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Phase-Equilibria Behavior of Systems Carbon Dioxide-2-Methylnaphthalene and Carbon Dioxide-*n*-Decane-2-Methylnaphthalene

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The phase-equilibria behavior of the binary system CO_2–2-methylnaphthalene was studied for liquid-vapor, solid-liquid-vapor, and liquid-liquid-vapor systems. The liquid-liquid-vapor equilibria of the ternary system CO_2–2-methylnaphthalene–*n*-decane were studied in detail. As well as locating where the liquid-liquid-vapor loci exist in pressure-temperature space, the distribution of hydrocarbons in the individual liquid phases was measured. The ternary liquid-liquid-vapor locus exhibited either *UCST* or *K*-type termination at the upper end of the locus, depending upon initial mole ratio of hydrocarbons. Furthermore, the CO_2-richer phase (L2) preferred selectively *n*-decane. Also presented are the data on the depression in the freezing point of 2-methylnaphthalene upon the addition of *n*-decane at 1 atm and on the elevation in the freezing point of 2-methylnaphthalene owing to pressurization.

The ability of CO_2 to induce partial miscibility in hydrocarbons suggests separation processes with liquid CO_2 as a selective solvent. Its potential utility in this respect is attractive because of its availability and other desirable properties like low toxicity and noncorrosiveness. Moreover, the recovery of CO_2 from the hydrocarbons by mere depressurization, which eliminates the need of expensive distillation processes, makes the use of CO_2 particularly attractive.

To make full use of CO_2 as an extractive solvent, it is desirable to understand the ability of CO_2 to separate different chemical groupings such as lower vs. higher (i.e., with respect to molecular weight) alkanes, aromatics vs. alkanes, and single-ring aromatics vs. multiple-ring aromatics. For this reason, well-defined ternary prototype systems are being studied in detail. These studies would also provide accurate data for correlative purposes. The data obtained on the ternary system are such as to facilitate modeling these systems by solution models. Since most models use binary interactions in describing the ternary data, the binary systems constituting the ternary system are also studied. It is hoped that a model based only on binary interaction forces, obtained from relatively simple experiments such as solubility limits or freezing-point depressions, can a priori determine the types of systems to which this process might find application. As more systems are studied and their data analyzed, a model or models should evolve that will serve this purpose and, furthermore, aid in the design of the separation process.

Huie and coworkers (*4, 5*) studied in detail the system CO_2–*n*-decane–*n*-eicosane. One of the goals of the Huie study was to answer questions regarding the selectivity of CO_2 for lower vs. higher alkanes. Huie's results show that CO_2 can separate lower alkanes from the higher alkanes. These results are in agreement with other studies (*3*).

The Huie study is being extended to systems containing CO_2, alkanes, and aromatics. Of interest here is the selectivity of CO_2 for alkanes vs. aromatics. The study on the prototype system CO_2–*n*-decane–2-methylnaphthalene is now completed. Two out of the three components of this ternary system are the same as that of Huie's system. The third component, namely, 2-methylnaphthalene, has a similar melting point to that of *n*-eicosane. Work on other prototype systems which are parallel to this system and extend Huie's work is also being done (*17*). Furthermore, efforts are being made to correlate the phase-equilibria behavior of these systems by use of well-known solution models.

Phase-equilibria studies on a variety of CO_2–alkane systems have been conducted by a large number of investigators (*1, 6, 10–16*). However, there are little data on the systems of CO_2 and aromatics. The authors believe that the only study done on the systems of CO_2 and aromatics is by Francis (*2*). The partial miscibility characteristics of CO_2 in a variety of hydrocarbon solvents, including aromatics, have been investigated by Francis in an extensive scanning study. He has presented only compositional data on these systems in the form of triangular diagrams at room temperature. In view of the lack

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of precise compositional, volumetric, and state information, the Francis data are not particularly suitable for correlative purposes.

On the basis of the data available, it was deemed necessary that detailed multicomponent, multiphase data would be needed to understand sufficiently the selectivity behavior of liquid carbon dioxide.

Experiments Performed

The following experiments were performed to answer the questions regarding the selectivity of CO_2 for alkanes vs. aromatics and to provide the data for use in correlations as mentioned earlier:

Liquid-vapor isotherms were performed on the binary and ternary systems: namely, CO_2–2-methylnaphthalene and CO_2–*n*-decane–2-methylnaphthalene. (Detailed data on the binary system CO_2–*n*-decane are available (*7, 14*); hence, this system is not investigated here.)

