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# Excess Molar Enthalpies for Dimethyl Carbonate with *o*-Xylene, *m*-Xylene, *p*-Xylene, Ethylbenzene, or Ethyl Benzoate at 298.15 K and 10.2 MPa

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A flow-type isothermal microcalorimeter was used to measure excess molar enthalpies, $H^E$, for the binary mixtures of dimethyl carbonate with *o*-xylene, *m*-xylene, *p*-xylene, ethylbenzene, or ethyl benzoate at 298.15 K and 10.2 MPa. The experimental $H^E$ values are positive for all the binary mixtures over the entire composition range. In comparison with the values of $H^E$ at 0.1 MPa, the excess enthalpies were found to decrease with increasing pressure for these binary systems, except for dimethyl carbonate + ethylbenzene. The maximum $H^E$ values are in the range 356.8 J·mol$^{-1}$ to 652.8 J·mol$^{-1}$, which are located at about $x_i = 0.5$. The experimental $H^E$ data were correlated accurately with the Redlich−Kister polynomial equation. The Peng−Robinson and the Cubic Chain-of-Rotators equations of state with the binary interaction parameters determined from $H^E$ data at atmospheric pressure were adopted to calculate the excess enthalpies at 10.2 MPa. The Cubic Chain-of-Rotators equation with two binary interaction parameters yielded better results.

## Introduction

Excess enthalpy data, $H^E$, are fundamentally important to understanding the intermolecular interactions between dissimilar molecules and can also be applied to test the reliability of thermodynamic models. The present study is undertaken to investigate the enthalpic behavior of the mixtures containing dimethyl carbonate (DMC). DMC is a potential substitute for methyl *tert*-butyl ether gasoline additive, due to its high oxygen content, low toxicity, and high biodegradability.[1] It is also widely used as a solvent in many liquid extraction processes. Lien et al.[2] measured the heats of mixing for the binary systems of DMC with *o*-xylene, *p*-xylene, *m*-xylene, ethylbenzene, or ethyl benzoate at 298.15 K under atmospheric pressure. As an extension, similar measurements were made in this work at 10.2 MPa for the same five binary systems to reveal the pressure effect on $H^E$. No literature data are found for these systems at comparable conditions. With the binary interaction parameters determined from the excess enthalpy data at atmospheric pressure, the Peng−Robinson (PR)[3] and the Cubic Chain-of-Rotators (CCOR)[4] equations of state were used to estimate the excess enthalpies at 10.2 MPa. The reliability of these equations of state was tested with the experimental results of this work.

## Experimental Section

***Chemicals.*** Dimethyl carbonate (99 mass %), *o*-xylene (99 mass %), *m*-xylene (99+ mass %), *p*-xylene (99 mass %), and ethylbenzene (99.8 mass %) were purchased from Arcos (USA). Ethyl benzoate (99+ mass %) was supplied by Aldrich (USA). The densities of these chemicals at 298.15 K were measured by an Anton Paar densimeter, DMA 4500, with an accuracy of ±5 × 10$^{-5}$ g·cm$^{-3}$. The densities are compared with literature values in Table 1, indicating that the agreement is satisfactory. All the chemicals were used without further purification.

**Table 1. Densities of Pure Components at 298.15 K, Calibration Constants, and Data Sources of $H^E$ Used for Determination of Calibration Constants**

| component | $\rho$/g·cm$^{-3}$ this work | $\rho$/g·cm$^{-3}$ lit. | calibration constants, $a_{0i}/a_{1i}$ | $i$ | ref[a] |
|---|---|---|---|---|---|
| dimethyl carbonate | 1.0631 | 1.063 40[5] | 2.9913<br>0.5466 | 1 | 9[b] |
| *o*-xylene | 0.8750 | 0.875 96[6] | 3.0941<br>0.6996 | 2 | 10[c] |
| *m*-xylene | 0.8600 | 0.859 90[6] | 3.1421<br>0.4348 | 2 | 10[d] |
| *p*-xylene | 0.8567 | 0.856 68[6] | 3.1832<br>0.5284 | 2 | 11[e] |
| ethylbenzene | 0.8625 | 0.862 6[7] | 3.0606<br>0.6291 | 2 | 12[f] |
| ethyl benzoate | 1.0413 | 1.041 4[8] | 3.4119<br>0.0010 | 2 | 13[g] |

[a] Data sources of $H^E$ used for determination of calibration constants. [b] Dimethyl carbonate + hexane and dimethyl carbonate + benzene. [c] Benzene + *o*-xylene. [d] Benzene + *p*-xylene. [e] Benzene + *m*-xylene. [f] Methanol + ethylbenzene. [g] Hexane + ethyl benzoate.

