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Kinetics study of CO$_2$ absorption in potassium carbonate solution promoted by diethylenetriamine

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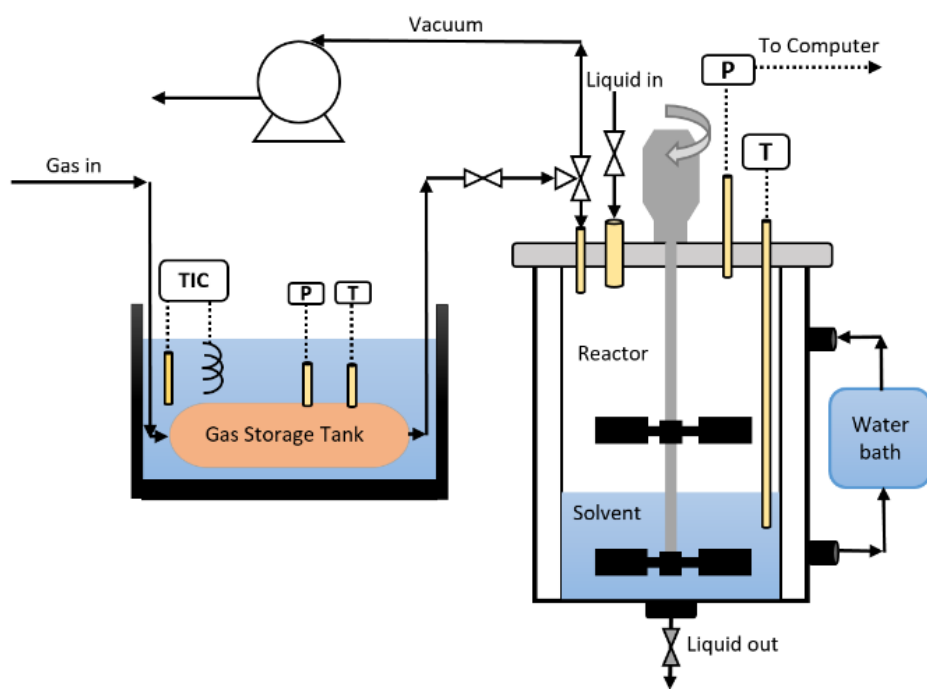
Kinetics study of CO_2 absorption in potassium carbonate solution promoted by diethylenetriamine

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Abstract

In this work, characterization and kinetics of CO_2 absorption in potassium carbonate (K_2CO_3) solution promoted by diethylenetriamine (DETA) were investigated. Kinetics measurements were performed using a stirred cell reactor in the temperature range of 303.15-323.15 K and total concentration up to 2.5 kmol/m^3. The density, viscosity, physical solubility, CO_2 diffusivity and absorption rate of CO_2 in the solution were determined. The reaction kinetics between CO_2 and K_2CO_3 + DETA solution were examined. Pseudo-first order kinetic constants were also predicted by zwitterion mechanism. It was revealed that the addition of small amounts of DETA to K_2CO_3 results in a significant enhancement in CO_2 absorption rate. The reaction order and activation energy were found to be 1.6 and 35.6 kJ/mol, respectively. In terms of reaction rate constant, DETA showed a better performance compared to the other promoters such as MEA, EAE, proline, arginine, taurine, histidine and alanine.

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1. Introduction

The chemical manufacturing plants release a huge amount of greenhouse gases, particularly carbon dioxide (CO_2), into the atmosphere which causes environmental issues e.g. climate change [1, 2]. Therefore, the reduction of CO_2 emissions is essential to mitigate impacts of climate change. Chemical absorption is one of the most efficient technologies for capture of CO_2 from gas streams [3]. The operational efficiency of the chemical absorption process significantly depends on solvent's characteristics such as absorption rate, loading capacity, regeneration energy, corrosion tendency, thermal and chemical stability [4]. The alkanolamines, particularly monoethanolamine (MEA) is usually used as an absorbent for CO_2 capture [5]. However, MEA has several drawbacks such as corrosion issues, high regeneration energy, low CO_2 loading capacity, high volatility and toxicity [5]. Therefore, research on other type of solvents with better absorption performance is essential.

