

# Measurement and Prediction of the Solubility of CO<sub>2</sub> in Ester Mixture

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## ABSTRACT

*The solubility of CO<sub>2</sub> in ester mixtures under high pressures are studied in this article. The constant-volume method is used to determine the solubility of CO<sub>2</sub> in DMC + diethyl carbonate system, DMC + propyl acetate system, DMC + propylene carbonate system, and DMC + ethylene carbonate system from 282.0 K to 303.0 K. It is found that the solubility of CO<sub>2</sub> in four mixed solvents follows the Henry's law and the linear compound has a greater ability to dissolve CO<sub>2</sub> than the cyclic compound at the same temperature. Furthermore, a modified equation is proposed to fit the solution data and a better equation is obtained in this paper. This will be useful for the future research in the screening of a potential physical solvent for CO<sub>2</sub> capture.*

**Keywords:** CO<sub>2</sub> Solubility, Ester Mixture, Solubility Prediction

## 1. Introduction

There are several ways to separate and capture CO<sub>2</sub> from fossil fueled power plant which can be classified into three general categories: pre-combustion capture, post-combustion capture and oxyfuel strategy. [1] The selection of a technology for a given capture route depends on the features of the gas treated. Based on the method used for CO<sub>2</sub> removal, separating methods can be broadly classified as solvent absorption, adsorption, membrane and cryogenic fractionation. [2] Compared with other methods, solvent absorption process, especially physical solvent, tends to be a favored choice when the partial pressure of CO<sub>2</sub> is high. In addition, physical solvent can be stripped by reducing operation pressure with little heat.

Methanol, [3-6] propylene carbonate [7,8] and polyethylene glycol dimethyl ether [9,10] are commonly used physical solvent in CO<sub>2</sub> capture process. A general trend in CO<sub>2</sub> capture research is to develop a new solvent that has the potential for dramatic cost reduction. Dimethyl carbonate (DMC), a "green" absorbent, with advantages of high CO<sub>2</sub> loading, has been proved to be an effective and low cost absorbent for CO<sub>2</sub> capture. [11] But the characteristics of low boiling point and high freezing point limit its application in industry. According to the colligative properties of solutions, adding a non volatile solute, can increase the boiling point and decrease the

freezing point of a solvent, which means a decreasing loss of the solvent and an enlargement application range respectively in CO<sub>2</sub> capture process. Based on the above, the absorption capacity of DMC combined with other additive is studied in this paper.

## 2. Experiment Materials

CO<sub>2</sub> with a volume fraction of 0.9999 was supplied by BeiWen Gas in Beijing. DMC (C<sub>3</sub>H<sub>6</sub>O<sub>3</sub>, with a mass fraction of 0.999, made in China), propylene carbonate (C<sub>4</sub>H<sub>6</sub>O<sub>3</sub>, with a mass fraction of 0.999, made in China), ethylene carbonate (C<sub>3</sub>H<sub>4</sub>O<sub>3</sub>, with a mass fraction of 0.999, made in China), diethyl carbonate (C<sub>5</sub>H<sub>10</sub>O<sub>3</sub>, with a mass fraction of 0.999, made in China), propyl acetate (C<sub>5</sub>H<sub>12</sub>O<sub>2</sub>, with a mass fraction of 0.999, made in China) were all obtained from Aladdin-Reagent Company in Shanghai. All components were used without further purification. The apparatus, experimental procedure and data processing were described in detail by Xia Gui. [11] Four mixed solvent (DMC + ethylene carbonate, DMC + propyl acetate, DMC + diethyl carbonate, DMC + propylene carbonate) were prepared, in which the amount of DMC is 50% by volume in order to reduce errors in the measurement.

## 3. Pure Compound Properties

Experimental results for the phase equilibrium of CO<sub>2</sub> in DMC at high pressure are given as bellow. The molecu-

lar formula, Hildebrand parameter (Mpa<sup>1/2</sup>), and dielectric constant for the five pure solvents are provided in Table 1 [12-14].

#### 4. Results and Discussions

The temperature T, the CO<sub>2</sub> partial pressure p at the equilibrium state, CO<sub>2</sub> mole fraction x<sub>i</sub> in liquid-phase and the estimated uncertainties u<sub>i</sub>, for the ternary systems CO<sub>2</sub> + DMC + propylene carbonate, CO<sub>2</sub> + DMC + ethylene carbonate, CO<sub>2</sub> + DMC + diethyl carbonate and CO<sub>2</sub> + DMC + propyl acetate are presented in Table 2, Table 3, Table 4, Table 5 and plotted in Figure 1, Figure 2, Figure 3 and Figure 4.

