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Gas–Liquid Equilibrium for the Helium–Ethylene and Helium–Propylene Systems below 260 K and 120 Atm

James D. Garber and Waldemar T. Ziegler*

School of Chemical Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332

Gas–liquid phase equilibrium measurements are reported for the helium–ethylene and helium–propylene systems for a series of isotherms below 260 K up to pressures of 120 atm. Gas-phase compositions expressed in terms of the enhancement factor have been computed for several models by using thermodynamic relations and have been compared with the experimental results. The interaction parameter K_{12} has been computed from experimental values of the second virial interaction coefficient.

This work is a continuation of phase equilibrium studies made in this laboratory on binary systems involving helium and hydrogen gases as one component of the system. The present work is concerned with gas–liquid phase equilibrium measurements for the helium–ethylene and helium–propylene systems (*1*) below 260 K and up to pressures of approximately 120 atm. Other helium systems which have been studied by using techniques similar to those used in this work are He–Ar (*2*, *3*), He–CO_2 (*4*), and He–CF_4 and He–$CClF_3$ (*5*–*7*).

In all instances the experimental results have been compared with gas-phase equilibrium values computed from various models by appropriate thermodynamic relations.

Experimental Section

All phase equilibrium measurements have been made by using a single-pass flow-type apparatus described by Kirk (*8*) and Kirk and Ziegler (*9*). In this apparatus which consists of a thermostated copper equilibrium cell in which the pure liquid condensable component (in this instance ethylene or propylene) can be condensed, after which the pure helium gas is bubbled through the liquid until the composition of the exit helium gas mixture becomes constant at which time the liquid phase is sampled to determine its composition. The detailed operating procedures used were similar to those described by Kirk and Ziegler (*9*). Equilibrium was considered to have been reached when three successive gas-phase samples taken at approximately 10-min intervals showed the same composition within the experimental uncertainty of the measurements (approximately ±3% of the determined composition in mole fraction). When this state was reached for a fixed pressure and temperature of the equilibrium cell, liquid samples were taken from the cell and analyzed for helium.

The composition analyses were made by using two separate gas chromatographs (Perkin-Elmer 154 vapor fractometers). Analyses of the gas-phase samples to determine the hydrocarbon content made use of an activated silica gel column for ethylene and a dimethylsulforlane on fire-brick column for propylene with helium as the carrier gas. The liquid phase was analyzed for helium by using a molecular sieve column (Linde 5A) with argon as the carrier gas. The chromatographs were calibrated on a peak height basis as described by Kirk (*8*, *9*). The experimental phase equilibrium measurements made consisted of five or six isotherms at five to six different pressures in the range up to approximately 120 atm. The experimental results are given for the helium–ethylene system in Table I and for the helium–propylene system in Table II. The temperature of the equilibrium cell could be controlled to approximately ±0.03 K. All temperatures were measured with a capsule-type platinum resistance thermometer calibrated by the U.S. National Bureau of Standards on the International Practical Temperature Scale of 1948 (IPTS-48). All temperature in Tables I and II are given on the International Practical Temperature Scale of 1968 (IPTS-68), the conversion of Barber (*10*) having been used to convert the IPTS-48 to IPTS-68.

The two Bourdon gages used to measure the pressures (one for lower pressures and the second for the higher pressures) have been calibrated by Kirk (*8*, *9*). The calibration was verified by measuring the vapor pressures of pure argon and carbon dioxide. The uncertainty in the pressure reported in Tables I and II is estimated to be $\pm^1/_2$% of the indicated pressure.

From a consideration of the uncertainty in the temperature and pressure measurements and the scatter of the data in preparing the calibration curves for the chromatographs, the overall uncertainty of the gas-phase composition for the helium–ethylene system is estimated to be $\pm 4^1/_2$% of the reported values for the 129.98 and 150.00 K isotherms and ±3% of the quoted values for the higher temperature isotherms. For the helium–propylene gas-phase data y_1 is estimated to be uncertain to $\pm 4^1/_2$% for the 200.00 and 212.49 K isotherms and ±3% for the higher temperature isotherms.

One of the questions which arises with the use of the flow-through type of cell is whether or not equilibrium has been reached, as contrasted with a steady state which may or may not represent a true equilibrium state. In the present experi-

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Table I. Experimental Values of Equilibrium Gas- and Liquid-Phase Compositions for the Ethylene (1)-Helium (2) System

T, K	P, atm	P_{01}, atm	gas phase $100y_1$ mol % ethylene	liquid phase $100x_2$ mol % helium	$\phi_1 = Py_1/P_{01}$
129.98	119.65	0.04348	0.0453	0.1745	1.247
	100.05		0.0521	0.1471	1.199
	80.45		0.0639	0.1284	1.183
	63.16		0.0770		1.118
	60.28			0.0948	
	41.93		0.1090	0.0652	1.059
	25.35		0.2010	0.0401	1.033
150.01	119.24	0.2697	0.253	0.3887	1.117
	99.91		0.293	0.3303	1.086
	99.78			0.3272	
	80.59		0.360	0.2671	1.077
	60.24		0.477		1.066
	42.07		0.683	0.1493	1.065
	22.95		1.241	0.0805	1.056
162.00	119.10	0.6320	0.588	0.5877	1.108
	99.61		0.699	0.5063	1.102
	81.27		0.850	0.4145	1.093
	60.38		1.133	0.3091	1.082
	42.01		1.605	0.2205	1.067
	22.91		1.957	0.1181	1.072
173.99	118.77	1.3035	1.239	0.8572	1.129
	99.71		1.446	0.7235	1.106
	80.66		1.778	0.5903	1.100
	60.24		2.363	0.4506	1.092
	41.87		3.390	0.3081	1.089
	22.92		6.040	0.1692	1.062
188.02	119.17	2.6637	2.593	1.231	1.160
	99.51		2.987	1.065	1.116
	80.72		3.726	0.8662	1.129
	60.24		4.952	0.6446	1.120
	44.35		6.553	0.4545	1.091
	44.20		6.659	0.4516	1.105
	22.80		12.82	0.2469	1.097
202.01	119.51	4.8793		1.725	
	119.30		4.904		1.199
	99.64		5.842	1.447	1.193
	81.13		7.000		1.164
	60.13		9.424	0.8969	1.161
	42.58		13.17	0.6118	1.148
	24.61		22.07	0.3171	1.113
216.04	119.09	8.2378	8.633		1.248
	89.77		10.21	1.945	1.236
	83.11		12.19		1.230
	60.25		16.23	1.174	1.187
	42.29		22.89	0.8153	1.175

