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Solid−Liquid Equilibria of *n*-Alkanes in *N*,*N*-Dimethylformamide

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Solid−liquid equilibria of 11 binary mixtures of *n*-alkanes (octadecane, nonadecane, eicosane, heneicosane, docosane, tricosane, tetracosane, pentacosane, hexacosane, heptacosane, and octacosane) in *N*,*N*-dimethylformamide (DMF) have been measured from 275 K to the melting temperatures of the *n*-alkane. The solubility results have been correlated by the Redlich−Kister (two and three parameters) equation and the NRTL equation.

Introduction

The knowledge of solid−liquid equilibria is of technical and theoretical interest. As it forms the basis for any crystallization process, it is important in different parts of the chemical industry such as separation of thermolabile compounds, refrigeration processes, pipeline design, and obtaining high-purity products (Wynn, 1992). Several authors (Fiege et al., 1996; Hofman et al., 1998; Rolinska et al., 1997; Domanska et al., 1993; Domanska and Venkatesu, 1998a,b) have investigated the optimization and design of appropriate solvents to be used as separating agents for extractive distillation and separations of the petroleum fractions into their pure components. Addition of the solvent DMF to the model mixtures makes the phase behavior fairly complex, so that in order to design efficient large-scale separation processes in the petroleum industry, experimental information on phase equilibria and related thermodynamic properties is required (Walas, 1985). The theoretical importance of solid−liquid equilibria is connected with the development of prediction models. Middle-distillate fuels contain high molar-mass normal alkanes with chain lengths in range C_{18} to C_{28}. These hydrocarbons crystallize as wax crystals at low temperatures, thus posing a constantly recurring problem in the petroleum industry. Refiners have sought many solutions, most of them relying on purely empirical hypotheses. The polymorphism, the melting solid transition temperatures, and the associated enthalpy changes of the long-chain, normal primary alkanols and normal alkanes have been a source of controversy for may decades. The solid−solid transformations for long-chain compounds were confirmed by earlier solubility measurements for 1-alkanols (Domanska and Rolinska, 1993; Domanska 1996) and *n*-alkanes (Domanska and Kinaz, 1980; Domanska and Domanski, 1991).

This paper presents experimental solid−liquid equilibrium data for *N*,*N*-dimethylformamide with octadecane, nonadecane, eicosane, heneicosane, docosane, tricosane, tetracosane, pentacosane, hexacosane, heptacosane, and octacosane measured by a dynamic method. The experimental data are correlated by using the Redlich−Kister (two and three parameters) equation and the NRTL equation.

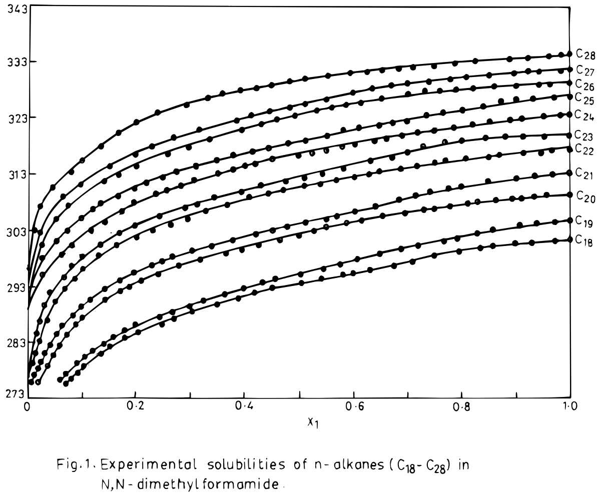

Figure 1.

Experimental Section

Materials. Commercially available *n*-alkanes were directly used without purification. The purity (98%−99%, depending on chain length *n*) was used for solid−liquid equilibrium (SLE) measurements. Hexacosane was taken from Aldrich, and other *n*-alkanes were from Fluka AG. *N*,*N*-Dimethylformamide (Fluka) was purified as described previously (Ramadevi et al., 1996; Venkatesu and Rao, 1998). The physical properties of the pure components are collected in Table 1.