**Table 1. Hildebrand parameter (Mpa<sup>1/2</sup>) and dielectric constant for DMC, propylene carbonate, ethylene carbonate, diethyl carbonate and propyl acetate.**

compound	molecular formula	Hildebrand parameter/Mpa <sup>1/2</sup>	dielectric constant
DMC	C <sub>3</sub> H <sub>6</sub> O <sub>3</sub>	20.2	3.1
propylene carbonate	C <sub>4</sub> H <sub>6</sub> O <sub>3</sub>	27.2	64.4
ethylene carbonate	C <sub>3</sub> H <sub>4</sub> O <sub>3</sub>	30.1	89.6
diethyl carbonate	C <sub>5</sub> H <sub>10</sub> O <sub>3</sub>	18	2.8
propyl acetate	C <sub>5</sub> H <sub>12</sub> O <sub>2</sub>	17.8	5.6

**Table 2. Mole fraction (x<sub>i</sub>), equilibrium pressure (p), and uncertainties (u<sub>i</sub>) of CO<sub>2</sub> in DMC + propylene carbonate from 282.81 K to 313.75 K.**

p/MPa	x <sub>i</sub>	u <sub>i</sub>	p/MPa	x <sub>i</sub>	u <sub>i</sub>
T = 282.81 K					
0.2592	0.0549	0.0009	1.5513	0.2879	0.0021
0.4156	0.0860	0.0021	1.6656	0.3106	0.0027
0.7905	0.1512	0.0017	1.8961	0.3465	0.0016
1.0601	0.1981	0.0027	2.0455	0.3735	0.0032
1.3298	0.2502	0.0028	2.2550	0.4068	0.0028
T = 298.69 K					
0.3311	0.0423	0.0009	1.8901	0.2415	0.0024
0.5826	0.0744	0.0018	2.2296	0.2849	0.0014
0.9543	0.1219	0.0017	2.4782	0.3166	0.0012
1.3130	0.1677	0.0025	2.7687	0.3537	0.0017
1.6656	0.2127	0.0021	3.1073	0.3971	0.0023
T = 313.75 K					
0.3669	0.0362	0.0008	2.7123	0.2678	0.0024
0.8047	0.0795	0.0018	3.4600	0.3416	0.0019
1.0921	0.1078	0.0017	3.8245	0.3776	0.0021
1.5709	0.1551	0.0020	4.2680	0.4214	0.0023
2.2200	0.2191	0.0012			

**Table 3. Mole fraction (x<sub>i</sub>), equilibrium pressure (p), and uncertainties (u<sub>i</sub>) of CO<sub>2</sub> in DMC + ethylene carbonate from 282.83 K to 313.67 K.**

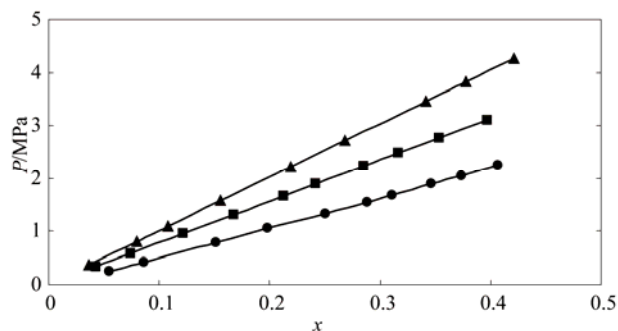
p/MPa	x <sub>i</sub>	u <sub>i</sub>	p/MPa	x <sub>i</sub>	u <sub>i</sub>
T = 282.83 K					
0.1884	0.0317	0.0010	1.2136	0.1980	0.0012
0.3772	0.0709	0.0016	1.3791	0.2246	0.0019
0.5557	0.0987	0.0022	1.5516	0.2508	0.0017
0.7222	0.1233	0.0016	1.7863	0.2854	0.0016
0.9990	0.1680	0.0023	1.9778	0.3142	0.0023
T = 298.78 K					
0.1724	0.0179	0.0008	1.9874	0.2067	0.0017
0.4269	0.0444	0.0016	2.3351	0.2430	0.0021
0.8038	0.0836	0.0022	2.5806	0.2685	0.0019
1.0643	0.1107	0.0016	2.7079	0.2818	0.0023
1.3620	0.1417	0.0012	3.0797	0.3204	0.0019
1.7089	0.1778	0.0017			
T = 313.67 K					
0.2974	0.0231	0.0008	1.610	0.1243	0.0013
0.4798	0.0371	0.0017	2.1497	0.1661	0.0021
0.8023	0.0621	0.0017	2.5945	0.2004	0.0023
1.0939	0.0845	0.0024	3.2326	0.2496	0.0023
1.3283	0.1026	0.0013	3.6383	0.2811	0.0018