Table II. Experimental Values of Equilibrium Gas- and Liquid-Phase Compositions for the Propylene (1)-Helium (2) System

T, K	P, atm	P_{01}, atm	gas phase $100y_1$ mol % propylene	liquid phase $100x_2$ mol % helium	$\phi_1 = Py_1/P_{01}$
175.00	118.83		*a*	0.4284	
	118.62			0.4238	
	100.39			0.3664	
	80.38			0.2918	
	60.04			0.2325	
	42.21			0.1615	
	22.81			0.0876	
187.50	117.40		*a*	0.5961	
	99.16			0.5027	
	83.51			0.4317	
	59.97			0.3122	
	41.33			0.2232	
	23.10			0.1222	
200.00	118.42	0.26509	0.2300	0.8162	1.027
	99.50		0.2805	0.6858	1.053
	80.79		0.3468	0.5585	1.057
	60.24		0.4497	0.4310	1.022
	41.67		0.6438	0.2960	1.012
	23.03		1.180	0.1692	1.025
212.49	118.22	0.53216	0.4677	1.047	1.039
	100.46		0.5493	0.9216	1.037
	80.59		0.6848	0.7454	1.037
	60.24		0.9187	0.5560	1.040
	41.05		1.343	0.3858	1.036
	23.45		2.392	0.2250	1.054
224.99	118.69	0.9729	0.8722	1.365	1.064
	100.86		1.027	1.165	1.065
	80.40		1.272	0.976	1.064
	60.65		1.716	0.727	1.066
	40.98		2.528	0.481	1.065
	25.47		4.060	0.295	1.065
254.98	118.43	3.2232	3.113	2.259	1.144
	101.20		3.644	2.010	1.144
	80.32		4.570	1.596	1.139
	60.45			0.7270	
	60.65		5.979	1.182	1.125
	41.66		8.573	0.8197	1.108
	25.27		13.84	0.4651	1.085
239.99	119.92	1.8535	1.706	1.838	1.104
	100.86		2.026	1.540	1.102
	80.59		2.541	1.232	1.105
	60.25		3.390	0.9513	1.102
	41.13		4.935	0.6315	1.095
	26.09		7.602	0.3900	1.070

[a] Analysis discarded because of unexplained systematic error.

ments the flow rate of helium gas through the cell was approximately the same as was used by Kirk (*9*) in his studies of the H_2–CH_4 system. Under these conditions the residence time of helium in the cell is about 25 min. For the helium-ethylene system four experimental points were measured in the temperature range 150–200 K and the pressure range 42.3–119.3 atm in which the flow rate of the helium was 200 cm^3/min (that is, twice the flow rate used in the experiments reported in Table I). The equilibrium gas- and liquid-phase compositions found for these two flow rates agreed within the estimated uncertainty of the measurements (±3%). A similar test was made for the helium-propylene system along the 175.00 K isotherm at 119 and 23 atm. These results also agreed within estimated experimental uncertainty of ±3% with measurements made at the lower flow rate of 100 cm^3/h. From these studies it was concluded that the experimental phase compositions reported in Tables I and II are equilibrium values.

The helium used in this work was obtained from the U.S. Bureau of Mines and had a quoted purity of 99.997%. Ethylene and propylene were obtained from the Phillips Petroleum Co. and had a stated purity of 99.97% and 99.99%, respectively. All these gases were used without further purification.

Results

Gas–liquid phase equilibrium measurements were made for a series of pressures from 20 to 120 atm for seven isotherms at 129.98, 150.01, 162.00, 179.99, 188.02, 202.01, and 216.04 K for the helium–ethylene system and at 175.00, 187.49, 200.00, 212.49, 224.99, 239.99, and 254.98 K for the helium–propylene system. Gas-phase compositions along the two isotherms at 175.00 and 187.50 K were discarded because they appeared to contain a systematic error. (For discussion of this point see Garber (*1*).)

The experimental results are shown in Tables I and II. In these tables each gas-phase composition is an average of three successive measurements of the equilibrium gas stream. The liquid-phase measurements represent the average of replicate measurements of a single liquid sample.

The vapor pressures (P_{01}) of the pure liquid hydrocarbons are shown in Tables I and II for each temperature. For ethylene all values of P_{01} below 1 atm were obtained by linear inter-

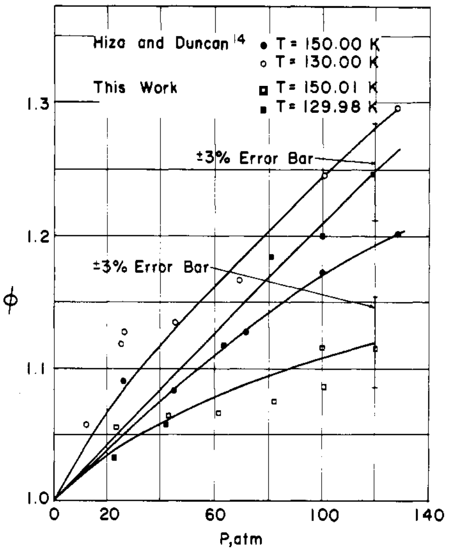

Figure 1. Experimental enhancement factor for ethylene in the helium-ethylene system at 130 and 150 K.

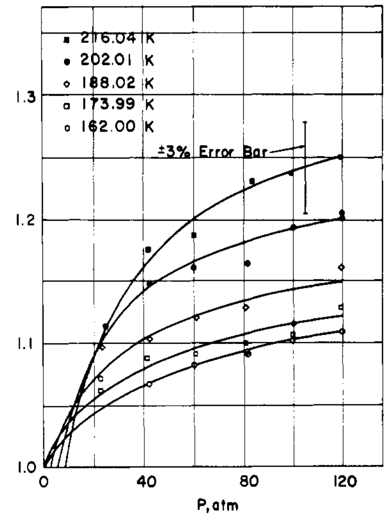

Figure 2. Experimental enhancement factor for ethylene in the helium-ethylene system at 162.00, 173.99, 188.02, 202.01, and 216.04 K.