The depression in the freezing point of 2-methylnaphthalene upon the addition of *n*-decane was measured at several compositions at atmospheric pressure.

The three-phase solid-liquid-vapor and liquid-liquid-vapor loci were determined for the binary system CO_2–2-methylnaphthalene.

The liquid-liquid-vapor regime of the ternary system CO_2–*n*-decane–2-methylnaphthalene was studied in detail.

Pressure-temperature data were obtained for the solid-liquid locus of pure 2-methylnaphthalene.

Experimental

A detailed description of the experimental equipment and procedure is given elsewhere (*4, 5, 7, 8, 17*). The only significant innovation introduced in our experimental procedure is the analysis of samples withdrawn from coexisting liquid phases for hydrocarbon mole ratio with a refractometer.

Table I. Smoothed Values of Pressure, Composition, and Molar Volume for Liquid-Vapor Isotherm on Carbon Dioxide–2-Methylnaphthalene System at 34°C

$\bar{V}_{CO_2} =$ 43.3 cc/g-mol
$\bar{V}_{2mn} =$ 142.5 cc/g-mol

Press, atm	CO_2 liquid mole fraction	Liquid molar vol, cc/g-mol
15.0	0.093	133.3
20.0	0.125	130.1
25.0	0.157	126.9
30.0	0.190	123.7
35.0	0.223	120.4
40.0	0.256	117.1
45.0	0.289	113.8
50.0	0.322	110.6
55.0	0.360	107.1
60.0	0.388	104.5
65.0	0.416	101.7
	Raw experimental data	
8.66	0.0543	136.02
10.86	0.064	136.33[a]
16.69	0.1043	130.52
19.84	0.1233	130.20[a]
26.56	0.1677	124.18
34.20	0.2202	120.89[a]
39.90	0.2531	116.75
49.10	0.3182	111.91[a]
53.31	0.3442	108.61
55.63	0.3611	107.71[a]
61.47	0.3973	103.70
68.84	0.4344	103.31[a]

[a] Duplicate run.

The compositions were determined to an estimated accuracy of 0.002 mole fraction. Temperatures were measured and maintained to an estimated accuracy of ±0.02°C. The maximum error in measuring pressures was estimated to be ±0.07 atm. Liquid volumes were judged to be accurate to ±0.02 ml. The maximum error in reported molar volumes is estimated to be ±0.5 cc/g-mol.

The pressures and temperatures for the elevation of the freezing point of 2-methylnaphthalene owing to pressure were determined to an estimated accuracy of ±200 psig and ±0.1°C, respectively.

Materials

The CO_2 used in this study was obtained from the Matheson Co. as "Coleman grade" material with a stated minimum purity of 99.99%. Oxygen, carbon monoxide, and hydrogen were the major impurities.

The CO_2 was prepared for use by flashing it from the cylinder at room temperature to the 3000-cc storage reservoir maintained at 0°C. In doing this, it was hoped that the resulting liquefaction of the CO_2 would make it free from impurities, the impurities remaining in the vapor space. Subsequent venting of the vapor phase in the reservoir would then remove these impurities.

The Kuenen criterion of purity (the difference between the bubble-point and the dew-point pressure divided by the critical pressure) was less than 0.01. The properties vapor pressure, sublimation pressure, and critical and triple point were within 0.3 atm and 0.1°C of the literature values (*9*).

The *n*-decane and 2-methylnaphthalene used in this study were, respectively, the Humphrey-Wilkinson and Aldrich products labeled as having 99% minimum purity. The melting points of *n*-decane and 2-methylnaphthalene were observed to be −29.60° and 33.45°C, respectively. Both the *n*-decane and the 2-methylnaphthalene were used without further purification.

Results

CO_2–2-methylnaphthalene binary system. Liquid-vapor isotherms were performed on this binary system at four different temperatures, the lowest temperature being just above the melting point of 2-methylnaphthalene. The smoothed values of pressure, liquid-phase composition, and molar volume obtained from these liquid-vapor isotherms are presented in Tables I–IV. The partial molar volumes were independent of the composition over the range of mole fractions encountered. The raw data of Table I are given to provide a sense of the actual experimental data variation encountered.