***Apparatus and Procedure.*** A flow-type isothermal microcalorimeter (model 4400 IMC, Calorimeter Science Corporation, USA) was employed in this study to measure the enthalpy changes of mixing. Its schematic diagram and the procedure of the operations have been detailed in ref 14. The apparatus consists of a flow-mixing cell, a reference cell, a thermostatic water bath, a data acquisition system, and two liquid pumps (model PU-1580, JASCO, Japan) with an on-line degasser. The temperature of the bath was controlled to within ±0.05 K. The heat of mixing can be measured to within a sensitivity of ±0.015 J·mol$^{-1}$.

The molar ratio of the two mixing streams was calculated from the molecular weights, densities, and the volumetric

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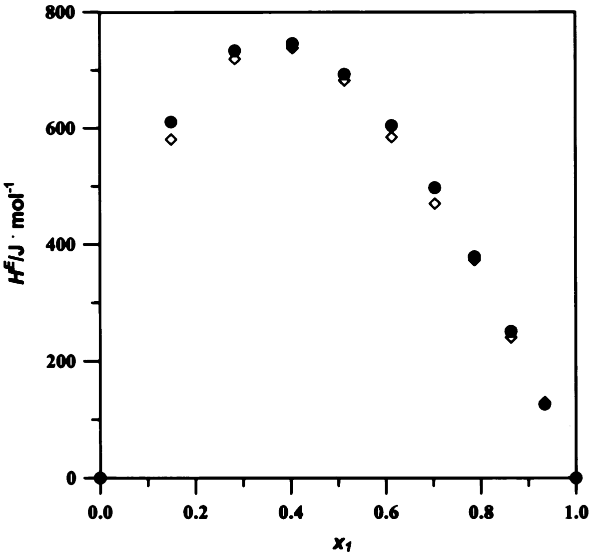

**Figure 1.** Comparison of literature data[15] (●) with data of this work (◇). Excess molar enthalpies, $H^E$, for 2-methyl-1-propanol + heptane at 298.15 K and 10.2 MPa.