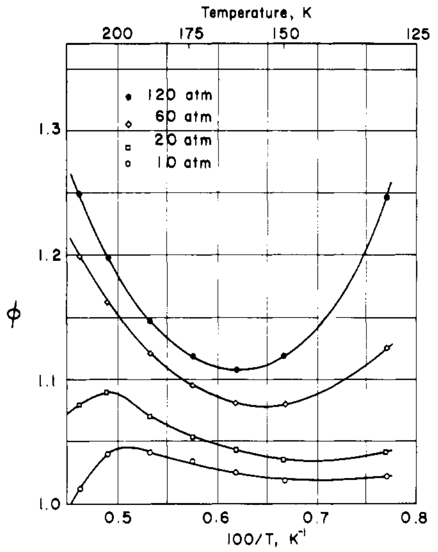

Figure 3. Smoothed experimental enhancement factors of ethylene in helium at constant pressures (10, 20, 60, and 120 atm).

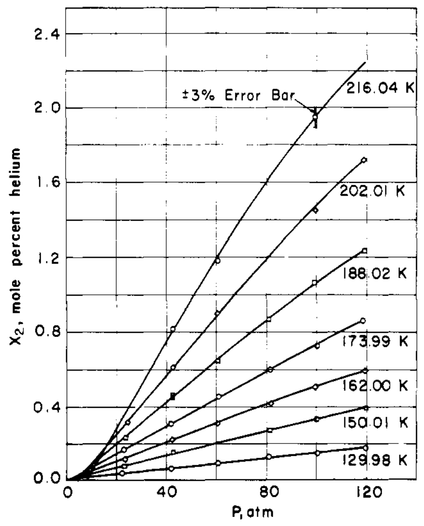

Figure 4. Experimental solubility of helium in liquid ethylene.

polation of the tabular values given by Ziegler et al. (*11*) and above this pressure the values of Michels and Wassenaar (*12*) were used. The vapor pressures of propylene were computed by using the following Antoine equation taken from Rossini (*13*).

$$\log P(\text{torr}) = 6.81960 - \frac{785.00}{T(^\circ\text{C}) + 247.00}$$

Discussion of Results

Hiza and Duncan (*14*) have reported equilibrium gas-phase compositions for the ethylene-helium system over the range 91-150 K and at pressures up to about 130 atm and have reported the enhancement factors $\phi = py_i/P_{0i}$ for the isotherms at 130.00 and 150.00 K as a function of pressure. Their results for these isotherms are compared with results obtained in the present work at 129.98 and 150.00 K in Figure 1. Hiza and Duncan estimated the accuracy of their gas-phase ethylene measurements to be ±2% except at the lowest concentration where they estimated the accuracy to be probably ±3%. Examination of Figure 1 shows that the compositions reported by us (with an estimated uncertainty of ±3%) agree with the results of Hiza and Duncan (*14*) to within the estimated uncertainty of the two sets of data. Figure 1 also shows the utility of using the enhancement factor as an empirical parameter for representing experimental gas-phase equilibrium data since the enhancement factor is expected to vary smoothly with pressure. Experimental enhancement factors of ethylene in the gas phase of the helium-ethylene system at higher temperatures are shown in Figure 2. No other data appear to have been published in this temperature region.

The experimental curves shown in Figures 1 and 2 were used to derive a set of smoothed enhancement factors for ethylene in the helium-ethylene systems at 10, 20, 60, and 120 atm. These smoothed values of ϕ_1 are plotted in Figure 3 vs. $1/T$. This set of curves shows an interesting set of maxima and minima. Hiza and Duncan (*14*) have suggested that such a minimum might exist in this system.

The experimental liquid-phase data for the helium-ethylene system as a function of temperature and pressure are shown in Figure 4. The reverse solubility (increase of solubility of a gas in a liquid with increasing temperature) characteristic of compressed gas-liquid systems involving helium is apparent.

Experimental gas-liquid phase equilibrium data for the helium-propylene system are shown in Table II and have been plotted in Figure 5. No other experimental data for this system appear to have been published. Smoothed values of the enhancement factor at 10, 20, 60, and 120 atm have been taken from these curves and the curves of Figure 6 constructed. The

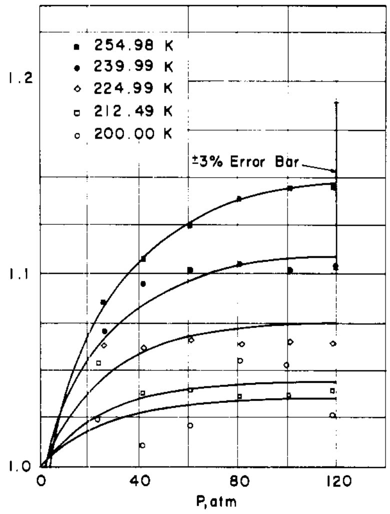

Figure 5. Experimental enhancement factors of propylene in helium at 200.00, 212.49, 224.99, 239.99, and 254.98 K.

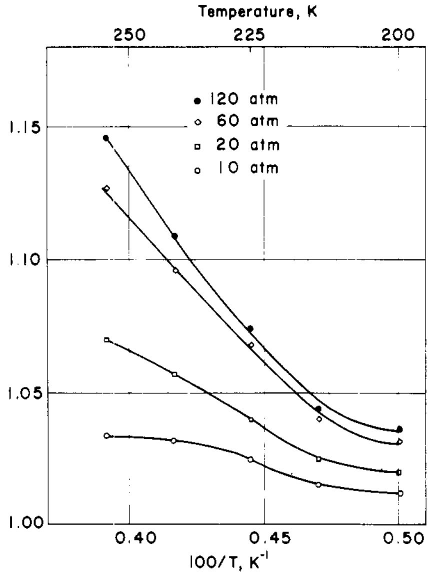

Figure 6. Smoothed experimental enhancement factors of propylene in helium at constant pressures (10, 20, 60, and 120 atm).

general shape of the curves of Figure 6 is similar to those of the helium–ethylene system shown in Figure 3. The solubility of helium in liquid propylene as a function of pressure at various temperatures is shown in Figure 7.