The data on the depression in the freezing point of 2-methylnaphthalene upon CO_2 pressurization (S1-L1-V locus) are reported in Table V.

Table VI presents the smoothed values of compositions, pressure, temperature, and molar volume of the binary system CO_2–2-methylnaphthalene along the liquid-liquid-vapor locus.

The termination points of the L1-L2-V locus were observed in separate runs in which one or the other of the two liquid phases was present in a trace amount. The differences between the observed pressure and temperature at the Q1-point or the *K*-point in runs where L1 or L2 was in trace amount are within experimental accuracy as reported in Table VI. This would indicate that the CO_2 used is very pure.

2-Methylnaphthalene-n-decane binary system. The data on the depression in freezing point at atmospheric pressure of 2-methylnaphthalene upon the addition of *n*-decane are presented in Table VII.

CO_2-2-methylnaphthalene-n-decane ternary system. Liquid-vapor isotherms were performed on this ternary system at 50°C and two different initial mole ratios (Tables VIII and IX).

The liquid-liquid-vapor region of this ternary system was investigated in detail. As well as locating where the liquid-liquid-vapor region exists in pressure-temperature space, the mole ratios of hydrocarbons in the individual liquid phases were measured.

Smoothed values of pressure and temperature of the L1-L2-V locus for this ternary system are presented in Table X for various starting mole ratios of *n*-decane to 2-methylnaphthalene.

At lower starting mole ratios of *n*-decane to 2-methylnaphthalene, the termination points are *Q*-type at the lower end and *K*-type at the upper end of the L1-L2-V

Table II. Smoothed Values of Pressure, Composition, and Molar Volume for Liquid-Vapor Isotherm on Carbon Dioxide-2-Methylnaphthalene System at 51°C

$\bar{V}_{CO_2}$ = 46.5 cc/g-mol
$\bar{V}_{2mn}$ = 144.5 cc/g-mol

Press, atm	CO_2 liquid mole fraction	Liquid molar vol, cc/g-mol
15.0	0.085	136.2
20.0	0.111	133.6
25.0	0.137	131.1
30.0	0.161	128.7
35.0	0.185	126.4
40.0	0.209	124.0
45.0	0.235	121.5
50.0	0.260	119.0
55.0	0.285	116.6
60.0	0.310	114.1
65.0	0.335	111.7

Table III. Smoothed Values of Pressure, Composition, and Molar Volume for Liquid-Vapor Isotherm on Carbon Dioxide-2-Methylnaphthalene System at 75°C

$\bar{V}_{CO_2}$ = 47.1 cc/g-mol
$\bar{V}_{2mn}$ = 147.6 cc/g-mol

Press, atm	CO_2 liquid mole fraction	Liquid molar vol, cc/g-mol
15.0	0.064	141.4
20.0	0.085	139.2
25.0	0.106	137.0
30.0	0.126	135.0
35.0	0.146	132.8
40.0	0.165	130.6
45.0	0.185	128.9
50.0	0.202	127.0
55.0	0.223	125.2
60.0	0.241	123.2
65.0	0.259	121.4

Table IV. Smoothed Values of Pressure, Composition, and Molar Volume for Liquid-Vapor Isotherm on Carbon Dioxide-2-Methylnaphthalene System at 100°C

$\bar{V}_{CO_2}$ = 50.0 cc/g-mol
$\bar{V}_{2mn}$ = 150.0 cc/g-mol

Press, atm	CO_2 liquid mole fraction	Liquid molar vol, cc/g-mol
15.0	0.054	144.4
20.0	0.073	142.6
25.0	0.090	140.8
30.0	0.107	139.1
35.0	0.122	137.6
40.0	0.141	135.8
45.0	0.157	134.3
50.0	0.173	132.8
55.0	0.189	131.2
60.0	0.205	130.1
65.0	0.221	128.2

Table V. Smoothed Values of Pressure, Temperature, Composition, and Molar Volume of Solid-Liquid 1-Vapor Locus for System Carbon Dioxide-2-Methylnaphthalene

