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Isothermal Phase Equilibria and Excess Molar Enthalpies for Binary Systems with Dimethyl Ether at 323.15 K

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Isothermal vapor−liquid equilibria (VLE) and excess molar enthalpies (H^E) for the binary systems of dimethyl ether (DME) with *n*-decane, *n*-dodecane, 1-octene, 1-decene, benzene, toluene, and water were measured at 323.15 K with a computer-driven static apparatus and an isothermal flow calorimeter, respectively. None of the measured binary systems show azeotropic behavior at this temperature. The system DME + water shows a miscibility gap. The experimental VLE and H^E data were correlated with different G^E models and the Redlich−Kister polynomial.

Introduction

Dimethyl ether (DME), the simplest of all ethers, has recently been gaining attention worldwide as a clean-burning substitute for liquefied petroleum gas, liquefied natural gas, diesel, and gasoline as a powerful generation fuel. Besides, DME is used as an aerosol propellant in the cosmetic industry to replace CFC propellants because it is nontoxic. Additionally, DME may be used in gas turbines for power generation, for household purposes (cooking, heating), and also as a hydrogen source for fuel cells.[1]

DME is physically similar to liquefied petroleum gas which primarily contains propane and butane, and its heating characteristics are similar to natural gas. It easily becomes liquid and can be easily stored and shipped because it is not a cryogenic liquid. It boils at 298 K under atmospheric pressure or condenses at less than 600 kPa at 298 K.[2]

DME is currently manufactured by dehydration of methanol derived from natural gas, coal, or biomass. Even though DME is available today in relatively small quantities in the field of the propellant market, large and economical supplies of DME for power generation as a fuel alternative should be prepared. It means at least 1 to 2 million t/y DME production capacity, which requires a scale-up by a factor of 100 of existing plants.[2] Therefore, the thermodynamic behavior of systems with DME is of great interest.

As a continuation of our research program involving measurement of phase equilibria and mixture properties, we reported vapor−liquid equilibria (VLE) and H^E data for the systems of DME + C_1 to C_4 alkanols.[3] In the present work, we report the isothermal VLE and H^E at 323.15 K for the systems of DME with some hydrocarbon and water, i.e., decane, dodecane, 1-octene, benzene, toluene, and water. In the case of H^E, the system DME + 1-decene was additionally measured. As far as we know, there is no measured data for the same systems and conditions except VLE data for the system DME + water.[4]

Experimental Section

Materials. All the chemicals including DME were used as commercial grade and supplied by Sigma-Aldrich Co. Ltd. The mass fraction purity of DME, specified by the manufacturer, was greater than $w = 99.5$ %. The purity of other chemicals was better than $w = 99.8$ % by gas chromatographic analysis. The chemicals except DME were dried over molecular sieves with a pore diameter of 0.4 nm and used without any further purification. Besides, doubly distilled water of deionized water was used for the measurement.

Apparatus and Procedure. A computer-driven static apparatus was used for the isothermal VLE measurements ($P-x$ data). The evacuated VLE cell kept at constant temperature in a thermostatic oil bath was charged with purified and degassed sample compounds using motor-driven injection pumps with automatic valves. It has been previously described in more detail elsewhere.[5,6] The pressure in the equilibrium cell was monitored using a Digiquartz pressure sensor (model 245A, Paroscientific). The temperature was measured with a Pt100 resistance thermometer (model 1506, Hart Scientific).

The overall compositions were determined from the known quantities of liquids injected into the equilibrium cell. Then, true liquid-phase compositions were obtained by solving mass and volume balance equations taking into account the vapor−liquid equilibrium ($P-x$) behavior. The experimental uncertainties of this device are as follows: $\sigma(T) = 0.03$ K, $\sigma(P) = 20$ Pa $+ 0.0001$ (P/Pa), and $\sigma(x_i) = 0.0005$.