Comparison of Experimental Results with Theoretical Models

The prediction of gas–liquid phase equilibria such as those determined in this work from thermodynamic considerations has been discussed by numerous investigators. Kirk, Ziegler, and Mullins (*15*) have discussed such calculations for the CH_4 (1)–H_2 (2) system, for instance, under conditions where the condensed phase is either a pure solid or a pure liquid. These discussions have often made use of a quantity, ϕ_i, designated as the enhancement factor and referring generally to the condensable component of such binary systems. The enhancement factor is defined by the relations

$$\phi_i \equiv Py_i/P_{01} \equiv y_i/y_i^\circ \qquad (1)$$

where y_i is the gas-phase composition (in mole fraction) of the condensable component i at the phase equilibrium P and T and P_{01} is the vapor pressure of the pure condensable phase at T. If one assumes the gas phase at P, T to be an ideal gas mixture,

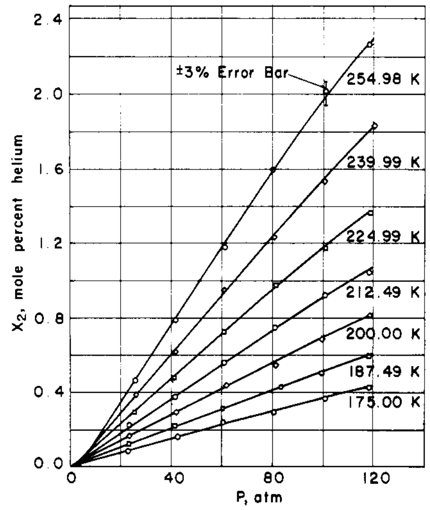

Figure 7. Experimental solubility of helium in liquid propylene.

then y_i° is the mole fraction which one would expect if the partial pressure of i in the gas phase were P_{0i}. Both forms of ϕ_i given in eq 1 have been used to describe such binary gas-phase equilibria (see for instance Dokoupil et al. (*16*) and Rowlinson and Richardson (*17*)). The quantity ϕ_i is called the enhancement factor because it is almost always greater than unity. Thermodynamic expressions for ϕ_i can be developed from the thermodynamic requirement that the chemical potential of a component i must be equal in both the gas and the condensed phase at equilibrium (or, alternately, that the fugacities of the components must be equal).

The condition of equality of chemical potential or fugacity for the condensable component (designated as component 1) in the two phases leads to the following exact thermodynamic expression for ϕ_i (see also ref 9, 15, and 18).

$$\ln \phi_1 = \ln \frac{PV_m}{P_{01}V_{01}} + \frac{1}{RT}\int_{P_{01}}^{P} V_1^c\, dP + \frac{1}{RT}\int_{V_{01}}^{\infty}\left[\left(\frac{\partial P}{\partial n_1}\right)_{V,T} - \frac{RT}{V}\right] dV - \frac{1}{RT}\int_{V_m}^{\infty}\left[\left(\frac{\partial P}{\partial n_1}\right)_{V,T,n_2} - \frac{RT}{V}\right] dV + \ln \gamma_1 x_1 \qquad (2)$$

The calculation of ϕ_i from eq 2 requires that the properties (vapor pressure and equation of state for the pure condensed phase and the saturated vapor of component 1) be known, that an equation of state for the gas-phase mixture be known in order to evaluate the last integral, and finally that the solubility x_1 and the activity coefficient (γ_1) of component 1 in the condensed phase be known or some reasonable assumptions be made concerning the condensed phase solution.

If the virial equation of state in the form

$$\frac{PV_m}{RT} = 1 + \frac{B_m}{V_m} + \frac{C_m}{V_m^2} \qquad (3)$$

with

$$B_m = y_1^2 B_{11} + 2y_1y_2B_{12} + y_2^2 B_{22} \qquad (4)$$

and

$$C_m = y_1^3 C_{111} + 3y_1^2 y_2 C_{112} + 3y_1 y_2^2 C_{122} + y_2^3 C_{222} \qquad (5)$$

Table III. Smoothed Experimental and Theoretical Enhancement Factors of Ethylene in Helium and the Smoothed Experimental Solubility of Helium in Liquid Ethylene