**Table 2. Excess Molar Enthalpies, $H^E$, of Binary Mixtures Containing Dimethyl Carbonate at 298.15 K and 10.2 MPa**

| $x_1$ | $H^E$/J·mol$^{-1}$ | $x_1$ | $H^E$/J·mol$^{-1}$ |
|---|---|---|---|
| Dimethyl Carbonate (1) + *o*-Xylene (2) | | | |
| 0.1005 | 136.3 | 0.5515 | 610.0 |
| 0.1501 | 240.8 | 0.6004 | 591.0 |
| 0.2014 | 345.1 | 0.6499 | 559.5 |
| 0.2511 | 426.9 | 0.6999 | 516.6 |
| 0.2992 | 496.8 | 0.7502 | 458.0 |
| 0.3516 | 548.7 | 0.8006 | 384.7 |
| 0.3994 | 586.1 | 0.8511 | 296.5 |
| 0.4510 | 603.2 | 0.8997 | 193.0 |
| 0.5009 | 616.7 | | |
| Dimethyl Carbonate (1) + *m*-Xylene (2) | | | |
| 0.0503 | 77.6 | 0.5506 | 646.6 |
| 0.1007 | 200.4 | 0.5995 | 631.7 |
| 0.1511 | 313.7 | 0.6521 | 604.3 |
| 0.2016 | 389.1 | 0.7018 | 565.2 |
| 0.2519 | 468.8 | 0.7517 | 506.9 |
| 0.3005 | 531.6 | 0.8017 | 432.6 |
| 0.3505 | 577.2 | 0.8516 | 342.5 |
| 0.4015 | 608.5 | 0.9014 | 230.5 |
| 0.4505 | 641.7 | 0.9509 | 105.4 |
| 0.4998 | 648.7 | | |
| Dimethyl Carbonate (1) + *p*-Xylene (2) | | | |
| 0.0504 | 66.8 | 0.5524 | 637.7 |
| 0.1026 | 188.3 | 0.6013 | 620.7 |
| 0.1515 | 293.0 | 0.6507 | 597.6 |
| 0.2020 | 356.0 | 0.7004 | 552.1 |
| 0.2524 | 440.2 | 0.7503 | 489.1 |
| 0.3011 | 498.1 | 0.8002 | 432.4 |
| 0.3511 | 572.5 | 0.8502 | 346.8 |
| 0.4022 | 613.4 | 0.9017 | 235.0 |
| 0.4514 | 635.7 | 0.9511 | 107.4 |
| 0.5016 | 640.2 | | |
| Dimethyl Carbonate (1) + Ethylbenzene (2) | | | |
| 0.0501 | 100.8 | 0.5510 | 651.6 |
| 0.1004 | 212.5 | 0.6023 | 636.1 |
| 0.1491 | 268.1 | 0.6517 | 608.7 |
| 0.2011 | 361.6 | 0.7014 | 569.8 |
| 0.2514 | 446.0 | 0.7514 | 518.0 |
| 0.3000 | 512.5 | 0.8014 | 448.8 |
| 0.3528 | 571.9 | 0.8514 | 366.9 |
| 0.4009 | 611.6 | 0.9013 | 270.6 |
| 0.4528 | 638.3 | 0.9508 | 146.0 |
| 0.5028 | 652.5 | | |
| Dimethyl Carbonate (1) + Ethyl Benzoate (2) | | | |
| 0.1518 | 107.4 | 0.5496 | 355.6 |
| 0.1995 | 165.7 | 0.5991 | 347.5 |
| 0.2486 | 220.7 | 0.6507 | 332.4 |
| 0.2988 | 265.6 | 0.6994 | 310.2 |
| 0.3499 | 297.9 | 0.7496 | 277.5 |
| 0.4015 | 325.5 | 0.8010 | 244.5 |
| 0.4507 | 345.0 | 0.8509 | 186.8 |
| 0.5000 | 356.8 | 0.8995 | 119.4 |

flow rates of components 1 and 2. In general, the total flow rates were kept at about 0.4 cm$^3$·min$^{-1}$, which is a preferable condition as a result of test runs.

The excess molar enthalpy was obtained from the following equation:

$$H^E = \frac{\epsilon_m(V_m - V_m^\circ)}{\dfrac{f_1\rho_1}{MW_1} + \dfrac{f_2\rho_2}{MW_2}} \tag{1}$$

where $f_i$, $\rho_i$, and MW$_i$ are the volumetric flow rate, density, and molecular weight of constituent $i$, respectively. The voltage generated from the mixing process, $V_m$, is indicated by the data acquisition system. The voltage of the baseline, $V_m^\circ$, was calculated from

$$V_m^\circ = \frac{f_1V_1^\circ + f_2V_2^\circ}{f_1 + f_2} \tag{2}$$

where $V_i^\circ$ is the baseline voltage for component $i$. The baseline voltages were determined by charging with a pure compound at several different flow rates. In general, the value of $V_m^\circ$ is much smaller (<0.1%) than that of $V_m$. The variable $\epsilon_m$ in eq 1 is the calibration coefficient for mixing runs. Its value was estimated from the following equation:

$$\epsilon_m = \Phi_1\epsilon_1 + \Phi_2\epsilon_2 = \left(\frac{f_1}{f_1 + f_2}\right)\epsilon_1 + \left(\frac{f_2}{f_1 + f_2}\right)\epsilon_2 \tag{3}$$

where $\Phi_i$ and $\epsilon_i$ are the volumetric fraction and calibration coefficient for constituent $i$, respectively. It should be noted that the values of the calibration coefficients, $\epsilon_i$, vary with several factors, including temperature, volumetric flow rate, and the path of the streams. At a given operating temperature, the relationship between $\epsilon_i$ and the flow rate, $f_i$, for a specific stream was found to be linear over the entire experimental conditions of this study. Equation 3 can thus be expressed as

$$\epsilon_m = \left(\frac{f_1}{f_1 + f_2}\right)(a_{01} + a_{11}f_1) + \left(\frac{f_2}{f_1 + f_2}\right)(a_{02} + a_{12}f_2) \tag{4}$$

where ($a_{01}$, $a_{11}$) and ($a_{02}$, $a_{12}$) are the calibration constants for streams 1 and 2, respectively. The calibration constants for each constituent compound were determined by fitting the measurable $V_m$ to eq 1 with reference to reliable literature $H^E$ data of the related mixtures, prior to measuring a new system.