For the measurement of H^E (excess enthalpy or heat of mixing) data, a commercial isothermal flow calorimeter (model 7501, Hart Scientific) was used. This calorimeter consists of two solvent pumps (ISCO, LC 2600, 260 cm^3), a temperature-regulated flow cell equipped with a pulsed heater, and a Peltier cooler. The Peltier cooler operates at constant power, producing constant heat loss from the calorimeter cell. The temperature of the cell is kept constant by adjusting the frequency of the pulsed heater to compensate for the cooling from the Peltier cooler and the heat of mixing effect. The uncertainty of the experimental values was estimated as follows: $\sigma(T) = 0.03$ K, $\sigma(H^E) = 2\ \text{J}\cdot\text{mol}^{-1} + 0.01\ (H^E/\text{J}\cdot\text{mol}^{-1})$, $\sigma(x_i) = 0.0001$. This apparatus has already been described in detail.[7]

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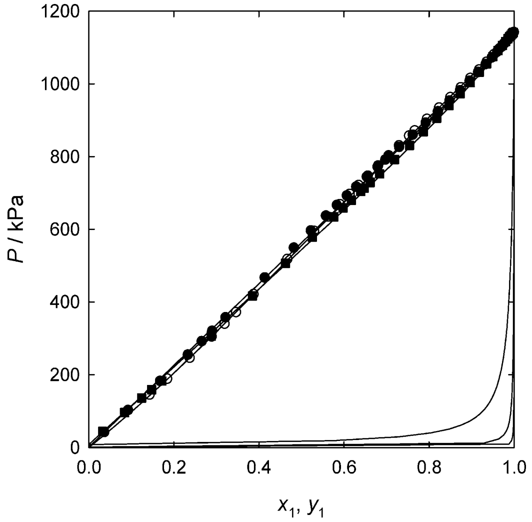

Figure 1. Isothermal vapor−liquid equilibria for the binary systems of DME (1) + *n*-decane (2), + *n*-dodecane (2), and + 1-octene (2) at 323.15 K: ●, DME + *n*-decane; ○, DME + *n*-dodecane; ■, DME + 1-octene. Solid curves were calculated from the van Laar equation.

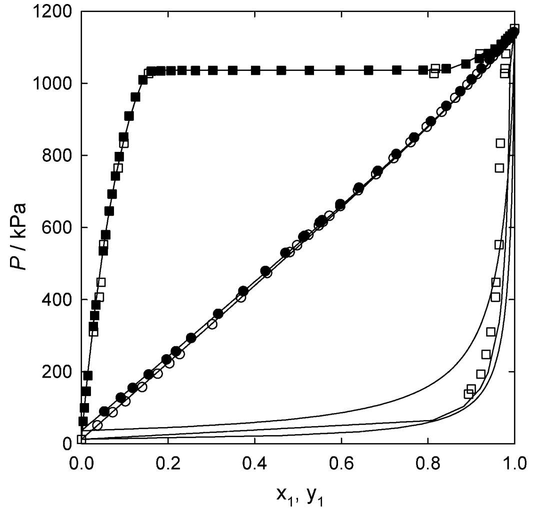

Figure 2. Isothermal vapor−liquid equilibria for the binary systems of DME (1) + benzene (2), + toluene (2), and + water (2) at 323.15 K: ●, DME + benzene; ○, DME + toluene; ■, DME + water; □, DME + water of Pozo et al.[4] Solid curves were calculated from the NRTL equation.

Table 1. Isothermal Vapor−Liquid Equilibria for the Binary Systems of DME (1) + *n*-Decane (2), DME (1) + *n*-Dodecane (2), DME (1) + 1-Octene (2), DME (1) + Benzene (2), DME (1) + Toluene (2), and DME (1) + Water (2) at 323.15 K

