Solid−Liquid Equilibria for Binary Mixtures Composed of Acenaphthene, Dibenzofuran, Fluorene, Phenanthrene, and Diphenylmethane

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The liquidus lines were determined with a solid-disappearance method for binary mixtures composed of acenaphthene, dibenzofuran, fluorene, phenanthrene, and diphenylmethane. While the first four substances are model compounds of wash oil, which has widely been used as a solvent to remove aromatics from coal oven gas, diphenylmethane is a high-boiling and low-melting compound that is a potential additive to modify the performance of wash oil. Each of the seven binaries appears to be a simple eutectic system. as evidenced by the experimental results. The Wilson and the NRTL models were employed to correlate the solid−liquid equilibrium data. Both activity coefficient models were found to represent accurately the nonideality of the liquid-phase for the investigated systems.

Introduction

The fractionation cuts obtained from coal tar have many industrial applications. One of these products, boiling range from 523 K to 573 K, was widely used as a solvent, called wash oil, to remove aromatics (benzene, toluene, and xylenes) from coal oven gas. Acenaphthene, dibenzofuran, fluorene, and phenanthrene are the major components of this fractionation cut. However, crystallization would take place in the recycled wash oil, leading to blockage of the stream, if the wash oil contains excessive acenaphthene. Blending a low-melting and high-boiling substance with the wash oil could be a feasible way to prevent solid formation in the recycled stream. Diphenylmethane is a potential modifier for this purpose due to its appropriate physical properties; the normal boiling temperature is 537.42 K, and the normal melting temperature is 298.2 K. Its molecular structure, containing two phenyl groups, is also expected to help in aromatics' removal.

The performance of the modified wash oil is governed by the phase behavior of the related mixtures. In the present study, solid−liquid equilibria (SLE) were measured for seven binary systems composed of acenaphthene, dibenzofuran, fluorene, phenanthrene, and diphenyl-methane with a solid-disappearance method over the entire composition range. The empirical equation of Ott and Goates (1983), the Wilson model (Wilson, 1964), and the NRTL model (Renon and Prausnitz, 1968) were applied to correlate these new SLE data. The enthalpy of fusion was also determined for diphenylmethane with a differential scanning calorimeter.

Experimental Section

Acenaphthene (99 mass %), dibenzofuran (99+ mass %), phenanthrene (98 mass %), and diphenylmethane (99+ mass %) were purchased from Aldrich (Milwaukee, WI). Fluorene (99+ mass %) was supplied by Fluka (Swiss). All

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these substances were used without further purification. Their melting points are compared in Table 1. The liquidus lines were determined by a solid-disappearance method, which has been verified previously for its reliability (Lee and Chi, 1993). The experimental procedure was described in detail elsewhere (Lee and Chi, 1993). Each mixture sample (about 3 g) was prepared by weighing pure compounds of interest to ± 0.1 mg and sealing them in a tiny glass vial. The solidified sample was then shaken vigorously in a visual thermostatic bath (Neslab, TV-4000, stability = ± 0.03 K) for observation of the solid disappearance. The bath temperature was measured with a Hart Scientific Microtherm (Model 1506) with a platinum RTD probe to ± 0.02 K. The uncertainty of the reported solid-disappearance temperatures was within ± 0.2 K under normal experimental conditions and ± 0.5 K in the vicinity of the eutectic point, as determined by repeatedly measuring the disappearance temperatures of the same sample.

Results and Discussion

The melting temperature (T_m) of each constituent compound measured in this study is compared with literature values in Table 1. The agreement is within ± 0.5 K. The solid-disappearance temperatures are listed in Table 2 for acenaphthene + dibenzofuran, + fluorene, and + phenanthrene and in Table 3 for diphenylmethane + acenaphthene, + dibenzofuran, + fluorene, and + phenanthrene. All seven binaries were found to be simple eutectic systems. These eutectic points are indicated in the tables. Figures 1 and 2 illustrate the SLE phase boundaries of acenaphthene + phenanthrene and phenanthrene + diphenylmethane, respectively. As seen from the tabulated values, blending diphenylmethane with each of the model compounds can effectively decrease its melting point. The lowest melting temperature (the eutectic temperature) of each diphenylmethane binary is lower than the melting temperature of each corresponding pure model compound, acenaphthene, dibenzofuran, fluorene, and phenanthrene, by 75.1 K, 66.1 K, 94.1 K, and 81.3K, respectively.