Press, atm	Temp, °C	CO_2 mole fraction L1	Liquid molar vol, cc/g-mol	Comment
1.0	33.45	0.0	142.5	2mn, mp
5.0	32.00	0.032	138.9	
10.0	29.75	0.065	135.6	
15.0	27.63	0.098	132.3	
20.0	25.50	0.133	128.8	
25.0	23.00	0.175	124.5	
30.0	20.00	0.230	118.9	
35.0	16.13	0.305	111.3	
40.0	11.58	0.407	101.0	
42.35	9.09	0.477	94.5	Q-point

Table VI. Smoothed Values of Pressure, Temperature, Composition, and Liquid Molar Volumes of Liquid 1-Liquid 2-Vapor Locus for System Carbon Dioxide-2-Methylnaphthalene

Temp, °C	Press, atm	CO_2 mole fraction		Liquid molar vol, cc/g-mol		Comment
		L1	L2	L1	L2	
9.09	42.32	...	...	...	...	Q-point (L2)
9.10	42.35	0.4772	0.9772	94.5	51.5	Q-point (L1)
11.00	44.20	0.4818	0.9772	94.0	52.0	
13.00	46.50	0.4859	0.9773	93.7	52.6	
15.00	48.75	0.4898	0.9774	93.4	53.1	
17.00	51.00	0.4936	0.9779	93.2	53.8	
19.00	53.70	0.4968	0.9780	93.0	54.7	
21.00	56.10	0.4996	0.9782	92.9	55.7	
23.00	58.60	0.5017	0.9791	93.0	56.8	
25.00	61.25	0.5025	0.9802	93.3	58.2	
27.00	64.25	0.5018	0.9815	93.6	59.8	
29.00	67.20	0.5000	0.9831	94.6	61.8	
31.00	70.02	0.4967	0.9850	95.5	64.0	
33.00	73.50	0.4912	0.9869	96.9	66.7	
35.00	76.80	0.4825	0.9890	99.0	70.0	
36.00	78.50	0.4740	0.9904	100.4	71.8	
36.29	79.13	...	...	...	...	K-point (L2)
36.33	79.15	0.4702	...	100.87	...	K-point (L1)

Table VII. Smoothed Data on Depression in Freezing Point of 2-Methylnaphthalene upon Addition of *n*-Decane at Atmospheric Pressure

Temp, °C	Mole fraction 2-methylnaphthalene
33.45	1.0
30.0	0.939
25.0	0.841
20.0	0.730
15.0	0.623
10.08	0.521
5.0	0.418

Table VIII. Smoothed Values of Pressure, Composition, and Molar Volume for Liquid-Vapor Isotherm at 50°C of Ternary System Carbon Dioxide–2-Methylnaphthalene–*n*-Decane

$(N_{10}/N_{2mn}) = 0.3704$

Press, atm	Liquid mole fraction		Liquid molar vol, cc/g-mol
	CO_2	2mn	
15.0	0.098	0.658	148.8
20.0	0.131	0.634	145.1
25.0	0.164	0.610	141.4
30.0	0.197	0.586	137.7
35.0	0.230	0.562	134.0
40.0	0.261	0.539	130.6
45.0	0.294	0.515	126.9
50.0	0.327	0.491	123.2
55.0	0.361	0.466	119.4
60.0	0.396	0.441	115.5
65.0	0.431	0.415	111.6

Table IX. Smoothed Values of Pressure, Composition, and Molar Volume for Liquid-Vapor Isotherm at 50°C of Ternary System Carbon Dioxide–2-Methylnaphthalene–*n*-Decane

$(N_{10}/N_{2mn}) = 1.6873$

Press, atm	Liquid mole fraction		Liquid molar vol, cc/g-mol
	CO_2	2mn	
15.0	0.121	0.327	164.0
20.0	0.161	0.312	158.9
25.0	0.201	0.297	153.7
30.0	0.240	0.283	148.7
35.0	0.280	0.268	143.6
40.0	0.318	0.254	138.7
45.0	0.357	0.239	133.6
50.0	0.397	0.225	128.5
55.0	0.438	0.209	123.2
60.0	0.477	0.195	118.2
65.0	0.518	0.179	112.9

loci, whereas at higher starting mole ratios of *n*-decane to 2-methylnaphthalene, a *UCST*-type termination at the upper end of the L1-L2-V locus was observed, the lower end termination being *Q*-type again.

The transition from *K*-type termination of the L1-L2-V locus to *UCST*-type termination occurs when the initial starting mole ratio of *n*-decane to 2-methylnaphthalene is between 0.46–0.5.