P/kPa	x_1	P/kPa	x_1	P/kPa	x_1	P/kPa	x_1	P/kPa	x_1	P/kPa	x_1
DME (1) + *n*-Decane (2)											
1.18	0.0000	357.84	0.3215	717.49	0.6285	893.13	0.7919	1088.67	0.9601	1134.52	0.9944
42.14	0.0361	467.48	0.4133	744.78	0.6535	924.24	0.8206	1101.95	0.9702	1137.61	0.9966
102.84	0.0923	549.53	0.4820	772.11	0.6787	954.00	0.8476	1112.74	0.9783	1140.52	0.9987
183.17	0.1674	596.50	0.5219	791.77	0.6970	982.15	0.8725	1121.47	0.9848	1142.31	1.0000
255.37	0.2328	637.43	0.5573	799.59	0.7043	1008.53	0.8952	1127.49	0.9892		
292.52	0.2655	666.77	0.5831	826.60	0.7296	1054.42	0.9331	1088.67	0.9601		
320.81	0.2900	693.02	0.6065	860.44	0.7614	1072.61	0.9476	1131.75	0.9923		
DME (1) + *n*-Dodecane (2)											
0.20	0.0000	372.26	0.3461	697.09	0.6133	857.44	0.7530	1015.92	0.8970	1122.96	0.9850
145.15	0.1428	421.66	0.3873	721.75	0.6342	871.67	0.7659	1039.51	0.9175	1132.65	0.9922
189.14	0.1843	518.33	0.4666	747.22	0.6559	903.62	0.7950	1061.01	0.9357	1137.67	0.9959
246.81	0.2371	595.76	0.5296	775.24	0.6801	934.56	0.8234	1080.83	0.9520	1140.36	0.9978
305.36	0.2889	634.84	0.5616	803.24	0.7046	963.44	0.8498	1096.75	0.9648	1142.39	0.9993
340.40	0.3191	669.18	0.5900	830.81	0.7290	990.60	0.8744	1110.92	0.9758	1143.39	1.0000
DME (1) + 1-Octene (2)											
8.30	0.0000	416.31	0.3845	704.14	0.6391	905.31	0.8181	1091.70	0.9621	1139.71	0.9967
43.95	0.0327	506.29	0.4624	713.33	0.6475	940.23	0.8468	1105.58	0.9721	1141.77	0.9981
96.08	0.0842	578.09	0.5254	728.98	0.6618	973.36	0.8733	1116.41	0.9799	1144.36	1.0000
135.64	0.1243	634.29	0.5756	752.82	0.6836	1003.88	0.8969	1124.16	0.9855		
158.77	0.1476	658.88	0.5979	791.95	0.7192	1031.86	0.9181	1129.61	0.9894		
183.00	0.1716	680.05	0.6171	830.88	0.7540	1054.70	0.9351	1133.45	0.9922		
306.27	0.2877	416.31	0.3845	868.84	0.7871	1074.79	0.9498	1137.53	0.9951		
DME (1) + Benzene (2)											
35.47	0.0000	256.43	0.2180	573.71	0.5122	757.00	0.6842	1011.00	0.9006	1122.89	0.9842
89.84	0.0528	293.28	0.2531	576.98	0.5152	803.47	0.7268	1040.75	0.9232	1130.01	0.9894
127.73	0.0907	360.33	0.3157	614.10	0.5500	849.57	0.7680	1066.21	0.9423	1134.77	0.9929
155.03	0.1182	423.21	0.3735	620.11	0.5556	894.50	0.8067	1087.07	0.9578	1139.56	0.9964
192.04	0.1551	479.24	0.4248	664.53	0.5975	937.20	0.8422	1103.74	0.9701	1141.87	0.9981
233.89	0.1962	529.14	0.4708	710.47	0.6408	977.55	0.8745	1115.01	0.9784	1144.40	1.0000
DME (1) + Toluene (2)											
12.22	0.0000	248.71	0.2268	605.48	0.5478	836.45	0.7599	1093.36	0.9634	1142.00	0.9984
51.06	0.0366	331.39	0.3019	615.75	0.5573	879.23	0.7968	1108.10	0.9740	1144.23	1.0000
87.32	0.0719	405.71	0.3681	632.17	0.5724	920.60	0.8314	1118.20	0.9813		
117.69	0.1016	473.80	0.4286	659.15	0.5975	959.47	0.8627	1125.19	0.9863		
157.19	0.1401	531.81	0.4806	703.05	0.6384	995.82	0.8910	1131.44	0.9908		
194.34	0.1758	550.71	0.4978	747.97	0.6800	1052.62	0.9337	1135.57	0.9938		
223.13	0.2030	579.55	0.5240	792.48	0.7206	1075.16	0.9502	1139.80	0.9968		
DME (1) + Water (2)											
12.29	0.0000	535.17	0.0503	962.25	0.1248	1035.47	0.3492	1040.49	0.8421	1131.07	0.9893
62.40	0.0040	579.77	0.0557	1009.74	0.1418	1035.67	0.4017	1053.42	0.8873	1137.09	0.9945
99.72	0.0071	645.59	0.0642	1033.34	0.1612	1035.67	0.4612	1068.77	0.9183	1142.97	1.0000
145.84	0.0111	693.00	0.0708	1034.02	0.1830	1035.85	0.5263	1082.82	0.9399		
189.50	0.0149	743.26	0.0784	1034.86	0.2066	1036.11	0.5968	1094.91	0.9546		
325.55	0.0277	796.59	0.0872	1035.77	0.2321	1036.14	0.6628	1108.22	0.9692		
355.58	0.0307	851.37	0.0973	1035.38	0.2632	1036.35	0.7280	1119.11	0.9793		
385.07	0.0337	909.31	0.1101	1035.51	0.3032	1036.43	0.7890	1124.71	0.9844		

