

Solubilities of CO₂ in 1-Allyloxy-3-(4-Nonylphenoxy)-2-Propanol Polyoxyethylene Ethers

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Abstract

1-allyloxy-3-(4-nonylphenoxy)-2-propanol polyoxyethylene ethers (ANAPEs), a new type of absorbent, are polymeric surfactants with different adduct numbers. In this work, ANAPEs, including SN-10 with adduct number of 10 and SN-15 with adduct number of 15, were prepared for CO₂ absorption using the isochoric saturation method.

Densities of the ANAPEs at atmospheric pressure were measured by a 5.567 ± 0.004 cm³ pycnometer, which decreased with increased temperature.

Solubility data of CO₂ in ANAPEs were measured within the pressure range of 0–600.0 kPa and temperature range of 303.15–323.15 K at 10 K intervals and could be calculated on the basis of experimental data of p , x_{CO_2} and b_{CO_2} . The solubility of CO₂ in absorbents increased linearly with increasing pressure and decreased with increasing temperature at all the pressures. The solubility of CO₂ in SN-15 is the highest at all temperatures, but almost the same with SN-10 at 303.15 K over pressures ($p < 350$ kPa), which indicates physical dissolution process. Henry's constants were determined from solubility data. With increasing temperature, Henry's constants increased.

Thermodynamics of CO₂ absorption were calculated including enthalpy, entropy, and Gibbs energy. The absolute value of $\Delta_{\text{sol}}H$ based on H_x of SN-15 is largest at 303.15 K and indicates stronger SN-15/CO₂ interactions, consistent with solubility of CO₂ based on H_x . The negative enthalpy demonstrated exothermic process, which means the dissolution of CO₂ in ANAPEs is favourable enthalpically. The $\Delta_{\text{sol}}G$ shows positive value.

Keywords

Ionic liquids, solubility, Henry's law constant, thermodynamics, Gibbs energy

1 Introduction

As one of the most abundant greenhouse gases, CO₂ has achieved great attention in academic and industrial areas. A number of separation technologies have been applied to capture CO₂, and liquid absorption is the most widely used method.^{1,2} Aqueous solutions of alkanolamines, as the most effective choice in CO₂ absorption technologies, has great superiority in removing CO₂.^{3–5} However, the inherit weaknesses, such as corrosion, high regeneration energy, and secondary pollution, made it essential to develop the alternatives.^{6–8} In recent years, ionic liquids (ILs), with features of non-volatility, thermal stability, and tuneable properties,⁹ were proposed as the favoured options to overcome some of the problems with amines.^{10–12} In addition, it is also inevitable to develop more versatile alternatives with respect to the price and environmental benignness.^{13–15}

Research effects have been directed toward polymeric surfactants owing to their potential applications in many fields.^{16–21} Compared with ILs, the surfactants are cheap, low-toxic, and structurally tunable.²² In our previous work, we have investigated the solubility of CO₂ in a series of

fatty amine polyethylene ethers (FAPEs) with various oxoethylene (EO) chain numbers at $T = (303.15, 313.15$ and $323.15)$ K, and $p = (100$ to $550)$ kPa. The results demonstrated high solubility of CO₂ in FAPEs with high EO content. Currently, the surfactants systems used in CO₂ absorption are vacant and it is significant to develop and diversify CO₂ absorbents with different structures.²³

1-allyloxy-3-(4-nonylphenoxy)-2-propanol polyoxyethylene ethers (ANAPEs) are polymeric surfactants with different adduct numbers. This work introduces a new type of absorbent and provides the thermodynamics in polymeric surfactants with different adduct numbers. The solubility of CO₂ in SN-10 and SN-15 were studied in a pressure range of 0–600 kPa, and temperature range of 303.15–323.15 K. Henry constants were determined from the solubility data. Thermodynamics of CO₂ absorption were calculated, including enthalpy, entropy, and Gibbs energy.

2 Experimental

2.1 Materials

The raw materials of 1-allyloxy-3-(4-nonylphenoxy)-2-propanol polyoxyethylene ethers SN-10 (≥ 0.99995) and SN-15 (≥ 0.99995) were provided by Foshan Kedi Chemi-

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cal Company (Foshan, China). They were vacuum dried at 350 K for 24 h to remove volatile impurities. The ANAPes' densities at atmospheric pressure were gauged in detail in the temperature range of 303.15–323.15 K at 10 K intervals with a $5.567 \pm 0.004 \text{ cm}^3$ pycnometer. The pycnometer was calibrated beforehand, using double distilled water at 303.15 K and then immersed in an oil-bath. CO₂ with the mass fraction exceeding 0.99995 was provided by Jing-gong Special Gas Co., Ltd.