**Table 4. Mole fraction (x<sub>i</sub>), equilibrium pressure (p), and uncertainties (u<sub>i</sub>) of CO<sub>2</sub> in DMC + diethyl carbonate from 282.89 K to 313.58 K.**

p/MPa	x <sub>i</sub>	u <sub>i</sub>	p/MPa	x <sub>i</sub>	u <sub>i</sub>
T = 282.89 K					
0.2341	0.0596	0.0012	1.45	0.3589	0.0022
0.5199	0.1367	0.0015	1.57	0.3876	0.0017
0.8111	0.2060	0.0020	1.7	0.4115	0.0019
1.0201	0.2553	0.0014	1.85	0.4444	0.0017
1.2217	0.3051	0.0014	2.03	0.4902	0.0022
T = 298.75 K					
0.3311	0.0576	0.0011	1.8105	0.3150	0.0016
0.6298	0.1096	0.0012	2.5344	0.4410	0.0022
0.9113	0.1586	0.0016	2.7797	0.4837	0.0017
1.2789	0.2225	0.0013	2.9941	0.5210	0.0017
1.5403	0.2680	0.0024			
T = 313.58 K					
0.1618	0.0221	0.0011	1.6413	0.2234	0.0024
0.5417	0.0737	0.0012	1.9669	0.2678	0.0017
0.9820	0.1337	0.0016	2.3695	0.3226	0.0015
1.2507	0.1702	0.0022	2.7381	0.3727	0.0021

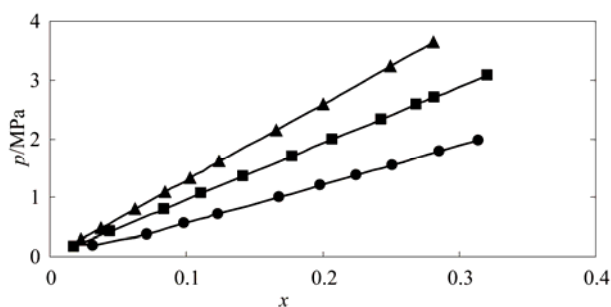
**Table 5. Mole fraction ( $x_i$ ), equilibrium pressure ( $p$ ), and uncertainties ( $u_i$ ) of CO<sub>2</sub> in DMC + propyl acetate from 282.86 K to 313.73 K.**

p/MPa	$x_i$	$u_i$	p/MPa	$x_i$	$u_i$
T = 282.86 K					
0.1869	0.05167	0.0013	0.9840	0.2637	0.0012
0.3474	0.0979	0.0017	1.1182	0.2943	0.0019
0.5358	0.1481	0.0013	1.2986	0.3322	0.0021
0.7113	0.1922	0.0022	1.4800	0.3722	0.0015
0.8647	0.2324	0.0020	1.7335	0.4239	0.0017
T = 298.79 K					
0.1627	0.0296	0.0009	1.5895	0.2894	0.0015
0.3892	0.0709	0.0017	1.7723	0.3227	0.0020
0.5476	0.0997	0.0016	2.0373	0.3713	0.0014
0.8071	0.147	0.0011	2.2797	0.4151	0.0019
1.0966	0.1997	0.0021	2.5771	0.4693	0.0013
1.3551	0.2467	0.0018			
T = 313.73 K					
0.2284	0.0324	0.0010	1.7740	0.2519	0.0011
0.5270	0.0748	0.0018	2.1766	0.3091	0.0022
0.7694	0.1093	0.0019	2.4870	0.3532	0.0021
1.0268	0.1458	0.0024	2.7946	0.3969	0.0015
1.3203	0.1875	0.0023			

