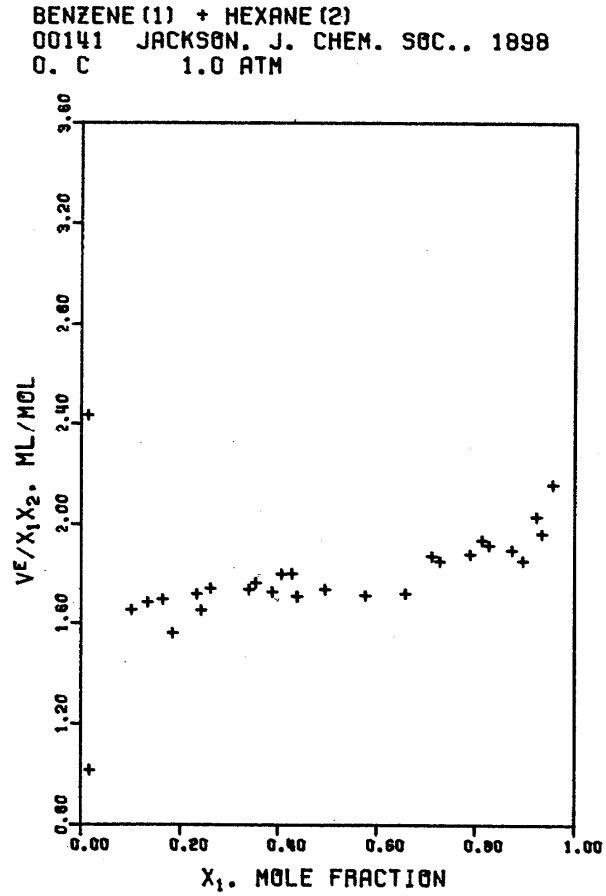

FIGURE 4. The best low-temperature set. 273.15 K. Jackson and Young 1898. MRL 141.

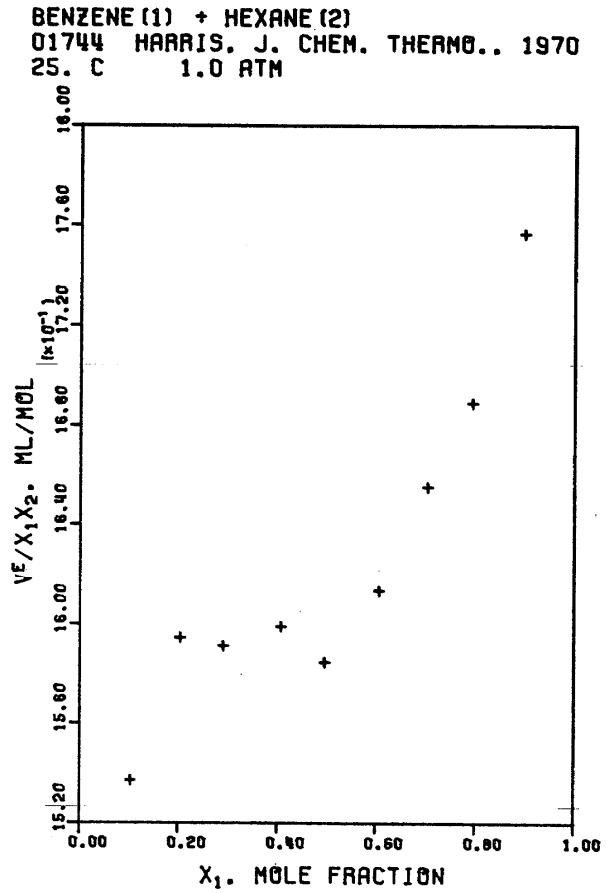

FIGURE 5. One of the two best sets at 298.15 K. Harris and Dunlop, 1970. MRL 1744.

273.15 K) and the 10295 (at 323.15 K) sets. Other than their C ratings, they are recommended by the fact that they fall on or close to a straight line through the selected values at 298.15 K.