The pressure-temperature projection of the ternary L1-L2-V loci is shown in Figures 1 and 2.

Sampling runs were performed for five different initial mole ratios at three different temperatures to assess the separational effect between the liquid phases in this ternary system.

The average absolute difference in reproducing mole ratios in replicate runs was ±0.009 for the L1 phase and ±0.012 for the L2 phase.

A measure of the separation of *n*-decane from 2-methylnaphthalene with CO_2 as an extractive solvent may be ascertained by computation of selectivity (β) on a CO_2-free basis:

$$\beta = (x_{10}/x_{2mn})_{L2}/(x_{10}/x_{2mn})_{L1}$$

where $x_{10} + x_{2mn} = 1$ in each phase.

Table XI gives the values of CO_2-free selectivities for various starting mole ratios of *n*-decane to 2-methylnaphthalene at 30°, 20°, and 15°C. The values of mole ratios, $(x_{10}/x_{2mn})_{L1}$ and $(x_{10}/x_{2mn})_{L2}$ used in calculating the selectivities, were smoothed by plotting them against initial mole ratios at each temperature. The standard deviations of the raw data from the smoothed values of mole ratios are ±0.015 for the L1 phase and ±0.042 for the L2 phase.

It appears from Table XI that the selectivities show a slight decrease with increase in initial mole ratio as well as with increase in temperature. The probable error in the selectivity values is estimated to be ±0.18.

Freezing-point elevation of 2-methylnaphthalene. Three

Table X. Smoothed Values of Pressure and Temperature of Liquid 1-Liquid 2-Vapor Locus for Ternary System Carbon Dioxide–*n*-Decane–2-Methylnaphthalene at Several Values of Initial Overall Mole Ratio ($r = N_{10}/N_{2mn}$)

Temp, °C	Press, atm							Comment
	r = 0.116	0.222	0.375	0.463	0.500	0.550	0.562	
1.7					34.58[a]			Q
1.9			34.68[a]					Q
3.52		36.37						Q
4.28	37.25							Q
5.00	37.86	37.70	37.50	37.48	37.45	37.12	36.99	
6.00					38.25	38.05	37.88	
7.00					39.19	39.00	38.82	
8.00					40.18	39.98	39.80	
9.00					41.20	40.99	40.82	
9.22							41.02	*UCST* ($x_{CO_2} = 0.834$)[b]
10.00	43.01	42.52	42.30	42.28	42.25	42.02		
10.33						42.35		*UCST* ($x_{CO_2} = 0.831$)
10.69					42.97			*UCST* ($x_{CO_2} = 0.791$)
15.00	48.21	47.71	47.39	47.20				
20.00	54.10	53.64	53.20	52.80				
25.00	60.55	60.55	59.49	59.05				
30.00	67.90	67.00	66.30	66.10				
35.00	75.49	74.40	73.91	73.50				
39.32	82.60							K
40.00		82.55	82.00	81.45				
41.20		84.90[a]						K
43.63			88.15					K
44.60				89.40[a]				K

[a] Estimated. [b] Estimated *UCST* CO_2 composition in the case of pure *n*-decane is 0.850 at −24.41°C and 16.16 atm.