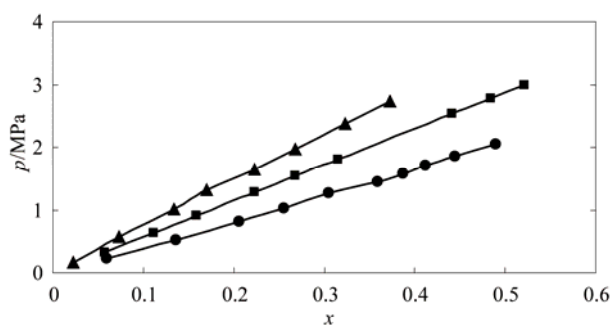


**Figure 1. Vapor-liquid equilibrium of the CO<sub>2</sub> + DMC + propylene carbonate system in this study.  $x$  is the mole fraction of CO<sub>2</sub> in liquid-phase and  $p$  is the partial pressure of CO<sub>2</sub> at equilibrium state: ●, 282.81 K; ■, 298.69 K; ▲, 313.75 K.**

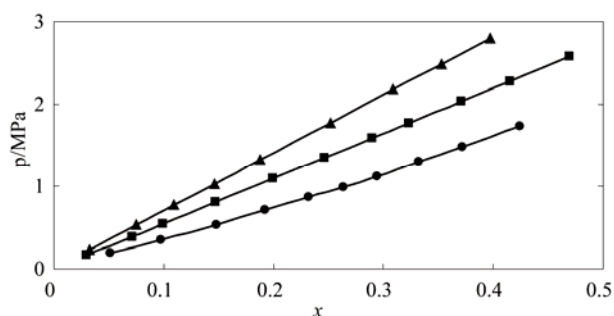
It can be seen from **Table 2** to **Table 5** and **Figure 1** to **Figure 4**, that the solubility of CO<sub>2</sub> in the four mixed solvents decreases with increasing temperature and decreasing pressure. And the solvent absorption capacity



**Figure 2. Vapor-liquid equilibrium of the CO<sub>2</sub> + DMC + ethylene carbonate system in this study.  $x$  is the mole fraction of CO<sub>2</sub> in liquid-phase and  $p$  is the partial pressure of CO<sub>2</sub> at equilibrium state: ●, 282.83 K; ■, 298.78 K; ▲, 313.75 K.**



**Figure 3. Vapor-liquid equilibrium of the CO<sub>2</sub> + DMC + diethyl carbonate system in this study.  $x$  is the mole fraction of CO<sub>2</sub> in liquid-phase and  $p$  is the partial pressure of CO<sub>2</sub> at equilibrium state: ●, 282.89 K; ■, 298.75 K; ▲, 313.58 K.**



**Figure 4. Vapor-liquid equilibrium of the CO<sub>2</sub> + DMC + propyl acetate system in this study.  $x$  is the mole fraction of CO<sub>2</sub> in liquid-phase and  $p$  is the partial pressure of CO<sub>2</sub> at equilibrium state: ●, 282.71 K; ■, 298.79 K; ▲, 313.73 K.**

depends on the gas partial pressure, which means that the behavior of the mixed solvents used in physical absorption also follows the Henry's law. Henry's law constants of mixed solvents are listed in **Table 6**.

Searching a solvent with high CO<sub>2</sub> absorption capacity by experiment alone would probably be very expensive and time consuming. [15] So this paper pays attention to discuss some aspects which may affect the identification

**Table 6. Comparison of the Hildebrand parameter and Henry's law constant in DMC + diethyl carbonate system, DMC + propyl acetate system, DMC + propylene carbonate system and DMC + ethylene carbonate system at 298 K in this study.**

compound	Hildebrand parameter of the mixed solvent/Mpa <sup>1/2</sup>	Henry's law constant at 298 K
DMC + ethylene carbonate	25.15	9.6612
DMC + propylene carbonate	23.7	7.8270
DMC + diethyl carbonate	19.1	5.7470
DMC + propyl acetate	19	5.4909

and selection of a potential physical solvent for CO<sub>2</sub> capture. The solubilization of a gas solute in a physical solvent can be divided into two steps [16]: first, the interactions among the solvent molecules must be broken to provide some cavities which can accommodate the solute molecules; second, the cavities formed in the first step are then filled with solute molecules and new interactions between the solute and solvent molecules are also created. In previous studies, it had been discussed that the ability to dissolve a gas solute of a solvent depended mainly on the interactions among the solvent molecules. [14] Thus, in this paper, the solvent – solvent interaction is considered to be a main factor in solvent screening, which can be described by Hildebrand solubility parameter of the solvent.

As can be seen from **Table 6**, the Henry's law constant clearly increases when the Hildebrand parameter increases, which also illustrates CO<sub>2</sub> solubility in physical solvents increases with the decreasing of Hildebrand parameter. This is mainly because in the gas dissolution process, the greater of the Hildebrand parameter value, the stronger of the solvent – solvent interactions, and the less cavities which can accommodate the solute molecules. So in order to increase the solubility of CO<sub>2</sub>, the interactions among solvent molecules must be minimized as much as possible which can create more cavities.