Procedure. Solid−liquid equilibrium temperatures were determined using a dynamic method (Domanska, 1986). Mixtures were prepared by mass, the error in mole fraction being estimated as $<5 \times 10^{-4}$. Mixtures of solute and solvent were heated very slowly (at <2 K h^{-1} near the equilibrium temperature) with continuous stirring inside a Pyrex glass cell, placed in a thermostat. The crystal disappearance temperatures, detected visually, were measured with a calibrated, totally immersed thermostat. The accuracy of the thermometer temperature measurement was ±0.01 K, and the experimental error was 0.05 K.

Results and Discussion

The experimental values of the temperatures at which the solid−solid phase transitions occur (all of them deter-

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Table 1. Physical Constants of Pure Compounds: T_{m1}, Melting Point (This Work); ΔH_{m1} Molar Heat of Fusion; ΔH_{tr1}, Molar Heat of Transition ($\alpha \rightarrow \beta$); ΔC_{p1}, Heat Capacity Change at the Melting Temperature; and T_{tr1}, Transition Temperature (This Work)

substance	T_{m1}/K	T_{tr1}/K	T^{d}_{tr1}/K	ΔH_{m1}/ kJ·mol^{-1}	ΔH_{tr1}/ kJ·mol^{-1}	ΔCp/ J·K^{-1}·mol^{-1}
octadecane	301.65			61.71[a]		50.0[b]
nonadecane	304.95	302.85		45.83[a]	13.81[a]	−332.8[b]
eicosane	309.80	309.55	309.35	66.93[c]	18.39[c]	54.0[b]
heneicosane	313.80	311.85	310.65	47.73[d]	15.49[d]	−100.0[b]
docosane	317.65	314.45	314.15	39.76[b]	36.35[b]	58.5[b]
tricosane	320.50	318.05	318.65	54.01[b]	21.77[b]	0.1[b]
tetracosane	323.80	320.65	321.25	57.31[b]	27.68[b]	66.6[b]
pentacosane	327.10	322.60	322.15	57.78[b]	26.08[b]	71.0[b]
hexacosane	329.70	326.50	326.45	63.92[b]	30.36[b]	78.3[b]
heptacosane	332.00	326.40	326.15	60.46[d]	28.97[d]	91.5[b]
octacosane	334.65	331.75	331.15	66.52[b]	33.66[b]	118.90[b]

[a] Van Oort and White (1985). [b] From linear extrapolation of data for odd (1967) and even-numbered alkanes (1991). [c] Claudy and Letoffe (1991). [d] Schaerer et al. (1955).

Table 2. Experimental Solid-Liquid Equilibrium Temperatures $T_{\alpha 1}$, $T_{\beta 1}$ for *n*-Alkanes(1) with *N*,*N*-Dimethylformamide (DMF) (2)