Prior to the measurements, several test runs have been made to ensure that eq 4 is still valid at high pressures. The results confirmed that the calibration constants were independent of pressure, at least up to 10.2 MPa. As an illustrative example, Figure 1 compares the experimental results with the literature values[15] for 2-methyl-1-propanol (1) + heptane (2) at 298.15 K and 10.2 MPa, in which the calibration constants were determined from $H^E$ data at atmospheric pressure. The comparison shows that the data treatment is appropriate for the mixing runs at pressures higher than atmospheric pressure. The calibration constants used in the data treatment of this study were thus taken from previous work at atmospheric pressure.[2] Table

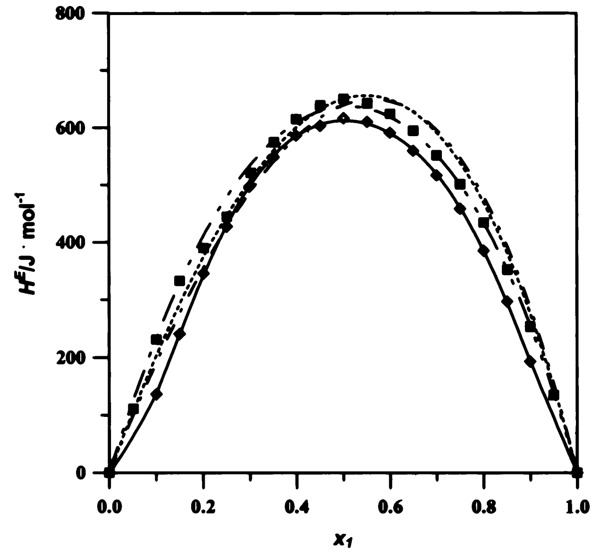

**Figure 2.** Comparison of excess molar enthalpies, $H^E$, for dimethyl carbonate + *o*-xylene at 298.15 K. Experimental data at atmospheric pressure (■) and 10.2 MPa (◇); calculated from the Redlich–Kister equation (—); calculated from the PR (- - -), CCOR$_1$ (— - —), and CCOR$_2$ (— -- —) EOS with the binary interaction parameters determined from $H^E$ data at atmospheric pressure.

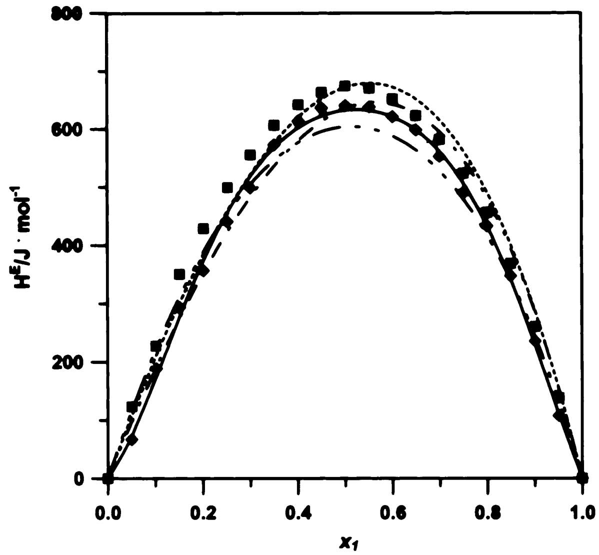

**Figure 4.** Comparison of excess molar enthalpies, $H^E$, for dimethyl carbonate + *p*-xylene at 298.15 K. Experimental data at atmospheric pressure (■) and 10.2 MPa (◇); calculated from the Redlich–Kister equation (—); calculated from the PR (- - -), CCOR$_1$ (— - —), and CCOR$_2$ (— - - —) EOS with the binary interaction parameters determined from $H^E$ data at atmospheric pressure.