Furthermore, according to Hildebrand and Hansen theory, [17-19] if the interactions between solute and solvent can be neglected, Hildebrand solubility parameter

$\delta$  can reasonably used to describe and estimate the gas solubility in solvent. A relationship between the logarithm of the gas solubility and the Hildebrand solubility parameter  $\delta$  has been given by Hildebrand and other co-workers, which can be described as follows: [20] where  $\delta_s$  is the solvent solubility parameter,  $\delta_i$  is the dissolved CO<sub>2</sub> gas solubility parameter,  $f_i/f_{i,l}$  is the ratio of the fugacities of the CO<sub>2</sub> gas in its pure gaseous state to its hypothetical liquid state,  $V_{i,l}$  is the molar volume of the hypothetical liquid.  $\epsilon_s$  is the dielectric constant of the solvent,  $\alpha$  is the polarizability of the solute, and  $Q$  is the quadrupole moment of the solute. For CO<sub>2</sub> at 25°C,  $Q = 4.1 \times 10^{-26}$  e.s.u,  $\alpha = 2.65 \times 10^{-24}$  e.s.u,  $\delta_i = 12.3$  MPa<sup>1/2</sup>,  $V_{i,l} = 55 \times 10^{-6}$  m<sup>3</sup> mol<sup>-1</sup> and  $f_{i,l} = 4.12$  Mpa at 1 atm. [21] For the mixed solvent, the parameter  $\delta_s$  is replaced by  $\delta_{\text{mixed}}$ .

$$f_{i,l}(at, p) = f_{i,l}(1atm) \exp \frac{V_{i,l}(P-1)}{RT} \quad (2)$$

$$\delta_{\text{mixed}} = \sum_j^n \phi_{s,j} \delta_{s,j} \quad (3)$$

$$\epsilon_{\text{mixed}} = \sum_j^n \phi_{s,j} \epsilon_{s,j} \quad (4)$$

where  $\phi_{s,j}$  is the volume fraction of the solvent  $j$ ,  $\delta_{s,j}$  is the solubility parameter of the pure solvent  $j$  and  $\epsilon_{s,j}$  is the dielectric constant of the pure solvent  $j$ .

But the Equation (1) cited in this paper is available only at low pressure, bigger error of calculation occurs when the pressure gets higher. It is obviously important to consider the pressure impact on the application of Equation (1). So some adjustment and modification should be made to the equation, pressure correction items were introduced in Equation (1), and a new equation was proposed at high pressure in this paper, which can be described as:

where  $A$  and  $B$  are two pressure correction factors. Each value of the correction factors  $A$  and  $B$  for the four mixed solvents are listed separately in **Table 7**.

The solvent added into the DMC can be divided into two groups in this paper. Propylene carbonate and ethylene carbonate can be considered as a group of cyclic compound, and propyl acetate and diethyl carbonate can be classified as a linear compound group. As can be seen in **Table 7**, the value correction factor  $B$  is about the same, but the value correction factor  $A$  differs greatly for

$$\ln x_i = \ln(f_i/f_{i,l}) - \frac{V_{i,l}}{RT} (\delta_s - (\epsilon_s - 1)(0.1225 - 8.43 \times 10^{21} \times \alpha + 2.52 \times 10^{24} \times Q) - \delta_i)^2 \quad (1)$$

$$\ln x_i = \ln \left( A \frac{f_i}{f_{i,l}} \right) - \frac{V_{i,l}}{RT} (\delta_s - (\epsilon_s - 1)(0.1225 - 8.43 \times 10^{21} \times \alpha + 2.52 \times 10^{24} \times Q) - \delta_i)^2 + BP \quad (5)$$