x_1	$T_{\beta 1}$/K	x_1	$T_{\beta 1}$/K	x_1	$T_{\beta 1}$/K	$T_{\alpha 1}$/K	x_1	$T_{\beta 1}$/K	$T_{\alpha 1}$/K
Octadecane (**1**) + DMF (**2**)									
0.0728	276.45	0.2522	286.90	0.5429	244.95		0.8359	300.25	
0.0842	277.35	0.2742	287.95	0.5842	295.65		0.8625	300.75	
0.0908	278.35	0.3074	288.95	0.6022	295.95		0.8994	301.15	
0.1028	279.40	0.3542	290.75	0.6324	296.40		0.9324	301.25	
0.1356	281.55	0.3924	291.45	0.6749	297.15		0.9530	301.35	
0.1542	282.95	0.4218	292.45	0.7152	297.85		0.9730	301.55	
0.1822	284.35	0.4529	293.40	0.7540	298.85		1.0000	301.65	
0.2042	285.65	0.5022	294.75	0.7952	299.45				
Nonadecane (**1**) + DMF (**2**)									
0.0653	276.65	0.2066	286.35	0.4621	294.55		0.7714	301.65	
0.0773	278.05	0.2439	287.75	0.5024	295.15		0.7928	301.85	
0.0923	279.65	0.2735	288.55	0.5459	296.15		0.8311		302.85
0.1055	280.55	0.2994	289.75	0.5769	297.35		0.8629		303.55
0.1249	281.85	0.3276	290.85	0.6135	298.35		0.9051		304.25
0.1426	283.05	0.3658	292.05	0.6518	299.25		0.9375		304.75
0.1601	284.15	0.4015	292.85	0.6929	300.15		0.9702		304.85
0.1833	285.15	0.4375	293.75	0.7241	300.75		1.0000		304.95
Eicosane (**1**) + DMF (**2**)									
0.0246	276.25	0.1942	294.05	0.4670	301.85		0.7798	307.95	
0.0359	279.35	0.2122	294.55	0.5026	302.75		0.8162	308.45	
0.0460	281.25	0.2446	295.85	0.5261	304.05		0.8346	308.95	
0.0568	283.05	0.2825	297.15	0.5573	304.40		0.8667	309.35	
0.0703	284.75	0.3049	297.85	0.5925	305.05		0.8971	309.45	
0.0865	286.65	0.3262	298.40	0.6185	305.55		0.9136		309.55
0.1025	288.35	0.3659	299.35	0.6556	306.15		0.9369		309.65
0.1213	289.55	0.3986	300.25	0.6978	306.75		0.9572		309.75
0.1460	291.25	0.4265	300.90	0.7342	307.45		1.0000		309.80
0.1726	292.75			0.7536	307.55				
Heneicosane (**1**) + DMF (**2**)									
0.0092	276.25	0.1186	290.75	0.3804	301.85		0.7184	309.65	
0.0148	277.95	0.1446	292.90	0.4263	303.05		0.7406	310.35	
0.0204	279.05	0.1529	293.55	0.4574	303.65		0.7792	311.05	
0.0296	279.75	0.1732	294.35	0.4786	304.35		0.8026	311.35	
0.0345	281.55	0.1940	296.05	0.5129	304.95		0.8578		311.85
0.0469	283.15	0.2304	296.95	0.5530	305.65		0.9026		312.75
0.0540	284.95	0.2518	297.95	0.5724	305.90		0.9218		312.85
0.0609	286.35	0.2742	298.55	0.6072	306.85		0.9426		313.15
0.0799	288.05	0.3063	299.75	0.6422	307.45		0.9659		313.45
0.0962	289.75	0.3424	300.75	0.6852	308.75		1.0000		313.80
Docosane (**1**) + DMF (**2**)									
0.0085	276.25	0.1021	296.55	0.4459	309.85		0.7504		315.55
0.0112	279.25	0.1324	298.75	0.4724	310.65		0.7740		316.05
0.0182	281.15	0.1685	300.75	0.5110	311.15		0.8059		316.35
0.0229	283.05	0.2076	302.35	0.5364	311.65		0.8523		316.65
0.0286	284.85	0.2304	303.55	0.5705	312.15		0.8724		316.95
0.0359	287.05	0.2652	304.45	0.6042	312.75		0.9184		317.35
0.0427	288.85	0.3005	305.65	0.6318	313.25		0.9475		317.45
0.0535	290.75	0.3325	306.85	0.6522	313.75		0.9690		317.55
0.0679	292.85	0.3744	308.05	0.6859		314.45	1.0000		317.65
0.0858	294.55	0.4129	309.15	0.7214		314.65			

Table 2. (Continued)