Table 1. Properties of Pure Compounds

substance	T_m/(K) this work	T_m/(K) lit.	$\Delta_{fus}H^a$ (kJ·mol^{-1})	$\Delta C_p{}^a$ (J·mol^{-1}·K^{-1})	$V_L{}^b$ (cm^3·mol^{-1})
acenaphthene	366.3	366.56[c] 366.6[d] 366.5[e]	21.476	14.855	149.8
dibenzofuran	355.7	355.65[c] 355.3[f] 355.3[g] 355.7[h]	18.600	9.6	152.5
fluorene	387.5	387.94[c] 387.9[d] 387.6[e] 387.9[f] 387.9[g]	19.591	1.444	163.7
phenanthrene[i]	372.5	372.3[c] 372.4[d] 372.8[e] 372.0[j]	16.474	12.586	168.1
diphenylmethane	298.2	298.39[c]	18.300[k]		172.5

[a] Taken from Coon et al. (1989) if not specifically noted. [b] Estimated from the modified Rackett equation (Spencer and Danner, 1972). [c] Tsonopoulos et al. (1986). [d] Finke et al. (1977). [e] Chio et al. (1985). [f] Domanska et al. (1993). [g] Sediawan et al. (1989). [h] Gupta et al. (1994). [i] $\Delta_p H_i = 1300$ J·mol^{-1} and $\Delta_p S_i = 3.85$ J·mol^{-1}·K^{-1} (Coon et al., 1989). [j] Mayer et al. (1990). [k] Determined in this study.

Table 2. Solid−Liquid Equilibria for Acenaphthene Systems

x_1	T (K)	δT^a (K)	x_1	T (K)	δT^a (K)
Acenaphthene (1) + Dibenzofuran (2)
0.1116	349.0	0.2	0.4216	328.2[b]	−0.3
0.1832	345.7	−0.5	0.4345	328.7	−0.1
0.2448	341.6	0.0	0.4533	329.8	0.4
0.3186	336.6	0.1	0.5055	334.1	0.3
0.3244	336.1	0.2	0.5668	339.2	−0.1
0.3525	333.7	0.4	0.6357	344.2	0.0
0.3958	330.3	0.1	0.7091	349.2	0.1
0.4052	329.5	0.0	0.8055	355.4	0.1
0.4124	328.6	0.2	0.9066	361.5	−0.1
0.4183	328.5	−0.3			
Acenaphthene (1) + Fluorene (2)
0.1562	376.7	0.1	0.5855	340.8	0.3
0.2735	367.8	0.0	0.5919	341.2	0.4
0.3667	360.3	−0.4	0.6539	346.2	0.0
0.4588	352.1	−0.5	0.6693	347.6	−0.4
0.5046	347.1	0.2	0.7139	350.0	0.2
0.5616	341.7	0.1	0.7841	354.6	0.0
0.5713	340.2[b]	0.3	0.8984	361.5	0.0
0.5765	340.5	−0.2			
Acenaphthene (1) + Phenanthrene (2)
0.0998	364.2	0.0	0.4708	333.6	0.4
0.2349	354.7	−0.1	0.4849	333.1[b]	0.1
0.3035	348.8	0.2	0.4889	333.4	0.1
0.3526	344.6	0.0	0.5557	338.4	0.1
0.4084	339.7	−0.2	0.5799	340.4	−0.1
0.4255	338.0	−0.1	0.6903	348.3	−0.3
0.4447	336.4	−0.2	0.7936	354.7	0.0
0.4591	334.8	0.2	0.8602	358.6	−0.1

[a] δT/K $= T_{(calc)} - T_{(expt)}$, where $T_{(calc)}$ was calculated from eq 1. [b] Eutectic temperature.