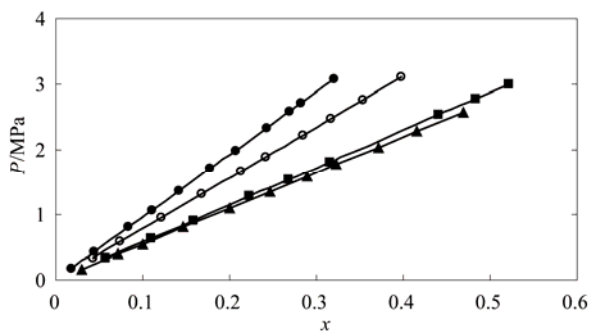
different solvents. It is also found that the Henry's law constant decreases as the correction factor A increases. Thus, the correction factor A brings about great influence on the solubility prediction of carbon dioxide in liquid solvent at high pressure. **Table 7** also shows that in the same group compound, the values of the correction factor A are similar. But for the different group, the correction factor A of the linear compound is larger than the cyclic compound, which confirms the greater solubility of CO<sub>2</sub> in linear compound mixed solvents. At the same time, it can be seen from **Figure 5**, that under the same temperature, linear compound added can greatly improved the solubility of CO<sub>2</sub> in DMC, which also indicates the larger of the correction factor A has a better absorption. In order to verify the accuracy of the Equation (5), comparisons of the measured ( $x_i$ ) and calculated ( $x_c$ ) of the CO<sub>2</sub> solubility in the four mixed solvents at 298 K were made in **Table 8** and **Table 9**.

## 5. Conclusions

Main conclusions of the study can be summarized as follows:

**Table 7. Correction factors of the DMC + diethyl carbonate system, DMC + propyl acetate system, DMC + propylene carbonate system and DMC + ethylene carbonate system at 298 K.**

Mixed solvent	A	B
propyl acetate + DMC	0.7468	0.0673
diethyl carbonate + DMC	0.7126	0.0686
propylene carbonate + DMC	0.5230	0.0688
ethylene carbonate + DMC	0.4259	0.0687



**Figure 5. Comparison of CO<sub>2</sub> solubility in mixed solvent system in this study.  $x$  is the mole fraction of CO<sub>2</sub> in liquid-phase and  $p$  is the partial pressure of CO<sub>2</sub> at equilibrium state: ■, DMC + diethyl carbonate system, 298.75 K; ▲, DMC + propyl acetate system, 298.79 K; ●, DMC + propylene carbonate system, 298.69 K; ◆, DMC + ethylene carbonate system, 298.78 K.**

**Table 8. Measured ( $x_i$ ) and calculated ( $x_c$ ) of the CO<sub>2</sub> solubility in DMC + diethyl carbonate system, DMC + propyl acetate system at 298 K.**

DMC + propyl acetate		DMC + diethyl carbonate	
$x_i$	$x_c$	$x_i$	$x_c$
0.0296	0.0296	0.0576	0.0575
0.0709	0.0708	0.1096	0.1095
0.0997	0.0997	0.1586	0.1586
0.147	0.1470	0.2225	0.2228
0.1997	0.1999	0.2680	0.2685
0.2467	0.2471	0.3150	0.3156
0.2894	0.2898	0.4410	0.4412
0.3227	0.3230	0.4837	0.4834
0.3713	0.3712	0.5210	0.5201
0.4151	0.4150		
0.4693	0.4686		

**Table 9. Measured ( $x_i$ ) and calculated ( $x_c$ ) of the CO<sub>2</sub> solubility in DMC + propylene carbonate system and DMC + ethylene carbonate system at 298 K.**

DMC + propylene carbonate		DMC + ethylene carbonate	
$x_i$	$x_c$	$x_i$	$x_c$
0.0423	0.0422	0.0179	0.0179
0.0744	0.0744	0.0444	0.0443
0.1219	0.1220	0.0836	0.0836
0.1677	0.1680	0.1107	0.1108
0.2127	0.2132	0.1417	0.1419
0.2415	0.2419	0.1778	0.1781
0.2849	0.2852	0.2067	0.2071
0.3166	0.3168	0.243	0.2432
0.3537	0.3536	0.2685	0.2685
0.3971	0.3961	0.2818	0.2817
		0.3204	0.3197

- 1) Under the pressure of 5 MPa and the temperature variations from 282 K to 313 K, the solubility data measured by constant-volume method of CO<sub>2</sub> in DMC + diethyl carbonate system, DMC + propyl acetate system, DMC + propylene carbonate system, and DMC + ethylene carbonate system decreases as temperature increases and pressure decreases.

- 2) The results show that CO<sub>2</sub> solubility in physical solvents increases with the decreasing of Hildebrand parameter. The greater of the Hildebrand parameter value, the stronger of the solvent – solvent interactions, and the less cavities which can accommodate the solute molecules.
- 3) It is also found by contrast that linear compound has a greater ability to dissolve CO<sub>2</sub> than the cyclic compound at the same temperature. Furthermore, the correction factor A brings about great influence on the solubility prediction of carbon dioxide in liquid solvent. This indicates that the correction factor A can be regarded as an main impact factor in the selection of a potential physical solvent for CO<sub>2</sub> capture.

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