Table 2. Regressed Parameters and Pressure Deviations for Systems of DME (1) + *n*-Decane (2), DME (1) + *n*-Dodecane (2), DME (1) + 1-Octene (2), DME (1) + Benzene (2), DME (1) + Toluene (2), and DME (1) + Water (2) at 323.15 K

model equation	A_{12}	A_{21}	α_{12}	ΔP/kPa
DME (1) + *n*-Decane (2)				
Margules	−0.0487	−0.0492		38.84
van Laar	−0.0104	−1.3302		26.56
Wilson	474.4339	−474.3808		39.11
NRTL	−37.8284	6.5524	0.3048	38.79
UNIQUAC	−152.4370	319.5013		38.16
DME (1) + *n*-Dodecane (2)				
Margules	−0.2092	−0.0971		25.50
van Laar	−0.2989	−0.1151		26.77
Wilson	251.6124	−21.2882		35.34
NRTL	−102.1879	40.4763	4.9028	26.42
UNIQUAC	−77.9854	239.7257		34.73
DME (1) + 1-Octene (2)				
Margules	−0.0107	−0.2795		56.86
van Laar	−0.0584	−0.8412		17.76
Wilson	97.9147	−97.9358		63.03
NRTL	40.3222	−40.3470	42.4392	17.76
UNIQUAC	−303.2635	472.4389		48.64
DME (1) + Benzene (2)				
Margules	0.0183	−0.2788		58.22
van Laar	−0.0665	−0.8466		13.23
Wilson	−246.1952	245.9898		67.26
NRTL	46.6072	−46.2807	35.3284	13.21
UNIQUAC	−630.9460	1112.3162		78.10
DME (1) + Toluene (2)				
Margules	0.0122	−0.2474		60.43
van Laar	−0.0564	−0.9254		15.57
Wilson	−116.0939	116.2200		65.87
NRTL	219.1782	−36.2000	49.6345	15.57
UNIQUAC	−541.3794	855.6668		67.41
DME (1) + Water (2)				
Margules	2.4979	2.2149		151.77
van Laar	2.5051	2.2217		150.64
Wilson	335.8253	2584.7223		305.67
NRTL	940.1671	1055.5143	0.3131	138.80
UNIQUAC	851.2268	96.6913		139.30

Results and Discussion

The measured isothermal VLE data ($P-x$) for the six binary systems of DME + decane, + dodecane, + 1-octene, + benzene, + toluene, and + water systems at 323.15 K are listed in Table 1. They are plotted in Figure 1 for DME with alkane and alkene systems and in Figure 2 for DME with aromatics and water systems.