Empirical Correlation

For interpolation purposes, the liquidus lines were correlated piecewise with the empirical equation of Ott and Goates (1983) given by

$$T = T^*[1 + \sum_{j=1}^{M} b_j(x_1 - x_1^*)^j] \qquad (1)$$

where x_1 is the mole fraction of component 1, T is the solid-disappearance temperature of the mixture, and b_j's are the coefficients to be determined by regression to the experi-

Table 3. Solid−Liquid Equilibria for Diphenylmethane Systems

x_1	T (K)	δT^a (K)	x_1	T (K)	δT^a (K)
Acenaphthene (1) + Diphenylmethane (2)
0.1000	294.0	0.0	0.2999	312.8	0.1
0.1194	293.1	0.0	0.3996	324.3	0.0
0.1396	292.2	0.1	0.4981	333.4	−0.1
0.1609	291.5	−0.1	0.5999	341.6	−0.1
0.1653	291.2[b]	0.0	0.6993	348.8	0.1
0.1799	294.0	0.0	0.7932	355.5	0.0
0.2005	297.8	0.0	0.8979	361.9	0.0
Dibenzofuran (1) + Diphenylmethane (2)
0.0997	294.5	−0.1	0.2494	294.8	0.2
0.1512	292.0	0.1	0.3481	308.3	0.0
0.1722	291.2	0.1	0.3957	313.4	0.1
0.1805	290.9	0.1	0.5000	323.0	0.1
0.1889	290.6	0.0	0.6013	331.1	−0.2
0.1964	290.4	−0.1	0.6987	337.9	−0.2
0.2094	289.8	0.0	0.8004	344.2	0.2
0.2157	289.6[b]	−0.1	0.8978	350.4	−0.1
0.2289	291.7	0.0			
Fluorene (1) + Diphenylmethane (2)
0.0498	296.3	0.0	0.1788	305.2	−0.2
0.0605	295.9	0.0	0.1981	309.0	0.2
0.0804	295.2	−0.1	0.3003	326.5	0.3
0.0996	294.3	0.1	0.4039	339.5	0.1
0.1096	293.8	0.2	0.5113	350.7	−0.5
0.1213	293.7	−0.3	0.5993	358.4	−0.2
0.1289	293.0[b]	−0.1	0.6987	366.5	0.4
0.1488	297.3	0.5	0.8407	377.0	−0.1
Phenanthrene (1) + Diphenylmethane (2)
0.0502	298.1	0.0	0.2509	305.3	−0.5
0.1305	294.6	0.0	0.2979	312.4	−0.6
0.1496	293.5	0.2	0.3989	324.0	0.6
0.1667	292.8	0.1	0.4974	333.4	0.3
0.1813	292.5[b]	−0.3	0.5981	342.8	0.3
0.1905	294.5	0.2	0.6959	350.8	−0.5
0.2021	297.0	−0.3	0.8969	364.2	0.1

[a] δT/K $= T_{(calc)} - T_{(expt)}$, where $T_{(calc)}$ was calculated from eq 1. [b] Eutectic temperature.

mental SLE data. The reference point of eq 1 for each branch of the liquidus line corresponds to x_1^* and T^*, which are the properties of the pure constituent compounds. Table 4 presents the best-fitted coefficients and the average absolute deviations (AAD) of the data correlation. The deviations of the calculated temperatures from the experimental values, δT, are listed in Tables 2 and 3.