P, atm	ϕ EXP	ϕ LJCL	ϕ KIH	ϕ KIHCK12	ϕ KIHEK12	ϕ BWR	$100x_2$, mol %
91.00 K (Hiza and Duncan[14])							
10	1.100	1.113	1.138	1.037	1.027	1.084	0.0[a]
20	1.173	1.229	1.289	1.074	1.053	1.176	0.0
30	1.238	1.349	1.452	1.110	1.079	1.272	0.0
40	1.296	1.471	1.628	1.146	1.103	1.368	0.0
50	1.348	1.596	1.817	1.180	1.127	1.473	0.0
60	1.395	1.721	2.020	1.215	1.150	1.574	0.0
95.00 K (Hiza and Duncan[14])							
20	1.147	1.197	1.258	1.066	1.047	1.154	0.0[a]
40	1.267	1.399	1.555	1.130	1.092	1.317	0.0
60	1.370	1.602	1.891	1.919	1.134	1.490	0.0
80	1.460	1.804	2.270	1.251	1.174	1.673	0.0
100	1.542	2.001	2.692	1.309	1.212	1.865	0.0
120	1.617	2.192	3.159	1.366	1.249	2.065	0.0
140	1.684	2.374	3.673	1.423	1.285	2.272	0.0
102.00 K (Hiza and Duncan[14])							
20	1.118	1.152	1.214	1.055	1.039	1.123	0.0[a]
40	1.217	1.301	1.454	1.108	1.076	1.250	0.0
60	1.302	1.446	1.719	1.158	1.110	1.382	0.0
80	1.378	1.585	2.010	1.207	1.143	1.518	0.0
100	1.447	1.716	2.328	1.255	1.174	1.659	0.0
120	1.510	1.838	2.674	1.301	1.205	1.803	0.0
140	1.567	1.951	3.048	1.347	1.234	1.950	0.0
112.00 K (Hiza and Duncan[14])							
20	1.080	1.116	1.180	1.053	1.040	1.102	0.0[a]
40	1.157	1.227	1.377	1.105	1.079	1.207	0.0
60	1.231	1.333	1.592	1.155	1.116	1.314	0.0
80	1.302	1.432	1.825	1.204	1.152	1.424	0.0
100	1.368	1.523	2.077	1.253	1.187	1.537	0.0
120	1.432	1.606	2.348	1.301	1.222	1.652	0.0
140	1.488	1.682	2.638	1.349	1.257	1.770	0.0
122.00 K (Hiza and Duncan[14])							
20	1.062	1.085	1.148	1.046	1.035	1.083	0.0[a]
40	1.117	1.163	1.306	1.089	1.068	1.164	0.0
60	1.168	1.234	1.475	1.130	1.099	1.247	0.0
80	1.216	1.297	1.655	1.172	1.129	1.330	0.0
100	1.260	1.354	1.846	1.212	1.159	1.414	0.0
120	1.302	1.404	2.048	1.252	1.189	1.499	0.0
140	1.342	1.320	2.263	1.293	1.219	1.584	0.0
130.00 K (Hiza and Duncan[14])							
20	1.065	1.068	1.130	1.042	1.033	1.073	0.032[b]
40	1.118	1.125	1.264	1.079	1.061	1.140	0.062
60	1.164	1.177	1.407	1.116	1.089	1.209	0.093
80	1.206	1.233	1.556	1.152	1.116	1.276	0.122
100	1.243	1.261	1.714	1.187	1.142	1.345	0.151
120	1.281	1.294	1.880	1.223	1.169	1.413	0.178
140	1.315	1.320	2.054	1.257	1.195	1.482	0.203
150.00 K (Hiza and Duncan[14])							
20	1.038	1.047	1.106	1.044	1.037	1.064	0.069[b]
40	1.076	1.076	1.205	1.074	1.061	1.112	0.139
60	1.110	1.098	1.307	1.103	1.084	1.160	0.206
80	1.142	1.116	1.413	1.131	1.106	1.207	0.270
100	1.170	1.129	1.523	1.159	1.127	1.253	0.331
120	1.193	1.138	1.637	1.187	1.149	1.298	0.390
140	1.211	1.143	1.754	1.216	1.171	1.343	0.442
129.98 K (This Work)							
20	1.042	1.068	1.130	1.042	1.033	1.073	0.032
40	1.084	1.125	1.264	1.079	1.061	1.140	0.062
60	1.126	1.177	1.407	1.116	1.089	1.209	0.093
80	1.167	1.223	1.557	1.153	1.116	1.276	0.122
100	1.208	1.261	1.714	1.187	1.142	1.345	0.151
120	1.247	1.293	1.881	1.223	1.169	1.413	0.178
150.01 K (This Work)							
20	1.036	1.047	1.106	1.044	1.037	1.064	0.069
40	1.060	1.076	1.205	1.074	1.061	1.112	0.139
60	1.080	1.098	1.307	1.103	1.084	1.160	0.206
80	1.096	1.116	1.413	1.131	1.106	1.207	0.270
100	1.109	1.129	1.523	1.159	1.127	1.253	0.331
120	1.120	1.138	1.637	1.187	1.149	1.298	0.389

Table III (*Continued*)

P, atm	ϕ EXP	ϕ LJCL	ϕ KIH	ϕ KIHCK12	ϕ KIHEK12	ϕ BWR	$100x_2$, mol %
			162.00 K (This Work)				
20	1.044	1.048	1.105	1.053	1.048	1.069	0.102
40	1.067	1.064	1.191	1.081	1.070	1.111	0.209
60	1.082	1.078	1.279	1.107	1.091	1.151	0.308
80	1.094	1.086	1.370	1.133	1.111	1.191	0.407
100	1.102	1.091	1.465	1.159	1.132	1.231	0.501
120	1.108	1.092	1.562	1.185	1.152	1.268	0.589
			173.99 K (This Work)				
20	1.054	1.056	1.110	1.067	1.063	1.080	0.147
40	1.080	1.067	1.188	1.095	1.086	1.119	0.297
60	1.096	1.072	1.268	1.120	1.106	1.155	0.445
80	1.107	1.073	1.351	1.145	1.126	1.190	0.591
100	1.115	1.070	1.434	1.170	1.147	1.224	0.731
120	1.120	1.065	1.522	1.195	1.166	1.257	0.862
			188.02 K (This Work)				
20	1.071	1.077	1.122	1.090	1.086	1.102	0.200
40	1.103	1.082	1.199	1.120	1.112	1.142	0.426
60	1.121	1.081	1.274	1.147	1.135	1.176	0.646
80	1.133	1.075	1.350	1.173	1.156	1.209	0.857
100	1.142	1.066	1.430	1.197	1.176	1.240	1.061
120	1.148	1.055	1.510	1.222	1.197	1.270	1.246
			202.01 K (This Work)				
20	1.090	1.100	1.135	1.112	1.110	1.120	0.250
40	1.139	1.110	1.220	1.154	1.148	1.170	0.569
60	1.163	1.104	1.297	1.186	1.177	1.208	0.883
80	1.179	1.093	1.375	1.215	1.200	1.241	1.186
100	1.190	1.079	1.454	1.242	1.224	1.271	1.472
120	1.198	1.062	1.536	1.270	1.247	1.300	1.739
			216.04 (This Work)				
20	1.080	1.113	1.134	1.122	1.121	1.127	0.282
40	1.159	1.143	1.245	1.194	1.189	1.204	0.742
60	1.199	1.139	1.335	1.238	1.229	1.251	1.180
80	1.221	1.135	1.423	1.276	1.264	1.290	1.587
100	1.237	1.122	1.513	1.312	1.294	1.325	1.954
120	1.249	1.086	1.608	1.346	1.324	1.357	2.250

[a] Condensed phase assumed by us to be pure ethylene. Liquid-phase compositon not determined by Hiza and Duncan. [b] Liquid composition values taken from present work.

is used to represent the gas phase and the pure condensable vapor, then eq 2 for ϕ_1 can be written in the form

$$\ln \phi_1 = \frac{1}{RT}\int_{P_{01}}^{P} V_1^{c}\,dP + 2\frac{B_{11}}{V_{01}} + \frac{3C_{111}}{2V_{01}^2} - \ln\frac{P_{01}V_{01}}{RT} - \frac{2}{V_m}(y_1B_{11} + y_2B_{12}) - \frac{3}{2V_m^2}(y_1^2C_{111} + 2y_1y_2C_{112} + y_2^2C_{122}) + \ln\frac{PV_m}{RT} + \ln\gamma_1x_1 \quad (6)$$

where V_m is the molar volume of the gas-phase mixture at equilibrium P and T as computed from eq 3, V_1^c is the molar volume of the pure condensed component and V_{01} represents the molar volume of pure saturated condensable vapor at T and P_{01}. When the condensed phase is solid, the assumption is often made that the equilibrium condensed phase is a pure solid phase (*15*, *18*). In this case the term $\ln \gamma_1x_1$ is zero. When the condensed phase is a liquid and the solubility of component 2 is expected to be small, the assumption that the liquid phase may be treated as a pure liquid phase (*15*) or an ideal solution (*3*) may be made in order to permit predictive calculation of ϕ_1 to be made from eq 2 or 6.