Although the systems DME with decane, dodecane, 1-octene, benzene, and toluene show nearly ideal behavior in the whole composition range, the system DME + water shows a miscibility gap. Graphically, a miscibility gap from $x_1 = 0.16$ to $x_1 = 0.83$ was determined. The experimental data were correlated with five common G^E models, i.e., Margules, van Laar, Wilson, NRTL, and UNIQUAC. The model parameters and mean deviations of P (ΔP) between experimental and calculated system pressures are listed in Table 2. The objective function used to calculate the pressure is

$$\mathrm{OF} = \sum\left(1 - \frac{P_{\mathrm{calcd}}}{P_{\mathrm{exptl}}}\right)^2 \leq \epsilon \qquad (1)$$

The binary interaction parameters (A_{ij}) of Wilson, NRTL, and UNIQUAC models mean

$$A_{ij} = (\lambda_{ij} - \lambda_{ii})/\mathrm{J{\cdot}mol^{-1}} \quad \text{(Wilson)} \qquad (2)$$

$$A_{ij} = (g_{ij} - g_{ii})/\mathrm{J{\cdot}mol^{-1}} \quad \text{(NRTL)} \qquad (3)$$

$$A_{ij} = (u_{ij} - u_{ii})/\mathrm{J{\cdot}mol^{-1}} \quad \text{(UNIQUAC)} \qquad (4)$$

Table 3. Excess Molar Enthalpies (H^E) for the Binary Systems of DME with Some Hydrocarbons and Water at 323.15 K

x_1	H^E J·mol^{-1}	x_1	H^E J·mol^{-1}	x_1	H^E J·mol^{-1}
DME (1) + *n*-Decane (2)					
0.1299	237.248	0.6541	633.618	0.8948	312.510
0.2396	408.717	0.6989	606.761	0.9190	252.422
0.3336	525.592	0.7393	571.385	0.9414	189.820
0.4149	597.104	0.7761	526.305	0.9623	127.427
0.4860	637.332	0.8097	478.921	0.9818	62.789
0.5487	653.719	0.8405	425.156		
0.6043	650.944	0.8687	371.203		
DME (1) + *n*-Dodecane (2)					
0.1484	280.311	0.6882	660.325	0.9085	306.214
0.2690	466.353	0.7304	624.770	0.9298	246.887
0.3688	584.831	0.7681	583.237	0.9494	184.600
0.4529	658.181	0.8019	532.099	0.9675	123.527
0.5247	687.748	0.8324	480.324	0.9844	60.713
0.5866	696.872	0.8601	423.704		
0.6407	686.963	0.8854	365.341		
DME (1) + 1-Octene (2)					
0.1071	104.072	0.6031	331.823	0.8724	174.100
0.2021	186.857	0.6509	321.974	0.9011	139.774
0.2868	248.119	0.6950	306.937	0.9281	106.053
0.3630	290.775	0.7358	283.868	0.9535	71.042
0.4317	317.356	0.7737	260.057	0.9774	35.240
0.4941	331.335	0.8089	232.657		
0.5510	334.998	0.8417	203.478		
DME (1) + 1-Decene (2)					
0.1265	130.133	0.6472	366.862	0.8920	173.725
0.2342	232.274	0.6924	350.701	0.9167	138.865
0.3269	299.693	0.7335	329.154	0.9397	103.389
0.4076	344.914	0.7708	301.404	0.9612	68.193
0.4784	366.930	0.8050	271.810	0.9812	33.734
0.5411	378.218	0.8363	241.011		
0.5971	377.521	0.8652	207.156		
DME (1) + Benzene (2)					
0.0637	−33.4	0.4628	−188	0.7950	−161
0.1256	−64.0	0.5140	−196	0.8379	−141
0.1857	−94.5	0.5638	−200	0.8799	−115
0.2442	−120	0.6124	−200	0.9208	−81.8
0.3011	−141	0.6597	−197	0.9609	−43.5
0.3565	−160	0.7059	−189		
0.4104	−176	0.7510	−179		
DME (1) + Toluene (2)					
0.0753	−48.8	0.5078	−223	0.8228	−170
0.1467	−90.6	0.5587	−226	0.8609	−147
0.2145	−128	0.6075	−228	0.8976	−118
0.2790	−158	0.6541	−224	0.9330	−83.1
0.3403	−180	0.6989	−216	0.9671	−44.3
0.3988	−199	0.7419	−206		
0.4545	−213	0.7831	−190		
DME (1) + Water (2)					
0.0137	−132	0.3054	−399	0.7649	308
0.0363	−317	0.4418	−188	0.8337	413
0.0808	−543	0.5543	−14.7	0.8713	400
0.1366	−646	0.6487	132	0.9114	348
0.2087	−547	0.7036	216	0.9542	243