Table 4. Correlation of Solid-Disappearance Temperatures with Eq 1

$x_{1,min}$	$x_{1,max}$	x_1^*	T^* (K)	b_1	b_2	b_3	b_4	AAD[a] (%)
				Acenaphthene (1) + Dibenzofuran (2)				
0.4216	1.0	1.0	366.3	0.1351	−0.0831			0.06
0.0	0.4216	0.0	355.7	−0.1826	0.2156	−0.5286		0.06
				Acenaphthene (1) + Fluorene (2)				
0.5317	1.0	1.0	366.3	0.0926	−0.4834	−1.3918	−1.5762	0.06
0.0	0.5317	0.0	387.5	−0.1632	−0.0840			0.07
				Acenaphthene (1) + Phenanthrene (2)				
0.4849	1.0	1.0	366.3	0.1390	−0.0710			0.03
0.0	0.4849	0.0	372.5	−0.2732	0.7075	−2.2750	2.1540	0.05
				Acenaphthene (1) + Diphenylmethane (2)				
0.1530	1.0	1.0	366.3	0.0809	−0.4313	−0.7537	−0.5674	0.01
0.0	0.1530	0.0	298.2	−0.1433	0.0097			0.02
				Dibenzofuran (1) + Diphenylmethane (2)				
0.2157	1.0	1.0	355.7	0.1188	−0.2731	−0.5313	−0.4786	0.04
0.0	0.2157	0.0	298.2	−0.1235	−0.0323			0.02
				Fluorene (1) + Diphenylmethane (2)				
0.1289	1.0	1.0	387.5	−0.1254	−1.2237	−1.9053	−1.1591	0.09
0.0	0.1289	0.0	298.2	−0.1299	0.0929	−0.8770		0.03
				Phenanthrene (1) + Diphenylmethane (2)				
0.1795	1.0	1.0	372.5	0.2233	0.0939	0.0658	−0.1375	0.08
0.0	0.1795	0.0	298.2	−0.1039	−0.1563			0.01

[a] AAD/% = $(100/n) \sum_{k=1}^{n} |T_{(calc),k} - T_{(expt),k}| T_{(expt),k}$, where n is the number of data points.
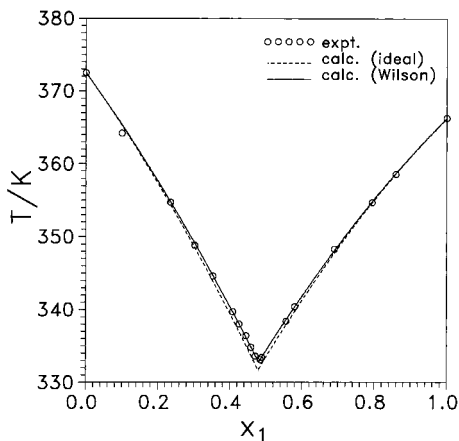

Figure 1. Solid−liquid phase equilibrium diagram for acenaphthene (1) + phenanthrene (2): (○) experimental values; (- - -) ideal solubilities (ideal I); (—) calculated from the Wilson model (Wilson I).

Figure 2. Solid−liquid phase equilibrium diagram for phenanthrene (1) + diphenylmethane (2): (○) experimental values; (- - -) ideal solubilities (ideal I); (—) calculated from the Wilson model (Wilson I).

Solid−Liquid Equilibrium Calculation

The new SLE data were also correlated with activity coefficient models to determine the optimized values of the binary interaction parameters, which are needed for predicting the SLE behavior of multicomponent mixtures. The criterion of SLE for a simple eutectic system in the presence of a solid-phase transition (Coon et al., 1989) can be expressed by

$$\ln a_i = \ln(x_i \gamma_i) = \frac{\Delta_{fus}H_i}{RT_{m,i}}[1 - (T_{m,i}/T)] -$$

$$\frac{\Delta C_{p,i}}{R}[1 - (T_{m,i}/T)] - \frac{\Delta C_{p,i}}{R}\ln(T_{m,i}/T) - \frac{\Delta_p H_i}{RT} + \frac{\Delta_p S_i}{R} \quad (2)$$