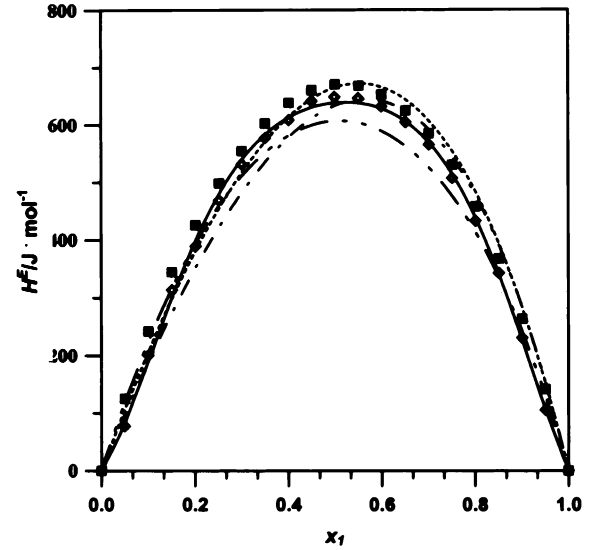

**Figure 3.** Comparison of excess molar enthalpies, $H^E$, for dimethyl carbonate + *m*-xylene at 298.15 K. Experimental data at atmospheric pressure (■) and 10.2 MPa (◇); calculated from the Redlich–Kister equation (—); calculated from the PR (- - -), CCOR$_1$ (— - —), and CCOR$_2$ (— - - —) EOS with the binary interaction parameters determined from $H^E$ data at atmospheric pressure.

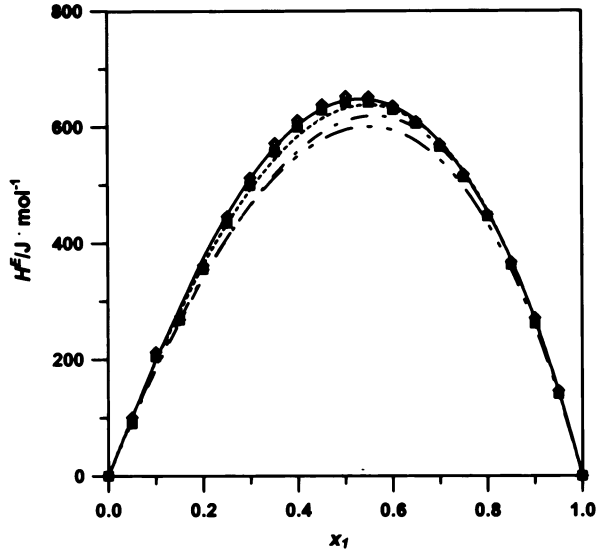

**Figure 5.** Comparison of excess molar enthalpies, $H^E$, for dimethyl carbonate + ethylbenzene at 298.15 K. Experimental data at atmospheric pressure (■) and 10.2 MPa (◇); calculated from the Redlich–Kister equation (—); calculated from the PR (- - -), CCOR$_1$ (— - —), and CCOR$_2$ (— - - —) EOS with the binary interaction parameters determined from $H^E$ data at atmospheric pressure.

1 lists the calibration constants and the sources of data used for the calibration. The accuracy of the reported $H^E$ in this study is estimated to be better than ±2%.

## Results and Discussion

The experimental results are reported in Table 2 for the binary systems of DMC with *o*-xylene, *m*-xylene, *p*-xylene, ethylbenzene, or ethyl benzoate at 298.15 K and 10.2 MPa. Figures 2–6 illustrate the variation of the excess molar enthalpies with the mole fraction of DMC. The results indicated that the excess enthalpies are positive (endothermic) over the entire composition range. The maximum $H^E$ values are located at about $x_1 = 0.5$. These maximum values are 652.5 J·mol$^{-1}$, 648.7 J·mol$^{-1}$, 640.2 J·mol$^{-1}$, 617.6 J·mol$^{-1}$, and 356.8 J·mol$^{-1}$ for the systems containing ethylbenzene, *m*-xylene, *p*-xylene, *o*-xylene, and ethyl benzoate, respectively. The values of $H^E$ at atmospheric pressure are also shown in the graphs for comparison. As reported by Lien et al.,[2] the maximum $H^E$ values of the corresponding systems at atmospheric pressure are 641.8 J·mol$^{-1}$, 670.7 J·mol$^{-1}$, 674.0 J·mol$^{-1}$, 650.3 J·mol$^{-1}$, and 386.6 J·mol$^{-1}$, respectively. This reveals that the excess enthalpies decrease with increasing pressure in the mixtures of DMC + *o*-xylene, + *m*-xylene, + *p*-xylene, and + ethyl benzoate but increase slightly with increasing pressure in the mixtures of DMC + ethylbenzene. The pressure effects on $H^E$ in the systems of DMC + xylenes are consistent with the behavior observed by previous investigators,[16–18] who concluded that $(\partial H^E/\partial P)_T$ values were usually negative for (polar + nonpolar) mixtures.