In the present work ϕ_1 has been calculated from eq 2 for a number of different gas-phase models (assumed equation of state for the gas-phase mixture). The liquid phase has been assumed to be an ideal solution ($\gamma_1 = 1$) and x_1 has been used from the experimental data. Becase the solubility of helium in the condensed liquid phase is usually quite small ($x_2 \leq 0.024$) in the present systems, the contribution of the term involving γ_1x_1 is quite small. The exact form of eq 2 used for each model will not be given but may be inferred from eq 2 and 3 or from expressions given for $\ln \phi_1$ by Kirk et al. (*15*) or Smith et al. (*18*). The present calculations are discussed in detail by Garber (*1*).

A comparison between the enhancement factor ϕ computed by using these different models for the gas phase and graphically smoothed experimental enhancement factors at various pressures is given in Table III for the helium–ethylene system and in Table IV for the helium–propylene system.

Lennard–Jones (6–12) Model (LJCL). In Tables III and IV the designation LJCL implies that the enhancement factor ϕ_1 has been computed by using eq 6 and that the virial coefficients B_{11}, B_{12}, and B_{22} and C_{111}, C_{112}, C_{122}, and C_{222} have been evaluated by using the classical Lennard-Jones (6–12) parameters for helium (component 2) and the hydrocarbons given in Table V and the methods described in Hirschfelder et al. (*19*) have been used except that the coefficients $C^{(j)}$ used in the evaluation of the third virial coefficients C_{ijk} made use of the first 21 values of $C^{(j)}$ coefficients given by Rowlinson et al. (*20*). The Lennard-Jones (6–12) parameters used in the LJCL model calculations are given in Table V.

Kihara Model (KIH). A second model used to calculate the virial coefficients B_{11}, B_{22}, and B_{12} in eq 6 made use of the Kihara intermolecular potential function as discussed by Prausnitz and Myers (*21*). The results of these calculations are designated (KIH) in Tables III and IV. The Kihara intermolecular potential function assumes that each molecule has an impenetrable core described by the three parameters M_0, S_0, and V_0 and involves a distance parameter ρ_0 and an energy parameter U_0. The

Table IV. Smoothed Experimental and Theoretical Enhancement Factors of Propylene in Helium and the Smoothed Experimental Solubility of Helium in Liquid Propylene

P, atm	ϕ EXP	ϕ LJCL	ϕ KIH	ϕ KIHCK12	ϕ KIHEK12	ϕ BWR	$100x_2$, mol %
175.00 K (This Work)							
20	*a*	1.004	1.094	1.022	1.019	1.035	0.076
40		0.9955	1.188	1.039	1.033	1.064	0.151
60		0.9796	1.287	1.057	1.048	1.093	0.224
80		0.9573	1.389	1.074	1.061	1.120	0.297
100		0.9297	1.494	1.090	1.075	1.145	0.368
120		0.8979	1.605	1.107	1.088	1.170	0.437
187.49 K (This Work)							
20	*a*	0.9976	1.085	1.024	1.021	1.034	0.107
40		0.9808	1.165	1.039	1.034	1.057	0.213
60		0.9581	1.249	1.055	1.047	1.080	0.318
80		0.9305	1.335	1.070	1.060	1.101	0.420
100		0.8989	1.424	1.085	1.071	1.121	0.519
120		0.8644	1.515	1.099	1.084	1.141	0.609
200.00 K (This Work)							
20	1.020	0.9967	1.081	1.029	1.027	1.037	0.142
40	1.029	0.9734	1.152	1.043	1.039	1.057	0.285
60	1.031	0.9455	1.225	1.057	1.050	1.074	0.428
80	1.035	0.9139	1.299	1.071	1.061	1.091	0.567
100	1.036	0.8794	1.374	1.085	1.073	1.108	0.700
120	1.036	0.8428	1.453	1.099	1.085	1.123	0.820
212.49 K (This Work)							
20	1.025	1.001	1.083	1.038	1.036	1.044	0.190
40	1.036	0.9726	1.147	1.051	1.048	1.061	0.379
60	1.040	0.9406	1.211	1.065	1.059	1.076	0.558
80	1.043	0.9057	1.277	1.078	1.070	1.091	0.738
100	1.044	0.8688	1.345	1.091	1.081	1.104	0.909
120	1.044	0.8306	1.414	1.104	1.092	1.116	1.066
224.99 K (This Work)							
20	1.040	1.011	1.089	1.051	1.049	1.056	0.233
40	1.060	0.9779	1.148	1.064	1.061	1.071	0.475
60	1.068	0.9422	1.207	1.077	1.072	1.084	0.714
80	1.072	0.9043	1.268	1.091	1.084	1.096	0.951
100	1.073	0.8651	1.329	1.103	1.095	1.107	1.175
120	1.074	0.8253	1.393	1.116	1.106	1.117	1.375
239.99 K (This Work)							
20	1.057	1.028	1.102	1.070	1.069	1.072	0.293
40	1.083	0.9922	1.159	1.086	1.083	1.088	0.613
60	1.096	0.9523	1.216	1.101	1.097	1.101	0.929
80	1.104	0.9109	1.272	1.115	1.108	1.112	1.239
100	1.108	0.8687	1.330	1.127	1.120	1.121	1.540
120	1.109	0.8266	1.388	1.141	1.132	1.130	1.830
254.98 K (This Work)							
20	1.070	1.051	1.119	1.093	1.092	1.098	0.359
40	1.107	1.014	1.180	1.116	1.114	1.119	0.775
60	1.127	0.9704	1.236	1.133	1.130	1.133	1.179
80	1.138	0.9251	1.292	1.149	1.144	1.144	1.590
100	1.144	0.8796	1.350	1.164	1.157	1.153	1.976
120	1.146	0.8353	1.409	1.179	1.171	1.161	2.284

[a] No results reported for the gas phase.

method of calculation used is that given in Prausnitz and Myers (*21*). The parameters used in the present calculations are given in Table V.