where R is the gas constant. a_i, γ_i, $T_{m,i}$, $\Delta C_{p,i}$, $\Delta_{fus}H_i$, $\Delta_p H_i$, and $\Delta_p S_i$ are the activity, the activity coefficient, the melting temperature, the specific heat difference between the subcooled liquid and solid states, the molar enthalpy of fusion, the molar enthalpy change of the solid-phase transition, and the molar entropy change of the solid-phase transition for compound i, respectively. Also in eq 2, x_i is
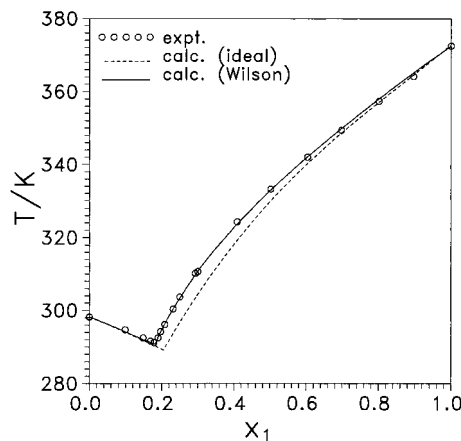
the solid solubility of compound i at temperature T. Among these five studied compounds, only phenanthrene undergoes a solid−solid λ transition. The solid-phase transition starts at 314.0 K and ends at 360.0 K (Coon et al., 1989). Its enthalpy and entropy changes of the solid-phase transition ($\Delta_p H_i$ and $\Delta_p S_i$) are 1300 J·mol^{-1} and 3.85 J·mol^{-1}·K^{-1}, respectively, if the equilibrium temperatures are below 314.0 K (Coon et al., 1989). The values of $\Delta_p H_i$ and $\Delta_p S_i$ were estimated by linear interpolation, when the equilibrium temperatures are within the temperature range of the λ-type transition (314.0 K < T < 360.0 K). All the properties needed in the SLE calculations are given in Table 1.

By assuming that $\gamma_i = 1$, x_i in eq 2 corresponds to the ideal solubility of the solid. Table 5 reports the calculated results with the ideal solution assumption. Two different treatments were used in the ideal solubility calculations. Case I (denoted as ideal I in Table 5) adopted the values of $\Delta C_{p,i}$, $\Delta_p H_i$, and $\Delta_p S_i$ from Table 1, while case II (denoted as ideal II in Table 5) assumed these values to be zero. The accuracy of predictions with ideal II (i.e., when the last four terms on the right-hand side of eq 2 are neglected)

Table 5. Results of SLE Calculations

			Wilson I		Wilson II		NRTL I		NRTL II	
mixture (1) + (2)	ideal I AAD[a] (%)	ideal II AAD[a] (%)	$\lambda_{12} - \lambda_{11}$ $\lambda_{21} - \lambda_{22}$ (J·mol^{-1})	AAD[a] (%)	$\lambda_{12} - \lambda_{11}$ $\lambda_{21} - \lambda_{22}$ (J·mol^{-1})	AAD[a] (%)	$g_{12} - g_{22}$ $g_{21} - g_{11}$ (J·mol^{-1})	AAD[a] (%)	$g_{12} - g_{22}$ $g_{21} - g_{11}$ (J·mol^{-1})	AAD[a] (%)
acenaphthene + dibenzofuran	0.27	0.20	2149.0 −1335.2	0.07	2331.2 −1509.2	0.07	−1120.1 1808.8	0.07	−1232.9 1868.0	0.07
acenaphthene + fluorene	0.14	0.12	−1428.6 2418.7	0.07	−1365.2 2215.5	0.07	2266.6 −1487.8	0.07	2098.0 −1432.6	0.07
acenaphthene + phenanthrene	0.27	0.20	1135.3 −804.9	0.08	785.2 −572.0	0.10	−391.3 694.0	0.08	−613.4 887.2	0.10
acenaphthene + diphenylmethane	0.15	0.09	11.3 127.0	0.06	−1006.9 1463.9	0.06	588.1 −428.7	0.06	1459.6 −1161.9	0.06
dibenzofuran + diphenylmethane	0.57	0.50	1682.9 −815.2	0.13	−828.1 1811.8	0.10	−362.1 1156.2	0.13	499.7 48.9	0.14
fluorene + diphenylmethane	0.40	0.38	−537.5 1045.0	0.08	−586.0 1097.9	0.08	1124.8 −633.0	0.08	332.6 −11.0	0.09
phenanthrene + diphenylmethane	1.22	1.38	582.0 126.9	0.12	1196.9 −286.4	0.12	−121.9 875.7	0.13	−199.9 1102.4	0.12