**Table 3. Correlated Results from the Redlich–Kister Equation**

| dimethyl carbonate + | $h_0$ / J·mol$^{-1}$ | $h_1$ / J·mol$^{-1}$ | $h_2$ / J·mol$^{-1}$ | $h_3$ / J·mol$^{-1}$ | $h_4$ / J·mol$^{-1}$ | $H^E$ AAD[a] / % |
|---|---|---|---|---|---|---|
| *o*-xylene | 2448.8 | 29.8 | 89.1 | 577.8 | −1647.0 | 0.5 |
| *m*-xylene | 2554.0 | 157.9 | 855.9 | 197.8 | −1901.3 | 1.9 |
| *p*-xylene | 2527.2 | 202.0 | 502.8 | 354.5 | −1521.6 | 2.8 |
| ethylbenzene | 2579.2 | 345.9 | 352.4 | 248.9 | | 1.6 |
| ethyl benzoate | 1403.0 | 198.6 | 106.7 | 503.0 | −1332.0 | 1.1 |

[a] $H^E$ AAD (%) $= (100/N)\sum_{i=1}^{N}|H^E_{i,\mathrm{calc}} - H^E_{i,\mathrm{expt}}|/H^E_{i,\mathrm{expt}}$.

**Table 4. Calculated Results of $H^E$ from the Equations of State**

| | PR | | CCOR$_1$ | | CCOR$_2$ | | |
|---|---|---|---|---|---|---|---|
| component 2 | $k_{aij}$ | $H^E$ AAD[a] (%) | $k_{aij}$ | $H^E$ AAD[a] (%) | $k_{aij}$ | $k_{cij}$ | $H^E$ AAD[a] (%) |
| *o*-xylene | 0.0247 | 15.1 | 0.0165 | 11.9 | −0.1100 | −0.1457 | 9.7 |
| *m*-xylene | 0.0226 | 9.8 | 0.0153 | 10.6 | −0.1200 | −0.1561 | 8.7 |
| *p*-xylene | 0.0220 | 13.0 | 0.0145 | 10.3 | −0.1195 | −0.1543 | 10.1 |
| ethylbenzene | 0.0210 | 2.5 | 0.0145 | 4.9 | −0.0232 | −0.0430 | 6.4 |
| ethyl benzoate | −0.0176 | 22.0 | −0.0444 | 37.6 | −0.1136 | −0.0702 | 19.8 |
| grand AAD (%) | | 12.5 | | 15.1 | | | 10.9 |

[a] $H^E$ AAD (%) $= (100/N)\sum_{i=1}^{N}|H^E_{i,\mathrm{calc}} - H^E_{i,\mathrm{expt}}|/H^E_{i,\mathrm{expt}}$.

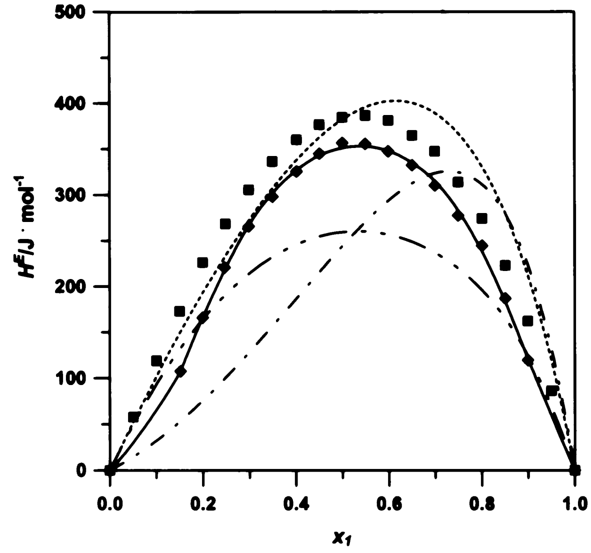

**Figure 6.** Comparison of excess molar enthalpies, $H^E$, for dimethyl carbonate + ethyl benzoate at 298.15 K. Experimental data at atmospheric pressure (■) and 10.2 MPa (◇); calculated from the Redlich–Kister equation (—); calculated from the PR (- - -), CCOR$_1$ (— - —), and CCOR$_2$ (— - - —) EOS with the binary interaction parameters determined from $H^E$ data at atmospheric pressure.