2.2 Apparatus

The stainless steel apparatus was an upgraded version, based on our previous glass equipment,²⁴ as shown in Fig. 1.

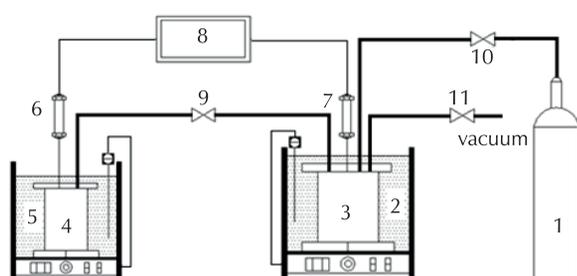


Fig. 1 – Schematic diagram of the CO₂ solubility apparatus: 1 – CO₂ gas cylinder; 2, 5 – thermostatic water bath and magnetic stirrer; 3 – CO₂ gas reservoir; 4 – CO₂ gas equilibrium cell; 6, 7 – pressure transmitter; 8 – digital indicator; 9, 10, 11 – valve

The apparatus included a CO₂ gas cylinder (1), two water baths (2, 5), a CO₂ gas reservoir (GR, 3) with magnetic stirrer, a CO₂ gas equilibrium cell (EC, 4) with magnetic stirrer, and two pressure transmitters (6, 7). The volumes of EC and GR were determined using the previous method²⁵ with the results of 141.61 cm³ and 370.99 cm³, respectively. The temperatures of water baths were accurately controlled with a precision of $\pm 0.05 \text{ K}$. The pressures were monitored by pressure transmitter (Fujian Wide Plus Precision Instruments Co., Ltd, Wide Plus-8, 0 to 600.0 kPa, with an accuracy of 0.1 % full scale).

2.3 Methods

The CO₂ solubility was measured using the *isochoric* saturation method.²⁵ The weights of the ANAPes were measured using an electronic analytical balance (Mettler-Toledo AL204) with an uncertainty of $2 \cdot 10^{-4} \text{ g}$. Temperatures were controlled by two water baths. After vacuum drying for 24 h at 350 K, approximately 15–40 g ANAPes were loaded into EC and degassed under vacuum at 343.15 K while stirring for 1 h. The entire system was controlled at a specified oven temperature using water baths, and evacuated to pressures p_1 for 1 h after cooling. The pressure then reached the scheduled value p_2 by feeding the CO₂ from gas cylinder into GR. By opening the valve between

EC and GR, the CO₂ was brought into EC and absorbed by ANAPes with magnetic stirring, which can facilitate CO₂ absorption. When the pressure of the EC reached equilibrium, which was normally after 4 h, the equilibrium pressures of CO₂ in EC and GR were recorded as p_3 and p_4 , respectively. The amount of absorbed CO₂ could then be calculated from the difference between the initial pressure of the GR and the final pressures of EC and GR. With the same procedure, the repeated experiments were fulfilled at the same equilibrium temperature.

3 Results and discussion

3.1 Density

The densities of the ANAPes at atmospheric pressure were measured by a $5.567 \pm 0.004 \text{ cm}^3$ pycnometer, which was calibrated in advance with double distilled water at 303.15 K. The results demonstrated that the densities of ANAPes decreased when increasing the temperature, as shown in Table 1.

Table 1 – Densities of ANAPes' solutions

T/K	$\rho/\text{g cm}^{-3}$	
	SN-10 	SN-15
303.15	1.0157	1.1037
313.15	1.0100	1.0299
323.15	1.0050	1.0234

3.2 Solubility data of CO₂ in ANAPes

Table 2 shows the solubility of CO₂ in ANAPes within the pressure range of 0–600.0 kPa at temperature range 303.15–323.15 K. In Table 2, p , x_{CO_2} and b_{CO_2} stand for CO₂ equilibrium pressure above the liquid absorbent, amount fraction of CO₂ in liquid phase, and molality of CO₂ in liquid phase, respectively.