^{a} AAD/% $= (100/n) \sum_{k=1}^{n} |T_{(calc),k} - T_{(expt),k}|/T_{(expt),k}$, where n is the number of data points.

was found to be even better than that of predictions with ideal I, except for phenanthrene + diphenylmethane. The ideal solution assumption, however, is oversimplified for some mixtures containing diphenylmethane. Figure 2, for example, shows that this assumption leads to a substantial underestimation of the equilibrium temperatures over the range of x_1 from 0.2 to 0.6 for phenanthrene + diphenylmethane.

The nonideality of the liquid phase should be taken into account in the SLE calculations to represent quantitatively the liquidus line over the entire composition range. For this reason, the Wilson (Wilson, 1964) and the NRTL (Renon and Prausnitz, 1968) models were used in this study to calculate the activity coefficient for each constituent compound in the mixture. The Wilson model is given by

$$\ln \gamma_i = 1 - \ln\left(\sum_{j=1}^{c} x_j \Lambda_{ij}\right) - \sum_{k=1}^{c}\left(x_k \Lambda_{ki} \Big/ \sum_{j=1}^{c} x_j \Lambda_{kj}\right) \quad (3)$$

with

$$\Lambda_{ij} = (V_{L,j}/V_{L,i}) \exp[-(\lambda_{ij} - \lambda_{ii})/RT] \quad (4)$$

where c is the number of components and $V_{L,i}$ is the liquid molar volume of component i. The molar volumes, listed in Table 1, were estimated from the modified Rackett model (Spencer and Danner, 1972). The NRTL model is given by

$$\ln \gamma_i = \frac{\sum_{j=1}^{c} \tau_{ji} G_{ji} x_j}{\sum_{k=1}^{c} G_{ki} x_k} + \sum_{j=1}^{c} \frac{x_j G_{ij}}{\sum_{k=1}^{c} G_{kj} x_k}\left(\tau_{ij} - \frac{\sum_{r=1}^{c} x_r \tau_{rj} G_{rj}}{\sum_{k=1}^{c} G_{kj} x_k}\right) \quad (5)$$

with

$$G_{ji} = \exp\left[\frac{-\alpha(g_{ji} - g_{ii})}{RT}\right] \quad (6)$$

where the nonrandomness parameter α was set to 0.5 for all binaries. After the parameters $(\lambda_{12} - \lambda_{11})$ and $(\lambda_{21} - \lambda_{22})$ of the Wilson model or $(g_{12} - g_{22})$ and $(g_{21} - g_{11})$ of the NRTL model are specified, the equilibrium temperature T at a given x_i can be solved from eq 2 by an iterative procedure. The optimal values of the model parameters were obtained by minimizing the objective function π over the entire composition range:

$$\pi = \sum_{k=1}^{n} [|T_{(calc),k} - T_{(expt),k}|/T_{(expt),k}]/n \quad (7)$$

where n is the number of data points. Table 5 lists the correlated results, indicating that both the Wilson and the NRTL models describe the nonideality of the liquid phases equally well. The calculated results from the Wilson model are compared with the experimental values and with the ideal solubilities in Figures 1 and 2 for acenaphthene + phenanthrene and phenanthrene + diphenylmethane, respectively. These two figures and the tabulated values in Table 5 demonstrate that better representation was achieved when the activity coefficient model was used to account for the nonideality of the liquid phase. Furthermore, Table 5 also shows that the calculated results are comparable between case I and case II for both models.

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