The interaction parameters $(U_0)_{12}$ and $(\rho_0)_{12}$ used to compute B_{12} were computed from the relations

$$(U_0)_{12} = (U_0)_1^{1/2}(U_0)_2^{1/2} \tag{7}$$

and

$$(\rho_0)_{12} = \frac{(\rho_0)_1 + (\rho_0)_2}{2} \tag{8}$$

For pure gases of small molecular weight such as helium it is necessary to consider quantum corrections to the second virial coefficient (*22*). Prausnitz and Myers (*21*) have suggested that for interaction between a quantum gas such as helium and a gas of relatively larger molecular weight such as ethylene, which we shall refer to as a nonquantum gas, it is necessary to include quantum effects in computing B_{ij}. This was done for the two helium systems discussed in the present paper by using the procedures given in Prausnitz and Myers (*21*).

For the KIH model calculations of ϕ_1, the third virial and third virial interaction coefficients in eq 3 and 6 have been calculated by using an empirical corresponding states correlation developed by Chueh and Prausnitz (*23*) for the third virial coefficient C of pure gases. This correlation is

$$C/V_c^2 = [0.232T_R^{-0.25} + 0.468T_R^{-5}][1 - \exp(1 - 1.89T_R^2)] + d\exp(-2.49 + 2.30T_R - 2.70T_R^2) \tag{9}$$

where

$$T_R = T/T_c \tag{10}$$

and V_c is the critical volume and T_c is the critical temperature of the substance.

Table V. Intermolecular Potential Parameters and BWR Parameters

parameter	He	C_2H_4	C_3H_6
LJCL (6–12)			
ref[a]	2	11	this work
e/k, K	6.96	150.76	167.66
b_0, cm^3/g-mol	22.9	227.06	425.14
KIHARA			
ref[a]	21	21	21
U_0/k, K	9.927	383.00	475.00
ρ_0, Å	2.921	2.950	3.439
M_0, Å	0.0	8.800	10.51
S_0, $Å^2$	0.0	3.480	5.260
V_0, $Å^3$	0.0	0.0	0.0
M	4.0026	28.0549	42.0823
BWR[d]			
ref[a]	32	33	33
A_0	1.308895 (−2)[b]	3.33958	6.11220
B_0	1.226171 (−2)	5.56833 (−2)	8.50647 (−2)
C_0	4.802198	1.31140 (5)	4.39182 (5)
a	5.759319 (−4)	2.59000 (−1)	7.74056 (−1)
b	3.352402 (−4)	8.6000 (−3)	1.87059 (−2)
c	2.440703 (−1)	2.1120 (4)	1.02611 (5)
α	2.592255 (−5)	1.78000 (−4)	4.55696 (−4)
γ	c	9.23000 (−3)	1.82900 (−2)

[a] Source of data. [b] Number in parentheses indicates powers of 10. [c] Values of γ is a function of temperature $= 3.850179 \times 10^{-3} - 2.332414 \times 10^{-5}(T) - 7.228731 \times 10^{-8}(T^2) + 6.765171 \times 10^{-10}(T^3)$. [d] Parameter units are L atm K g-mol.

Table VI. Imput Parameters for Calculation of Third Virial Coefficients Using the Method of Chueh and Prausnitz (*23*)

component	T_C, K	V_C, cm^3/mol	d_1	d_{12}
helium	10.47[b]	27.5[b]	0.0[b]	
ethylene	282.4[c]	129[c]	1.40[a]	0.70
propylene	165.0[c]	181[c]	1.80[a]	0.90

[a] These values were estimated by plotting third virial coefficient data from David and Hamanm (*24*) on the reduced curve presented by Chueh and Prausnitz (*23*). [b] Reference 23. [c] Reference 24.

Chueh and Prausnitz (*23*) recommend the use of this correlation for $T_R \geq 0.8$. The parameter d must be estimated for the particular substance for which the correlation is to be used.

For a quantum gas such as helium a modification of the strict corresponding states behavior is required. Chueh and Prausnitz (*23*) have presented a method for obtaining *effective* critical constants T_c and V_c which were used in our calculation. The effective critical temperature T_c and critical volume V_c used in eq 9 and 10 were

$$T_c = T_c^{\circ}/(1 + 21.8/MT) \qquad (11)$$

$$V_c = V_c^{\circ}/(1 + 9.9/MT) \qquad (12)$$

For helium we used $T_c^{\circ} = 10.47$ K and $V_c^{\circ} = 37.5$ cm^3/(g-mol) (*23*) with $M = 4.00$ as the molecular weight of helium.

To calculate C_{112} and C_{122}, the third virial interaction coefficients, in the various KIH models, we made use of the suggestion of Chueh and Prausnitz (*23*) regarding the use of eq 9. The input parameters used for computing the third virial coefficients from eq 9 are given in Table VI.

Kihara Core Models with K_{12} (Designated KIHCK12 and KIHEK12). In these models the geometric mixing rule for the energy parameter $(U_0)_{12}$ given in eq 7 has been modified by the factor $(1 - K_{12})$. Thus eq 7 becomes

$$(U_0)_{12} = (1 - K_{12})(U_{01})^{1/2}(U_{02})^{1/2} \qquad (13)$$

Chueh and Prausnitz (*23*) have shown that such a modification of the geometric combination rule for the energy parameters permits one to obtain a better fit of experimental high pressure phase equilibrium data for binary systems involving helium and hydrogen gases. Hiza and Duncan (*14*) and Hiza (*25*) have determined the value of K_{12} in eq 13 by a least-squares fit of experimental second virial interaction (B_{12}) data obtained from experimental phase equilibrium data for hydrogen and helium binaries together with the Kihara model. Hiza and Duncan (*14*) have also developed a semitheoretical relation for estimating the K_{12} parameter which has the form

Table VII. Values of K_{12} for the Helium–Ethylene and Helium–Propylene Systems

gas	ionization potential,[a] eV	calcd from eq 14	K_{12} (exptl)	
			this work	Hiza[b]
He	24.46			
C_2H_4	10.56	0.532	0.60	0.40
C_3H_6	9.80	0.595	0.63	

[a] Reference 26. [b] Reference 25.