CO₂ solubility in ANAPes could be calculated based on the experimental data of p , x_{CO_2} and b_{CO_2} . Due to the low pressure of ANAPes, its effect on CO₂ solubility could be neglected, and the gas phase was assigned to pure CO₂. Therefore, the amount of absorbed CO₂ can be calculated by

$$n_{\text{CO}_2} = n_0 - n_1 - n_2 \quad (1)$$

where n_{CO_2} is amount of absorbed CO₂ in ANAPes, n_0 is the initial amount of CO₂ in GR, n_1 and n_2 are equilibrium amounts of CO₂ in GR and EC, respectively. The values of n_0 , n_1 and n_2 were obtained from Soave-Redlich-Kwong (SRK) equation on the basis of experimental PVT data. The volume of liquid in EC was derived directly from the mass and density of ANAPes at different temperatures, and the volume expansion of liquid in EC could be neglected.

Table 2 – Solubility data of CO₂ in ANAPEs at different temperatures

System	303.15 K			313.15 K			323.15 K		
	p/kPa	$x_{\text{CO}_2}/10^{-3}$	$b_{\text{CO}_2}/10^{-3} \text{ mol kg}^{-1}$	p/kPa	$x_{\text{CO}_2}/10^{-3}$	$b_{\text{CO}_2}/10^{-3} \text{ mol kg}^{-1}$	p/kPa	$x_{\text{CO}_2}/10^{-3}$	$b_{\text{CO}_2}/10^{-3} \text{ mol kg}^{-1}$
SN-10	106.9	42.70	57.57	103.9	36.30	48.60	106.0	31.60	42.11
	205.5	80.00	112.2	209.0	70.10	97.27	213.5	61.80	84.96
	306.5	117.4	171.6	305.1	101.9	146.4	309.0	89.90	127.4
	408.7	152.9	232.9	394.2	130.4	193.5	418.9	120.2	176.3
	501.3	184.6	292.1	446.5	149.5	226.7	510.3	144.9	218.7
	117.0	46.40	48.89	118.0	43.10	45.27	114.3	36.40	37.89
SN-15	232.6	89.50	98.82	218.0	76.30	83.0	214.2	67.50	72.66
	324.5	123.5	141.7	313.9	112.1	126.8	313.5	98.2	109.4
	414.0	160.6	192.3	414.3	146.8	172.9	411.5	130.2	150.3
	528.0	198.2	248.4	512.1	168.4	203.4	517.1	170.3	206.2

The molar fraction (x_{CO_2}) and molality (b_{CO_2}) of CO₂ were obtained by the following equations:

$$x_{\text{CO}_2} = n_{\text{CO}_2} / (n_{\text{CO}_2} + n_{\text{ANAPE}}) \quad (2)$$

$$b_{\text{CO}_2} = n_{\text{CO}_2} / m_{\text{ANAPE}} \quad (3)$$

where n_{ANAPE} is the amount of ANAPE and m_{ANAPE} is the mass of ANAPE.

Fig. 2 shows the dependence of the CO₂ solubility on temperatures and pressures. It is evident that the solubility of CO₂ in SN-10 is positive linear with CO₂ pressure, and elevating temperature decreased the absorption of CO₂. Fig. 3 shows that the dependence of CO₂ solubility on pressure in SN-15 was similar to that in SN-10, but SN-15 had a higher capacity of CO₂ than SN-10. All these phenomena demonstrated that the absorption process of CO₂ in ANAPEs might be merely physical.²⁶

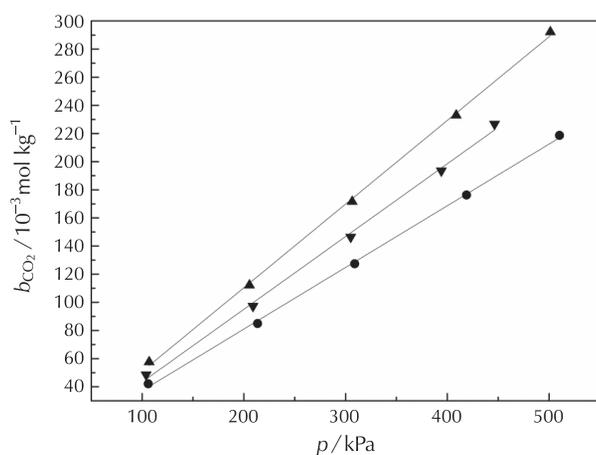


Fig. 2 – Solubility of CO₂ in SN-10 at different temperatures: ▲ – $T = 303.15 \text{ K}$; ▼ – $T = 313.15 \text{ K}$; ● – $T = 323.15 \text{ K}$, — — linear fit