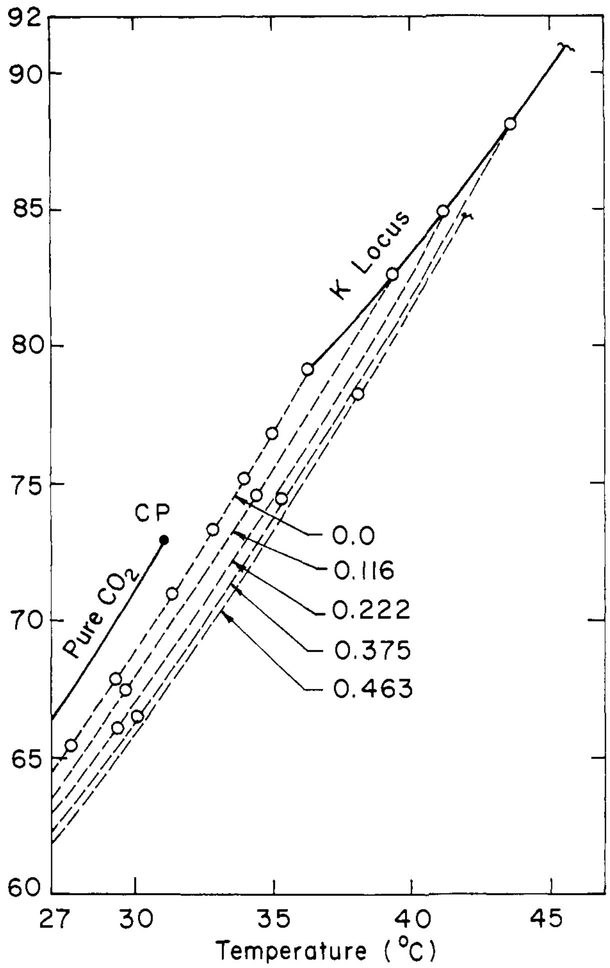

Figure 1. Upper end of liquid 1-liquid 2-vapor surface

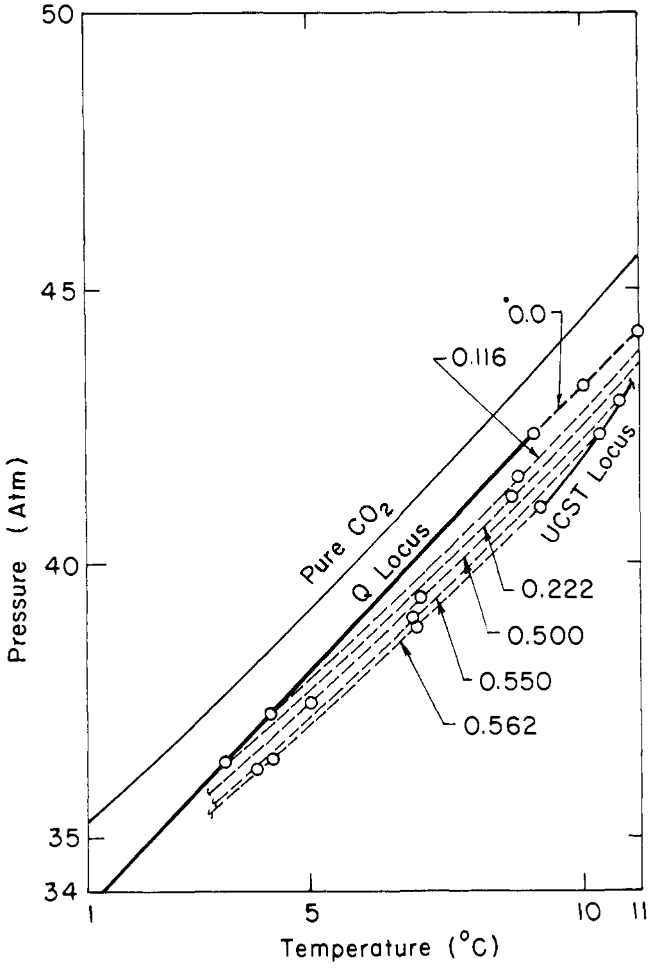

Figure 2. Lower end of liquid 1-liquid 2-vapor surface

Table XI. Selectivities (β) of Carbon Dioxide for *n*-Decane as Function of Initial Overall Mole Ratio (N_{10}/N_{2mn}) at 30°, 20°, and 15°C

Initial mole ratio, N_{10}/N_{2mn}	Selectivity		
	$T = 30°C$	$T = 20°C$	$T = 15°C$
0.15	2.64	2.67	2.76
0.20	2.27	2.40	2.53
0.25	2.06	2.24	2.40
0.30	1.93	2.14	2.31
0.35	1.84	2.07	2.25
0.40	1.78	2.01	2.20

Table XII. Experimental Data on Elevation in Freezing Point of 2-Methylnaphthalene upon Pressurization

Press, atm	Temp, °C
1.0	33.45
692.4	50.0
1343.9	69.2
2004.0	84.0

points were obtained on the solid-liquid locus of pure 2-methylnaphthalene (Table XII). These data are valuable for correlative purposes; they define the change in volume of 2-methylnaphthalene upon fusion as a function of temperature through use of the Clausius-Clapeyron equation.