Although for the systems DME + decane and + dodecane the best results are obtained by the van Laar and Margules equation, the NRTL equation gives the best correlation result for the other systems. The continuous lines in Figures 1 and 2 are the calculated values by the best model for each system. Additionally, the measured data were compared with the values published by Pozo and Streett.[4] Both data sets are in good agreement as shown in Figure 2.

The measured H^E data at 323.15 K for the same binary systems and additionally for the system DME with 1-decene are given in Table 3. The H^E data for the system DME + decane, + dodecane, + 1-octene, and + 1-decene show endothermic behavior as shown in Figure 3. The H^E values for the measured

Table 4. Fitted Parameters and Standard Mean Deviation of the Redlich−Kister Polynomial and Partial Excess Molar Enthalpies at Infinite Dilution for Systems of DME + Component 2 at 323.15 K

component 2	A_1	A_2	A_3	A_4	A_5	A_6	σ_{st}	$\bar{H}_1^{E,\infty}$	$\bar{H}_2^{E,\infty}$
n-decane	2571.2	691.6	191.6	151.2			0.952	1920.0	3605.6
n-dodecane	2722.2	834.4	224.6	158.9	105.0		1.154	2058.5	4045.1
1-octene	1328.0	269.4	44.5	54.6	−80.0		0.848	968.5	1616.5
1-decene	1490.7	370.8	80.2	65.6	−170.5		0.738	964.0	1836.8
benzene	−774.3	−276.6	−111.2	−124.7	19.8	70.6	0.683	−535.0	−1196.4
toluene	−883.3	−287.4	−167.2	−106.9	−10.9		0.675	−667.1	−1455.7
water	−353.3	2696.3	−658.8	6259.5	−912.4		15.393	−10880.3	7031.3

alkane (decane and dodecane) and alkene (1-octene and 1-decene) with DME increase with increasing chain length of the hydrocarbon. Although the H^E data for DME with benzene and toluene show negative values (see Figure 4), the H^E data with toluene are more strongly negative. The heterogeneous system DME + water shows endothermic and exothermic behavior. The measured H^E data of each binary system were correlated with the Redlich−Kister polynomial.[8]