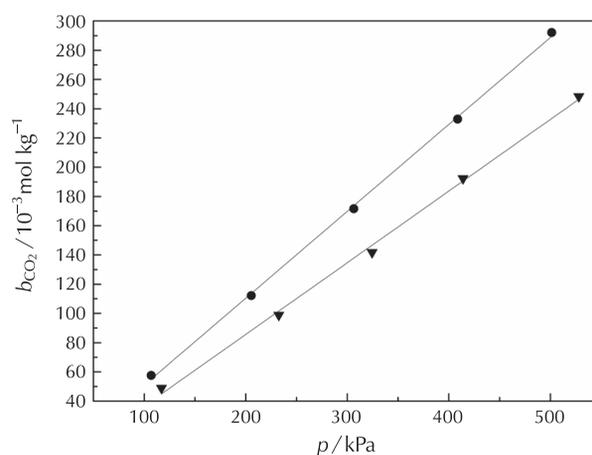


Fig. 3 – CO₂ solubility as a function of CO₂ equilibrium pressure at $T = 303.15 \text{ K}$ in: ▲ – SN-10; ▼ – SN-15, — — linear fit

3.3 Henry constants

Henry's constant, which is presented in terms of H_x or H_b , is the key parameter for designing the gas absorption process.²⁷ In this work, the gas phase was assumed to comprise pure CO₂ and its pressure was relatively low, which means the fugacity of gas was approximately equal to equilibrium pressure of CO₂.^{28,29} Therefore, Henry's constants of CO₂ in ANAPEs can be obtained from the experimental data of CO₂ solubility. As shown in Table 3, both H_x and H_b increased with the temperature, which means that the CO₂ solubility decreased with increasing temperature, as shown in Fig. 2. Additionally, Henry's constant H_x of CO₂ in SN-15 is lower than that in SN-10 under the same temperature, which means SN-15 possesses a higher CO₂ capacity than SN-10.

Table 3 – Experimentally determined Henry's constants (H_x , based on amount fraction; H_b , based on molarity) of CO₂ in solutions at various temperatures

Solutions	H_x /Mpa			H_b /Mpa kg mol ⁻¹		
	303.15 K	313.15 K	323.15 K	303.15 K	313.15 K	323.15 K
SN-10	2.668	2.996	3.488	1.749	2.027	2.376
SN-15	2.624	2.907	3.107	2.180	2.483	2.658

3.4 Thermodynamics

Temperature had a significant effect on Henry's constant, and thus an empirical equation was used to describe this relationship as

$$\ln \frac{H(T)}{0.1 \text{ MPa}} = \sum_{i=0}^n B_i (T/K)^{-i} \quad (4)$$

where B_i is the optimized coefficient and can be obtained using a linear regression of multiple-variables calculation. The values of B_0 – B_2 are listed in Table 4. The thermodynamic properties of the absorption process of CO₂ in ANAPes can then be calculated from the following equations:

$$\Delta_{\text{sol}}G = RT \ln \frac{H(T,p)}{p^\circ} \quad (5)$$

$$\Delta_{\text{sol}}H = R \left(\frac{\partial \ln[H(T,p)/p^\circ]}{\partial (1/T)} \right)_p \quad (6)$$

$$\Delta_{\text{sol}}S = \left(\frac{\Delta_{\text{sol}}H - \Delta_{\text{sol}}G}{T} \right) \quad (7)$$

where $\Delta_{\text{sol}}G$, $\Delta_{\text{sol}}H$, $\Delta_{\text{sol}}S$ are the standard Gibbs energy, enthalpy and entropy changes under the pressure of 0.1 MPa, respectively. The changes in thermodynamic properties at 303.15 K and 0.1 MPa are listed in Table 5.

$\Delta_{\text{sol}}H$ is related with the interaction between gas and liquid, and its negative value indicates that the absorption process of CO₂ is exothermic. According to the molecular points, $\Delta_{\text{sol}}S$ represents the degree of order between solute and solvent.³⁰ From Table 5, the absolute value of $\Delta_{\text{sol}}H$ of SN-15 is larger than that of SN-10, which means that there is a stronger interaction between CO₂ and SN-15. Additionally, a higher negative value of SN-15 indicates its interaction with CO₂. These account for the behaviour as shown in Fig. 3.