x_1	$T_{\beta1}$/K	x_1	$T_{\beta1}$/K	x_1	$T_{\beta1}$/K	$T_{\alpha1}$/K	x_1	$T_{\beta1}$/K	$T_{\alpha1}$/K
Tricosane (**1**) + DMF (**2**)									
0.0082	277.40	0.1041	298.75	0.3957	309.95		0.7328		318.05
0.0094	279.45	0.1352	300.85	0.4322	311.05		0.7722		318.95
0.0126	281.85	0.1540	301.90	0.4759	311.85		0.8048		319.05
0.0195	284.55	0.1702	302.65	0.5135	312.65		0.8451		319.95
0.0294	287.05	0.2034	304.15	0.5469	313.15		0.8847		320.05
0.0349	289.65	0.2419	305.75	0.5815	314.40		0.9137		320.15
0.0443	292.25	0.2922	307.15	0.6318	315.75		0.9562		320.35
0.0674	294.75	0.3165	307.85	0.6703	316.40		0.9744		320.40
0.0849	296.65	0.3479	309.25	0.7039	317.15		1.0000		320.50
Tetracosane (**1**) + DMF (**2**)									
0.0044	276.05	0.0618	299.05	0.3842	314.05		0.6924		320.65
0.0053	278.05	0.0809	301.65	0.4020	314.65		0.7249		320.95
0.0068	280.05	0.1158	303.55	0.4224	315.15		0.7553		321.40
0.0072	281.85	0.1402	304.75	0.4594	316.15		0.7879		321.75
0.0090	283.75	0.1780	306.95	0.4989	316.75		0.8246		322.25
0.0135	286.40	0.1985	308.05	0.5280	317.25		0.8521		322.85
0.0155	288.25	0.2394	309.75	0.5542	317.55		0.8903		323.05
0.0180	289.95	0.2622	310.75	0.5799	318.15		0.9182		323.40
0.0199	293.15	0.2872	311.17	0.6041	318.65		0.9502		323.65
0.0289	295.65	0.3164	312.15	0.6398	319.25		0.9826		323.75
0.0482	297.15	0.3574	313.65	0.6637	319.75		1.0000		323.80
Pentacosane (**1**) + DMF (**2**)									
0.0011	276.05	0.0334	298.40	0.3140	314.65		0.7028		323.00
0.0016	277.65	0.0496	301.25	0.3514	315.90		0.7351		323.55
0.0020	278.55	0.0740	303.15	0.3825	316.65		0.7689		324.25
0.0027	279.95	0.1029	305.65	0.4121	317.40		0.8025		324.75
0.0036	281.05	0.1289	307.05	0.4453	317.75		0.8398		325.60
0.0045	282.05	0.1349	307.95	0.4816	318.90		0.8772		326.25
0.0063	283.95	0.1674	309.55	0.5112	319.35		0.9004		326.65
0.0074	286.05	0.2025	310.75	0.5503	320.05		0.9348		326.90
0.0096	287.75	0.2246	311.85	0.5814	321.05		0.9689		327.05
0.0124	289.65	0.2529	312.65	0.6129	321.40		1.0000		327.10
0.0178	293.90	0.2756	313.75	0.6493	322.05				
0.0218	297.00			0.6759		322.60			
Hexacosane (**1**) + DMF (**2**)									
0.0013	276.30	0.0458	303.00	0.2976	318.05		0.6996		327.55
0.0018	279.10	0.0336	300.00	0.3349	319.55		0.7345		328.00
0.0027	282.15	0.0458	305.35	0.3615	320.35		0.7618		328.25
0.0035	283.35	0.0536	307.65	0.3962	321.25		0.7942		328.60
0.0046	284.85	0.0774	308.75	0.4318	322.15		0.8190		328.75
0.0057	287.30	0.0921	310.35	0.4626	323.05		0.8616		328.95
0.0087	289.95	0.1134	312.00	0.4942	323.75		0.9021		329.15
0.0095	291.05	0.1431	313.20	0.5318	324.65		0.9436		329.35
0.0125	293.45	0.1724	314.55	0.5626	325.15		0.9646		329.55
0.0189	296.10	0.2012	314.55	0.5849	325.65		1.0000		329.70
0.0248	298.75	0.2356	315.90	0.6142		326.40			
0.0336	300.85	0.2642	316.90	0.6534		326.90			
Heptacosane (**1**) + DMF (**2**)									
0.0009	275.75	0.0265	302.55	0.2711	319.85		0.6714		328.85
0.0011	278.60	0.0346	305.65	0.3162	321.15		0.7018		329.35
0.0015	279.75	0.0459	307.60	0.3569	322.15		0.7432		329.85
0.0018	280.15	0.0676	309.25	0.3878	322.85		0.7814		330.30
0.0020	281.40	0.0806	310.40	0.4106	323.95		0.8118		330.75
0.0028	284.05	0.0984	311.40	0.4486	324.55		0.8540		331.00
0.0034	285.80	0.1259	312.70	0.4818	325.35		0.8956		331.40
0.0043	288.15	0.1506	314.75	0.5159	325.90		0.9218		331.70
0.0055	290.35	0.1842	315.75	0.5425		326.40	0.9411		331.85
0.0086	293.70	0.2025	316.55	0.5776		327.05	0.9814		331.90
0.0100	297.15	0.2306	318.30	0.6018		327.85	1.0000		332.00
0.0196	299.55			0.6389		328.30			
Octacosane (**1**) + DMF (**2**)									
0.0005	276.65	0.0158	304.45	0.3318	326.65		0.6864		332.05
0.0009	280.00	0.0218	306.85	0.3642	327.75		0.7114		332.35
0.0012	282.65	0.0322	308.70	0.3917	328.25		0.7503		332.65
0.0015	285.35	0.0518	311.15	0.4219	328.75		0.7942		333.05
0.0018	286.75	0.0849	313.80	0.4500	329.20		0.8269		333.55
0.0026	290.00	0.1028	315.80	0.4803	329.95		0.8614		333.75
0.0035	291.85	0.1369	318.25	0.5146	330.15		0.8910		333.95
0.0046	293.95	0.1747	320.65	0.5518	330.95		0.9225		334.05
0.0058	295.40	0.2018	322.55	0.5920	331.35		0.9641		334.40
0.0070	297.35	0.2408	324.25	0.6269		331.75	0.9822		334.55
0.0089	299.55	0.2906	325.80	0.6576		331.85	1.0000		334.65