The data sets with C quality ratings are tabulated in Tables 7–10, and plotted in Figs. 4–7.

6. Experimental Measurements Needed

Reliable V^E measurements are needed at all temperatures, including 298.15 K. However, the benzene + cyclohexane system is already established as a test system for V^E measurements and there is no need to accumulate a similar large amount of 298.15 data for benzene + hexane. What are needed are A or B quality data sets at various temperatures over as wide a temperature range as the available experimental devices can support.

7. Pure Compound Densities

Pure compound volume values are needed to calculate V^E values from

$$V^E = V - x_1V_1 - x_2V_2, \quad (1)$$

when mixture density or volume values are reported. The authors' pure compound values are always used if they are given. If not, the computer program VECON which converts

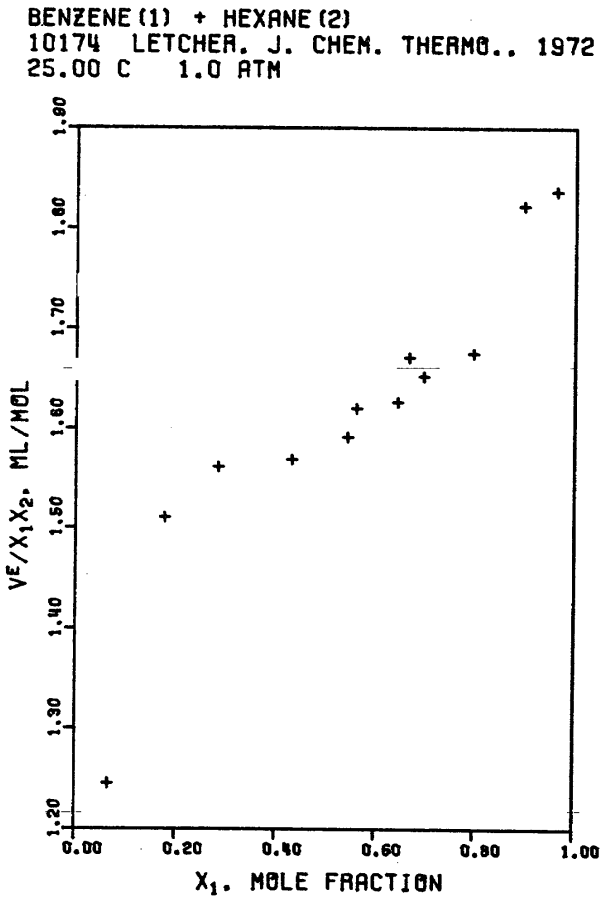

FIGURE 6. The other of the two best sets at 298.15 K. Letcher, 1972. MRL 10174.