Table VIII. Smoothed Values of B_{12} Extracted from Experimental Phase Equilibrium Data

T, K	B_{12}, cm^1/g-mol	T, K	B_{12}, cm^1/g-mol
Helium–Ethylene System			
90	−7.2 ± 3.5	160	22.3 ± 3.5
100	1.8 ± 3.5	170	23.7 ± 3.5
110	8.1 ± 3.5	180	24.6 ± 3.5
120	12.7 ± 3.5	190	25.3 ± 3.5
130	16.1 ± 3.5	200	25.9 ± 3.5
140	18.8 ± 3.5	210	26.3 ± 3.5
150	21.0 ± 3.5	220	26.7 ± 3.5
Helium–Propylene System			
200	32.0 ± 6	240	34.8 ± 6
210	33.4 ± 6	250	35.0 ± 6
220	34.0 ± 6	260	35.2 ± 6
230	34.5 ± 6		

$$K_{12} = 0.17(I_v - I_c)^{1/2} \ln (I_v/I_c) \qquad (14)$$

where I_v (in electron volts) is the first ionization potential of the more volatile component, say, helium, and I_c is the first ionization potential of the condensable component (say ethylene).

The model designated as KIHCK12 in Tables III and IV makes use of eq 14 to compute K_{12} for each system. The input data and value of K_{12} obtained are shown in Table VII. These values of K_{12} provided through eq 13 the means for the calculation of B_{12} and hence the enhancement factor. Thus the calculations for this model differ from those of the KIH model only in the fact that eq 13 replaces eq 7 in the calculations.

For the model designated as KIHEK12 the values of K_{12} used in the calculations were obtained from the experimental values for the second virial interaction coefficients B_{12} shown in Table VIII as determined by the method described below. The value of K_{12} for the helium–ethylene system so found was 0.60 and for the helium–propylene system was $K_{12} = 0.65$.

Benedict–Webb–Rubin Model (BWR). These calculations of the enhancement factor made use of eq 2 together with the assumption that the gas-phase mixture followed the BWR equation of state for mixtures in which the mixture parameters were computed by using the combination rules originally proposed by Benedict et al. (*27*) except that the parameter $(B_0)_{12}$ was computed by using a linear average. The values of the BWR parameters used in these calculations are given in Table V.

Discussion

Examination of Tables III and IV shows that for both the helium–ethylene and helium–propylene systems the values of ϕ_i (hence y_i) computed by using the KIHCK12 and KIHEK12 models give the best predictions of ϕ_1, with the predicted values at 100 atm agreeing with the experimental values to within about 5% above 130 K. For the helium–propylene system the (BWR) model also gives moderately good agreement.

Recently Pesuit (*30*) has discussed the estimation of K_{12} values for various types of binary systems and has concluded that the Hiza–Duncan relation (eq 14) is a useful relation for predictions of K_{12} interaction constants.

Extraction of the Second Virial Interaction Coefficient B_{12} from Gas–Liquid and Gas–Solid Phase Equilibrium Data

Reuss and Benakker (*28*), Rowlinson and Richardson (*17*), Mullins (*2*), and Chui and Canfield (*29*) among others have discussed the calculation of the second virial interaction coefficient B_{12} from experimental gas–liquid and gas–solid phase equilibrium data.

If the virial equation of state in the Leiden form (eq 3) through the third coefficient is used to describe the gas-phase mixture of a gas–liquid equilibrium, then eq 6 may be rearranged and solved for B_{12} to give

$$B_{12} = \frac{V_m}{2y_2}\left[\left(\frac{2B_{11}}{V_{01}} + \frac{3C_{111}}{2V_{01}^2}\right) - \ln Z_{01} - \frac{3}{2V_m^2}(y_1^2C_{111} + 2y_1y_2C_{112} + y_2^2C_{122}) + \frac{1}{RT}\int_{P_{01}}^{P} V_1^c\,dP - 2y_1\frac{B_{11}}{V_m} + \ln Z_m + \ln x_1 - \ln\frac{Py_1}{P_{01}}\right] \quad (15)$$

along a given experimental isotherm eq 15 is solved for B_{12} for each pressure by making use of experimental values for y_i, x_i, V_{01}, and V_1^c and P_{01} for the saturated phase and B_{11} and B_{22} and by assuming a model from which the third virial interaction coefficient C_{112} and C_{122} as well as C_{111} and C_{222} may be calculated. The volume V_m is also calculated by using these third virial coefficients together with an estimated value of B_{12}. The values of B_{12} so found from eq 15 are then plotted vs. the quantity $(P - P_{01})$ and the curve is extrapolated to $(P - P_{01}) = 0$ to obtain B_{12}. The values of B_{12} so obtained for each temperature were then plotted as a function of T (K) and a smooth curve was drawn through the data. Smoothed values were then read from this curve. Smoothed values of B_{12} for the two systems are shown in Table VIII. The uncertainty listed for each value is based primarily upon the effects on B_{12} of the uncertainty in the experimental values of y_i and x_i on the calculation of B_{12} from eq 15.

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Solubility of Some Alkali and Alkaline Earth Chlorides in Water at Moderate Temperatures

M. A. Clynne* and R. W. Potter II

U.S. Geological Survey, Menlo Park, California 94025

Solubilities for the binary systems, salt–H_2O, of the chlorides of lithium, rubidium, cesium, magnesium, calcium, strontium, and barium from near 0 °C to the saturated boiling point are reported. The experimental data and coefficients of an equation for a smoothed curve describing each system are listed in the tables. The data are improvements on those previously reported in the literature, having a precision on the average of ±0.09%.

Introduction

The solubilities of simple salts in pure water and aqueous solutions at moderate temperatures have a wide variety of applications. Geothermal energy, solution mining, radioactive waste disposal, and desalinization technologies all have need for accurate data on salt solubility. In addition, recently developed empirical models for the solubility of salts in complex brines require as input reliable binary and ternary solubility data (*1*). However, much of the older, available data lack the desired level of internal consistency (*1*, *2*). To improve the existing data base as well as to better understand the physical chemistry of concentrated electrolyte solutions, we have undertaken the measurement of the solubility of highly soluble salts. Previously we reported solubility data for NaCl, KCl, $CaCl_2$, Na_2SO_4, and K_2SO_4 (*3*). This paper reports on the solubilities of additional highly soluble salts in pure water from near 0 °C to the saturated