Table 3. Correlation of the Solubility Temperatures of *n*-Alkanes in DMF by the Redlich−Kister and NRTL Equations: Values of Paramenters and RMSDs[a]

		parameters					
		R−K[b]	R−K[c]	NRTL	deviations σ_T/K		
n-alkane	no. of data points	A_0, A_1/J·mol^{-1}	A_0, A_1, A_2/J·mol^{-1}	A_1, A_2/J·mol^{-1}	R−K[b]	R−K[c]	NRTL
octadecane	31	667.17 −198.66	649.21 −77.73 419.89	3182.50 −1917.43	0.39	0.30	0.37
nonadecane	31	610.75 −817.96	590.96 −527.93 810.01	6750.56 −3626.00	0.65	0.38	0.49
eicosane	37	−615.09 168.02	−639.60 341.07 346.70	909.58 −1440.58	0.48	0.45	0.49
heneicosane	39	712.41 −1304.31	612.30 −419.64 1415.91	8299.68 −4101.55	1.72	1.32	1.39
docosane	38	1323.97 499.88	1181.29 1237.22 1216.04	−2790.96 6101.09	1.01	0.59	0.91
tricosene	35	579.60 −357.59	490.35 131.44 784.16	1003.94 −198.76	0.93	0.66	1.00
tetracosane	43	617.92 2.53	545.05 557.49 763.45	249.53 375.33	1.08	0.96	1.08
pentacosane	44	690.68 −690.31	545.87 168.53 1206.64	2954.22 −1476.56	1.22	0.87	1.30
hexacosane	43	781.71 637.23	645.27 1400.91 1095.29	−3867.06 8223.67	0.88	0.55	0.70
heptacosane	45	1424.21 252.56	1208.32 968.65 1135.76	−1765.29 4181.66	1.15	0.79	1.11
octacosane	43	1545.52 933.07	1443.88 1390.82 650.43	−176.77 1183.19	1.04	0.95	1.40

[a] Root-mean-square deviations. [b] Redlich−Kisher eq 2 parameters. [c] Redlich−Kister eq 3 parameters.