## Data Correlation

The new $H^E$ data were fitted to the Redlich–Kister (R–K) equation:

$$H^E = x_1(1 - x_1)\sum_{i=0}^{m} h_i(1 - 2x_1)^i \tag{5}$$

The values of the coefficients $h_i$ were determined by a least-squares regression for each binary system. Table 3 reports the calculated results, in which $H^E$ AAD (%) is the percentage average absolute deviation, defined as

$$H^E \text{ AAD (\%)} = \frac{100}{N}\sum_{i=1}^{N}|H^E_{i,\mathrm{calc}} - H^E_{i,\mathrm{expt}}|/H^E_{i,\mathrm{expt}} \tag{6}$$

where $N$ is the number of experimental data points. As presented in Table 3 and Figures 2–6, the Redlich–Kister equation correlates well these new $H^E$ data.

The Peng–Robinson and the Cubic Chain-of-Rotators equations of state incorporated with the van der Waals one-fluid mixing rules were used for calculating the excess enthalpies at 10.2 MPa. The combining rules of the PR equation contain only one binary interaction parameter $k_{a12}$. Two versions of treatment were adopted in the enthalpy calculations to the CCOR equation. The first one, denoted as CCOR$_1$, also contains a single binary interaction parameter $k_{a12}$, the same as that for the PR equation. The second one, denoted as CCOR$_2$, has an additional binary interaction parameter $k_{c12}$ in the combining rule of $c_{ij}$. These binary interaction parameters, taken from Lien et al.,[2] were determined from the $H^E$ data at atmospheric pressure. The calculated results are shown in Table 4 and compared with the experimental values in Figures 2–6. The average absolute deviations (AADs) are reasonable for DMC + *m*-xylene, + *p*-xylene, and + ethylbenzene. However, the deviations are as high as 29% or larger for DMC + *o*-xylene and DMC + ethyl benzoate. In general, the CCOR equation with two adjustable interaction parameters (CCOR$_2$) yields better results.

## Conculsions

The excess enthalpies have been measured for the binary mixtures of dimethyl carbonate + *o*-xylene, + *m*-xylene, + *p*-xylene, + ethylbenzene, and + ethyl benzoate at 298.15 K and 10.2 MPa. It was found that the mixing process is endothermic for these five binary systems. The maximum $H^E$ values of the investigated systems followed the order ethylbenzene > *m*-xylene > *p*-xylene > *o*-xylene > ethyl benzoate. In comparison with the experimental results at atmospheric pressure, the excess enthalpies were found to decrease with increasing pressure, except for dimethyl carbonate + ethylbenzene. The Redlich–Kister equation correlated well these new $H^E$ data. By using the binary interaction parameters determined from $H^E$ data at atmospheric pressure, the Cubic Chain-of-Rotators equation of state with two binary interaction parameters yielded, generally, better results than the Peng–Robinson equation of state and the Cubic Chain-of-Rotators equation of state with a single binary interaction parameter.

## Literature Cited

(1) Pacheco, M. A.; Marshall, C. L. Review of Dimethyl Carbonate (DMC) Manufacture and Its Characteristics as a Fuel Additive. *Energy Fuels* **1997**, *11*, 2–29.

(2) Lien, P. J.; Lin, H. M.; Lee, M. J.; Venkatesu, P. Excess Molar Enthalpies of Dimethyl Carbonate with *o*-Xylene, *m*-Xylene, *p*-Xylene, Ethylbenzene or Ethyl Benzoate at 298.15 K. *J. Chem. Eng. Data* **2003**, *48*, 110−113.

(3) Peng, D. Y.; Robinson, D. B. A New Two-Constant Equation of State. *Ind. Eng. Chem. Fundam.* **1976**, *15*, 59−64.