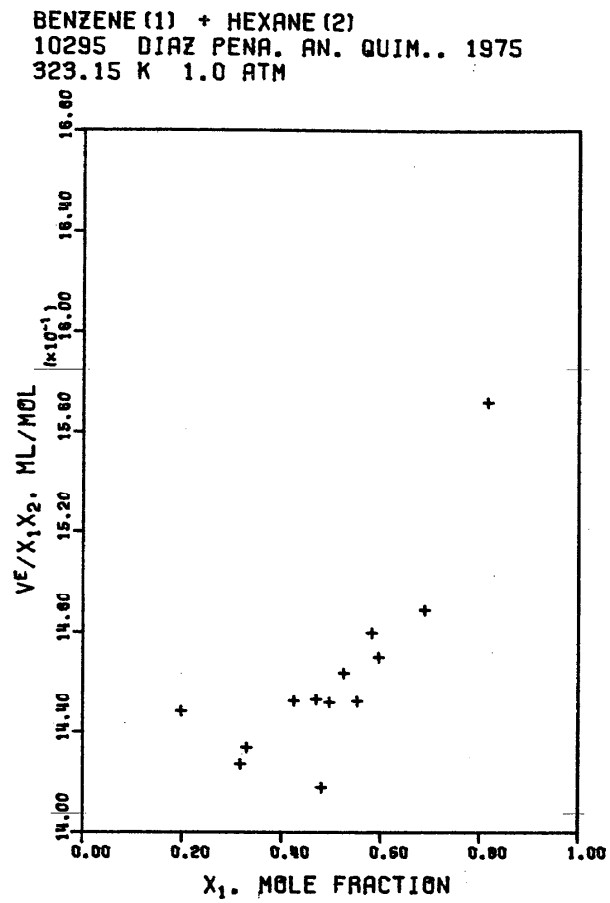

FIGURE 7. The best high-temperature set. 323.15 K. Diaz Pena and Nunez Delgado, 1975. MRL 10295.

the mixture or density data to V^E values accesses the pure compound data bank CDATA1 for the needed pure compound values. The general procedures used in selecting and correlating the pure compound liquid density data and storing it in CDATA1 have been described in a preceding paper.[4]

The pure compound volume values used for each set of data appear on the tabulation for that set. (See Tables 7–10 for examples.) When obtained from CDATA1, the values were calculated from a Francis equation correlation using the constants given in Table 11. The benzene correlation shown there is based on a fit of 279 selected experimental data points with a RMSD of 0.00018 g cm^3. The hexane correlation fitted 110 selected data points with a RMSD of 0.00019.

The total experimental data base for the benzene liquid density correlations in CDATA1 came from 73 primary literature documents each of which contributed one or more data points. The data points reported by 19 other primary literature sources were totally excluded from the correlation. The analogous numbers for hexane are 28 and 8.

8. Data Set Tabulations

Tabulations of all the data sets covered in this paper can be obtained from the Director, Thermodynamics Research Laboratory, Box 1144, Washington University, St. Louis, Missouri 63130. Copies of the V^E tables with the V^E/x_1x_2 versus x_1 plots on the back of the pages will be provided for $0.50 per set of data plus $5.00 for handling charges. An invoice will be mailed with the tables.

The tables will be provided only in a complete set for a given system, i.e., requests for tables for individual sets will not be processed.

9. Bibliography

Table 12 is the bibliography for excess volume data for the benzene(1) + hexane(2) system. The identifying number for each citation is the Laboratory's Master Reference List (MRL) number. The MRL numbers relate the citations in Table 12 to the various tables and figures used in this paper.

Some of the documents listed in Table 12 may report V^E data in a form which did not permit transcription and

Table 11. Pure compound density data[a]

Constant	Benzene (280 to 374 K)	Hexane (183 to 354 K)
A	0.11971445D+01	0.91393524D+00
B	0.96949888D-03	0.77406992D-03
C	0.11815362D+02	0.60000496D+01
E	0.64129077D+03	0.50989258D+03

[a]Density, g/ml = A - BT - C/(E-T)

processing. In most such cases, the data were presented only in graphical form. Those documents have been included in the bibliography in order to provide a complete coverage of the V^E literature for the benzene + hexane system.

TABLE 12. Bibliography for excess volume data

70. R.L. Schmidt, J. C. Randall, and H. L. Clever, J. Phys. Chem. **70**, 3912 (1966).
141. D.H. Jackson and S. Young, J. Chem. Soc., London **73**, 922 (1898).
165. A. R. Mathieson and J. C. J. Thynne, J. Chem. Soc., London, 3708 (1956).
171. H. Mohler, Helv. Chim. Acta **20**, 1447 (1937).
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10. Nomenclature

MRL	Acronym for Master Reference List.
RMSD	Acronym for root-mean-square deviation.
V	Liquid molar volume of mixture.
V_i	Liquid molar volume of component i.
V^E	Excess volume. Defined by Eq. (1).
x	Liquid mole fraction.

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