mined graphically) are collected in Table 1, and they are very close to the pure compounds' transition temperatures (Schaerer, 1955). The experimental results of the solid−liquid equilibrium temperatures (T) and the mole fractions (x_1) of the *n*-alkanes of two crystallographic forms α and β for the investigated systems are listed in Table 2 and are graphically represented in Figure 1. The solubilities of *n*-alkanes in DMF, investigated in this work, are characterized mainly by the following: (i) positive deviations from ideality were found, which means that the solubility is lower than the ideal one and the activity coefficients of the solute are positive ($\gamma_1 > 1$); (ii) the solubilities of *n*-alkanes in DMF decrease with increasing length of the carbon chain of the alkane. The solubility of a solute phase is expressed in a very general manner as follows.

$$-\ln x_1 = \frac{\Delta H_{\mathrm{m1}}}{R}\left(\frac{1}{T} - \frac{1}{T_{\mathrm{m1}}}\right) - \frac{\Delta C_{\mathrm{pm1}}}{R}\left(\ln\frac{T}{T_{\mathrm{m1}}} + \frac{T_{\mathrm{m1}}}{T} - 1\right) + \ln\gamma_1 \quad (1)$$

where x_1, γ_1, ΔH_{m1}, ΔC_{mp1}, T_{m1}, and T stand for mole fraction, activity coefficient, enthalpy of fusion, difference in solute heat capacity between the solid and liquid at the melting point, melting of the solute, and equilibrium temperature, respectively. If the solid−solid transition occurs prior to fusion, an additional term must be added to the right-hand side of eq 1 (Weimer and Prausnitz, 1965; Choi and McLaughlin, 1983). The solubility equation for temperatures below that of the phase transition must include the effect of the transition. The result for the first-order transition is

$$-\ln x_1 = \frac{\Delta H_{\mathrm{m1}}}{R}\left(\frac{1}{T} - \frac{1}{T_{\mathrm{m1}}}\right) - \frac{\Delta C_{\mathrm{pm1}}}{R}\left(\ln\frac{T}{T_{\mathrm{m1}}} + \frac{T_{\mathrm{m1}}}{T} - 1\right) + \frac{\Delta H^{1}_{\mathrm{tr}_1}}{R}\left(\frac{1}{T} - \frac{1}{T^{1}_{\mathrm{tr}}}\right) + \ln\gamma_1 \quad (2)$$

where $\Delta H^{1}{}_{\mathrm{tr}_1}$ and $T^{1}{}_{\mathrm{tr}_1}$ stand for enthalpy of transition and transition temperature of the solute, respectively.

The following correlation equations were used in solid−liquid equilibria calculations: Redlich−Kister equations (with two and three constants) and NRTL equations. The following equations for the excess Gibbs free energy were used: the Redlich−Kister expansion in the form

$$G^{\mathrm{E}} = x_1 x_2 \sum_{i=0}^{m-1} A(x_1 - x_2)^i \quad (3)$$

where m is the number of adjustable constants (A_i) which are temperature and composition independent, and the NRTL equation (Renon and Prausnitz, 1968).

$$G^{\mathrm{E}}/RT = x_1 x_2\left[\frac{A_1\exp(-\alpha A_1/RT)}{x_1 + x_2\exp(-\alpha A_1/RT)} + \frac{A_2\exp(-\alpha A_2/RT)}{x_2 + x_1 + \exp(-\alpha A_2/RT)}\right] \quad (4)$$

Two energetic parameters (A_1, A_2) have been fitted while the nonrandomness parameter α was fixed and set equal to 0.2.

The root-mean-square deviation in temperature defined by eq 5 was used as a measure of the correlation accuracy.

$$\sigma_T = \left[\sum_{i=1}^{n} \frac{(T_i^{\mathrm{cal}} - T_i)^2}{n}\right]^{1/2} \qquad (5)$$

where T_i^{cal} and T_i are respectively the calculated and experimental temperatures of the ith point and n is the number of experimental points, excluding that of the melting point. The calculated values of the equation parameters and the corresponding root-mean-square deviations are presented in Table 3. Generally, the parameters obtained using the Redlich−Kister (two and three parameters) and the NRTL equations are comparable with each other along the homologous series of alkanes from octadecane to octacosane. Analysis of the data indicates that the agreement between measured SLE temperature values and those calculated from the correlation equations is satisfactory in all the systems. For the 11 systems, the best results were obtained with the Redlich−Kister three-parameter equation.

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