(4) Lin, H. M.; Kim, H.; Guo, T, M.; Chao, K. C. Cubic Chain-of-Rotators Equation of State. *Fluid Phase Equilib.* **1983**, *13*, 143−152.

(5) Gonzalez, J. A.; Garcia, I.; Cobos, J. C.; Casanova, C. Thermodynamics of Binary Mixtures Containing Organic Carbonates. 4. Liquid−Liquid Equilibria of Dimethyl Carbonate + Selected *n*-Alkanes. *J. Chem. Eng. Data* **1991**, *36*, 162−164.

(6) Riddick, J. A.; Bunger, W. A. *Sakano, Organic Solvents*, 4th ed.; Wiley-InterScience: New York, 1986; Vol. II.

(7) Grolier, J.-P. E.; Faradjzadeh, A. *International Data Series, Selected Data on Mixtures, Ser. A*, Vol. 1980, No. 2; Thermodynamics Research Center, The Texas A & M University System: College Station, TX, 1980; p 143.

(8) Grolier, J.-P. E. *International Data Series, Selected Data on Mixtures, Ser. A*, Vol. 1973, No. 2; Thermodynamics Research Center, The Texas A & M University System: College Station, TX, 1973; p 109.

(9) Garcia, I.; Cobos, J. C.; Gonzalez, J. A.; Casanova, C. Thermodynamics of Binary Mixtures Containing Organic Carbonates. Excess Enthalpies of Dimethyl Carbonate + Hydrocarbons or + Tetrachloromethane. *J. Chem. Eng. Data* **1988**, *33*, 423−426.

(10) Singh, J.; Pflug, H. D.; Benson, G. C. Molar Execss Enthalpies and Volumes of Benzene-Isomeric Xylene Systems at 25 °C. *J. Phys. Chem.* **1968**, *72*, 1938−1943.

(11) Ott, J. B.; Marsh, K. N.; Stokes, R. H. Excess Enthalpies, Excess Volumes, and Excess Gibbs Free Energies for Benzene+*p*-Xylene at 288.15, 298.15, 308.15, and 318.15 K. *J. Chem. Thermodyn.* **1980**, *12*, 493−503.

(12) Van Ness, H. C.; Abbott, M. M. *International Data Series, Selected Data on Mixtures, Ser. A*, Vol. 1976, No. 1; Thermodynamics Research Center, The Texas A & M University System: College Station, TX, 1976; pp 9−10.

(13) Grolier, J.-P. E.; Ballet, D.; Viallard, A. Thermodynamics of Ester-Containing Ketones. Excess Enthalpies and Excess Volumes for Alkyl Acetates and Alkyl Benzoates + Alkanes, + Benzene, + Toluene, and Ethylbenzene. *J. Chem. Thermodyn.* **1974**, *6*, 895−908.

(14) Lien, P. J.; Venkatesu, P.; Lin, H. M.; Lee, M. J. Excess Molar Enthalpies of 1-Octanol with Ethylbenzene, Ethyl Benzoate, Acetophenone, Anisole or Methanol at 298.15 K. *J. Chem. Eng. Data* **2002**, *47*, 768−771.

(15) Oswald, G.; Schmittecker, D.; Wagner, D.; Lichtenthaler, R. N. Excess Enthalpies and Excess Volumes of Alkanol + *n*-Heptane Mixture at High Pressure. *Fluid Phase Equilib.* **1986**, *27*, 119−135.

(16) Heintz, A.; Lichtenthaler, R. N. Excess Enthalpies of Liquid Alkane Mixtures at Pressures up to 500 Bar. I. Experimental Results. *Ber. Bunsen-Ges. Phys. Chem.* **1980**, *84*, 727−732.

(17) Heintz, A. The Pressure Dependence of the Excess Enthalpies of Mixtures of 2-Propanol with Heptane and Isooctane up to 553 Bar. *Ber. Bunsen-Ges. Phys. Chem.* **1981**, *85*, 632−635.

(18) Gruszkiewicz, M. S.; Sipowska, J. T.; Ott, J. B.; Graham, R. C. Excess Enthalpies for (Butane + Chloromethane) at the Temperatures (298.15, 323.15 and 348.15) K and Pressure (5 and 15) MPa and at the Temperature 293.15 K and Pressure 15 MPa. *J. Chem. Thermodyn.* **1992**, *24*, 1057−1064.

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