Remarks

On the basis of the sampling results presented in Table XI, it appears that the CO_2-rich phase (L2) selectively extracts lower alkanes (in this context, *n*-decane) from aromatics (in this context, 2-methylnaphthalene), multi-ring aromatics to be precise.

When this study is considered in concert with Huie's study, one might further conclude that in the liquid-liquid-vapor region of a ternary system the CO_2-rich phase selectively extracts the substance which is more miscible with it in the temperature range of investigation. Indications are this trend also seems to hold for the other CO_2–alkane–aromatic system, namely CO_2–*n*-butylbenzene–*n*-eicosane which is presently under investigation at this site (*17*).

Thus, one might preliminarily conclude that it is more useful to consider the miscibility relationship of CO_2 with the individual hydrocarbons, rather than its chemical grouping when considering its selective solvent behavior. This hypothesis is being further studied by our laboratory at the present time.

The term "starting mole ratio" or "initial mole ratio" is synonymous with the mole ratio in L1, $(x_{10}/x_{2mn})_{L1}$, in this study. Very little hydrocarbon was present in phase L2 in preparing the data for Table X, by reason of limiting the amount of L2 in the cell to a trace quantity. Furthermore, the concentration of hydrocarbon in L2 is, by nature of the systems studied, small, which fact has bearing on the measurements used in preparing Table XI.

Nomenclature

K-point = singular point where L2 is in critical identity with V in the presence of L1 phase
L1 = CO_2-lean liquid phase
L2 = CO_2-rich liquid phase
N_i = number of moles of component i
P = pressure, atm
$Q1$ = quadruple point 1 (coexistence of the four-phase V-L1-L2-S1)
S1 = solid 2-methylnaphthalene
T = temperature, °C
$UCST$ = upper critical solution temperature of L1 and L2
V = vapor phase
$\bar{V}$ = partial molar volume, cc/g-mol
x_i = mole fraction of species i in the liquid phase

Greek Letter

β = extractive solvent (CO_2) free selectivity

Subscripts

2mn = 2-methylnaphthalene
10 = n-decane

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Calorimetric Investigation of Salt–Amide Interactions in Aqueous Solution

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Enthalpies of solution of the liquid amides, formamide, *N*-methylformamide, *N,N*-dimethylformamide, *N,N*-dimethylacetamide, and *N*-methylpropionamide and of acetamide (c) and *N*-methylacetamide (c) were determined in water and in solutions of electrolytes, mainly alkali and alkaline earth halides, over a range of salt molalities at amide molalities below 0.05*m* at 25°C. Enthalpies of transfer of the amides from water to salt solution were calculated from these data. With formamide, these quantities were negative for all salts investigated. For a series of salts having an ion in common, the transfer enthalpies of formamide became increasingly negative with increasing crystal radius of the other ion. Enthalpies of transfer of the other amides were more positive than for formamide, reflecting the contribution of added nonpolar groups. Enthalpies of transfer of *N,N*-dimethylacetamide differed markedly for the same salt from those of *N*-methylpropionamide, suggesting steric or charge distribution differences between structural isomers.

The thermodynamic quantities relating to the transfer of amides and peptides from water to salt solutions are useful in understanding processes involving the interaction of proteins and salts as, for example, the denaturation of proteins by electrolytes (*17*). Of particular interest is the relative importance of structural features of the amide and peptide molecules, such as number, kind, and arrangement of nonpolar groups, dipole moment, and hydrogen bonding ability, in affecting the sign and magnitude of these quantities. Although free energies of transfer for various compounds have been obtained (*7, 10, 11*), there has been little in the way of direct measurement of the enthalpy changes attending the transfer of amides and peptides from water to salt solution (*2*).

The present study had the aim of developing enthalpy of transfer data for a number of amides and peptides with a variety of salts. A qualitative comparison of the different structural influences could then be made as well as a correlation of various trends with salt type. Since Stern and his collaborators (*14, 15*) had measured enthalpies of transfer for several nonelectrolytes other than amides, a general comparison of trends for different nonelectrolytes was also possible.

Experimental

Materials. Formamide was obtained from either Nuclear Enterprises or Eastman Kodak Co. It was fractionally recrystallized twice under dry nitrogen (*18*). The recrystallized material had a melting point of 2.55°C. *N*-methylformamide (Eastman) was dried over phosphorus pentoxide, vacuum distilled, and stored over molecular

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