Table 4 – Values of coefficients B_0 , B_1 , and B_2 for the equation

Solutions	B_0	B_1	B_2
SN-10	15.51783	–6241.62	767099.3
SN-15	–12.31848	10650.53	–1796230

Table 5 – Calculated Gibbs energy, enthalpy and entropy of the solutions at 0.1 MPa and 303.15 K

Solution	ΔG /kJ mol ⁻¹	ΔH /kJ mol ⁻¹	ΔS /J mol ⁻¹ K ⁻¹
SN-10	8.27691	–9.81687	–59.6859
SN-15	8.23495	–9.97605	–60.0726

4 Conclusion

In this work, 1-allyloxy-3-(4-nonylphenoxy)-2-propanol polyoxyethylene ethers (ANAPes) with adduct numbers (10, 15) were prepared for CO₂ absorption within a pressure range of 0–600.0 kPa and a temperature range of 303.15–323.15 K using the isochoric saturation method. When increasing temperature, the solubility of CO₂ in absorbents decreased and Henry's constants increased. The absolute value of $\Delta_{\text{sol}}H$ based on H_x of SN-15 is the highest at 303.15 K and indicates stronger SN-15/CO₂ interactions, which is consistent with the solubility of CO₂ based on H_x . The negative enthalpy demonstrated an exothermic process, which means the dissolution of CO₂ in ANAPes is favourable enthalpically. The $\Delta_{\text{sol}}G$ shows positive value.

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List of abbreviations and symbols

ANAPE	– 1-allyloxy-3-(4-nonylphenoxy)-2-propanol polyoxyethylene ether
EC	– equilibrium cell
EO	– oxyethylene
FAPE	– fatty amine polyethylene ether
GR	– gas reservoir
IL	– ionic liquid
SRK	– Soave-Redlich-Kwong
b_{CO_2}	– molality of CO ₂ in liquid phase, mol kg ⁻¹

B_i	– optimized coefficient
$\Delta_{\text{sol}}G$	– standard Gibbs energy of solution, kJ mol ⁻¹
$\Delta_{\text{sol}}H$	– enthalpy of solution, kJ mol ⁻¹
$\Delta_{\text{sol}}S$	– entropy of solution, J mol ⁻¹ K ⁻¹
H_b	– Henry's constants based on molarity, Mpa kg mol ⁻¹
H_x	– Henry's constants based on amount of substance fraction, Mpa
m_{ANAPES}	– mass of ANAPE
n_0	– initial amount of CO ₂ in GR, mol
n_1	– equilibrium amount of CO ₂ in GR, mol
n_2	– equilibrium amount of CO ₂ in EC, mol
n_{ANAPES}	– amount of ANAPE
n_{CO_2}	– amount of absorbed CO ₂ in ANAPE, mol
P	– pressure, kPa
p_1	– residual gas pressure in EC, kPa
p_2	– pressure of scheduled value, kPa
p_3	– equilibrium pressures of CO ₂ in EC, kPa
p_4	– equilibrium pressures of CO ₂ in GR, kPa
p	– CO ₂ equilibrium pressure above the liquid absorbent, kPa
T	– temperature, K
x_{CO_2}	– amount fraction of CO ₂ in liquid phase
ρ	– density, g cm ⁻³

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SAŽETAK

Topljivost CO₂ u eterima

1-aliloksi-3-(4-nonilfenoksi)-2-propanola i polioksietilena

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Ispitana je topljivost ugljikova dioksida u eterima 1-aliloksi-3-(4-nonilfenoksi)-2-propanola i polioksietilena (ANAPE), (SN-10 i SN-15) u izohornim uvjetima pri rasponu tlakova 0–600 kPa i temperatura 303,15–323,15 K. Topljivost CO₂ raste s tlakom, a pri svim tlakovima opada s temperaturom. U cijelom temperaturnom rasponu topljivost je veća u SN-15, ali pri 303,15 K i tlakovima nižim od 350 kPa gotovo je izjednačena s topljivošću u SN-10 što ukazuje na fizikalni mehanizam otapanja. Određene su Henryjeve konstante i termodinamika apsorpcije, uključujući entalpiju, entropiju i Gibbsovu energiju. Prema negativnim vrijednostima entalpije otapanje CO₂ u eterima ANAPE je egzoterman proces.

Ključne riječi

Ionske tekućine, topljivost, Henryjeva konstanta, termodinamika, Gibbsova energija

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