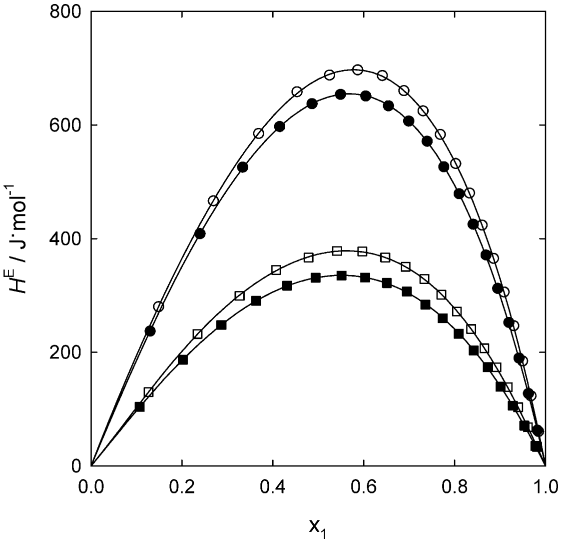

Figure 3. H^E for the binary systems DME (1) + *n*-decane (2), + *n*-dodecane (2), + 1-octene (2), and + 1-decene (2) at 323.15 K: ●, DME + *n*-decane; ○, DME + *n*-dodecane; ■, DME + 1-octene; □, DME + 1-decene. Solid curves were calculated from the Redlich−Kister polynomial.

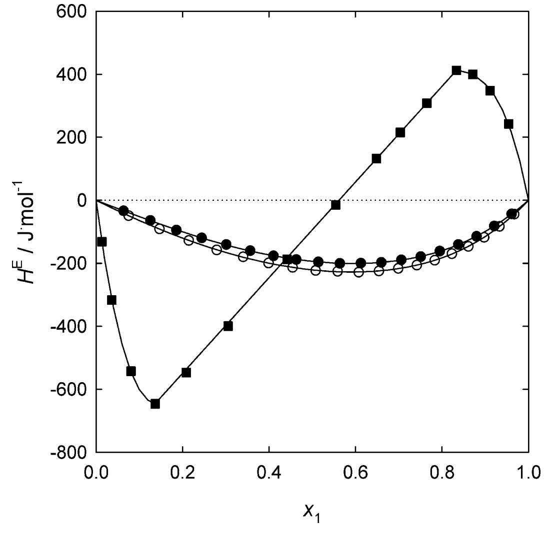

Figure 4. H^E for the binary systems DME (1) + benzene (2), + toluene (2), and + water (2) at 323.15 K: ●, DME + benzene; ○, DME + toluene; ■, DME + water. Solid curves were calculated from the Redlich−Kister polynomial.

$$H^E_{12}/\text{J·mol}^{-1} = x_1 x_2 \sum_{i=1}^{n} A_i (x_1 - x_2)^{i-1} \tag{5}$$

where A_i is the adjustable parameters and n is the number of fitted parameters.

The Redlich−Kister equation provides excellent correlation results for the measured H^E for all binary mixtures. The mean deviations of the correlated H^E values from the experimental data are approximately 1 J·mol^{-1} except for the system DME + water. The fitted parameters and the standard mean deviation using the Redlich−Kister polynomial are listed in Table 4. The calculated H^E values using Redlich−Kister parameters are furthermore described by the solid lines, shown in Figures 3 and 4. Besides, the partial excess molar enthalpies at infinite dilution were also calculated and given in Table 4. It can be directly calculated using eq 6 and the fitted Redlich−Kister parameters.

$$\begin{aligned} \bar{H}_1^{E,\infty}/\text{J·mol}^{-1} &= A_1 - A_2 + A_3 - A_4 + A_5 - A_6 \\ \bar{H}_2^{E,\infty}/\text{J·mol}^{-1} &= A_1 + A_2 + A_3 + A_4 + A_5 + A_6 \end{aligned} \tag{6}$$

Conclusions

Isothermal VLE ($P-x$) at 323.15 K for the binary mixtures of DME with *n*-decane, *n*-dodecane 1-octane, benzene, toluene, and water were determined by using a computer-driven static VLE apparatus. Additionally, heat of mixing (H^E) data at 323.15 K for the same binary systems and the DME + 1-decene system were measured using an isothermal flow calorimeter. The measured vapor−liquid equilibrium data show almost ideal behavior except for the DME + water system, and they show no azeotropic behavior. Although the measured excess enthalpies show positive values for DME with alkane and alkene systems, negative values are obtained for DME + aromatic systems. The measured VLE data were correlated well with various common G^E models, and the Redlich−Kister equation was applied for the correlation of the heat of mixing data. The system DME + water shows a miscibility gap.

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