Potassium carbonate (K_2CO_3) solution is an alternative to alkanolamines because of its advantages e.g. less corrosive, low volatility, low toxicity and less regeneration energy [6]. However, the main disadvantage associated with this solvent is the low reaction rate with CO_2 [7]. Several researchers showed that the addition of an amine with high absorption rate (as a promoter) to K_2CO_3 can improve its absorption performance. For instance, Bhosale et al. [8] studied the effect of addition of ethylaminoethanol (EAE) to K_2CO_3 solution on CO_2 absorption rate. They found that K_2CO_3 + EAE solution has a higher absorption rate than single K_2CO_3. The reaction order and activation energy were obtained to be 1 and 81.7 kJ/mol, respectively. Thee et al. [9] added proline and glycine to K_2CO_3 solution and compared their performances with several promoters. It was discovered that both proline and glycine enhanced the reaction kinetics between CO_2 and K_2CO_3. In addition, it was observed that proline has faster kinetics than DEA,

MEA and glycine but lower than sarcosine. Shen et al. [10] used arginine to improve the absorption rate of K_2CO_3 solution. The results revealed that arginine + K_2CO_3 solution has a better absorption rate compared to pure K_2CO_3. The primary amino group in the structure of arginine and its basic character are two main reasons for this positive effect [10]. Based on their observation, arginine showed a better performance than histidine, lysine and boric acid. Kim et al. [11] investigated the absorption rate of CO_2 in PZ + K_2CO_3 and 2MPZ + K_2CO_3 solutions. They observed that both 2-methylpiperazine (2MPZ) and piperazine (PZ) have similar absorption rate and can be considered as potential promoters to increase the CO_2 absorption rate in K_2CO_3. The performance of these blended solutions was shown better than MEA and AMP at high partial pressure of CO_2. Kang et al. [12] studied the absorption rate of CO_2 into K_2CO_3 promoted by sarcosine, and found that the addition of sarcosine to K_2CO_3 results in a significant enhancement in CO_2 absorption rate. The kinetics of CO_2 absorption in K_2CO_3 + MEA solution were evaluated by Thee et al. [13]. They reported kinetics parameters and compared its performance with several promoters. The reaction order and activation energy were determined to be equal to 1 and 44.9 kJ/mol, respectively. It was also concluded that the addition of promoters to potassium carbonate has a positive effect on rate of absorption of CO_2 and make it an interesting solvent for CO_2 capture. Generally, these promoters can be classified into three main categories, including inorganic, organic and enzymatic promoters. Based on above observation, it was found that organic promoters have better performance than other promoters. For this reason, the most widely studied promoters for potassium carbonate in the literature are organic promoters such as amines and amino acids. In addition, these studies showed that reaction kinetics between CO_2 and K_2CO_3 + promoter can be explained by zwitterion mechanism.

There is always a motivation in finding better promoters to enhance the CO_2 absorption rate of K_2CO_3 solution. Hartono et al. [14] measured the kinetics of CO_2 absorption diethylenetriamine (DETA) solution and revealed that DETA has a higher CO_2 absorption rate compared to MEA and AEEA. For this reason, DETA was chosen in this work as a promoter to be added to K_2CO_3 solution. Therefore, an aqueous blend of solutions of K_2CO_3 and DETA has been proposed as a new absorbent for CO_2 absorption. The objective of this work is to study experimentally the effect of the addition of DETA to K_2CO_3 on the CO_2 absorption rate and to compare its performance with other common promoters. Thus, the kinetics of the reaction between CO_2 and K_2CO_3 + DETA solution were studied. The density, viscosity, CO_2 solubility and diffusivity and the rate of absorption of CO_2 into solution were measured at 303.15, 313.15 and 323.15 K. Then, the kinetics parameters such as reaction rate constant, reaction order and activation energy were reported and the effect of temperature and concentration on these parameters was studied.

2. Kinetic study

The chemical reactions during the absorption of CO_2 in K_2CO_3 + DETA solution [15, 16] was described in the supporting information. To study the kinetics of CO_2 with K_2CO_3 + DETA, it is important that condition given in Eq. (1) be satisfied [17]. This condition shows that reaction regime is the fast pseudo-first-order reaction.

$$3 < H_a \ll E_i \tag{1}$$

Where the values of Hatta number (Ha) and enhancement factor (Ei) are calculated by:

$$H_a = \frac{\sqrt{K_{ov} \, D_{CO_2}}}{K_L} \tag{2}$$

$$E_i = 1 + \frac{D_{solvent} \, [solvent]}{b \, D_{CO_2} \, [CO_2]_i} \tag{3}$$

The CO_2 physical solubility in the solution was calculated by Henry's law:

$$[CO_2]_i = \frac{P_{CO_2}}{H_e} \tag{4}$$

The flux of the absorbed gas in solution can be obtained by [18]:

$$N_{CO_2} = E_A K_L ([CO_2]_i - [CO_2]) \tag{5}$$

In the fast pseudo-first-order reaction, E_A is equal to Hatta number, and consequently, the specific absorption rate can be determined by:

$$N_{CO_2} = \sqrt{K_{OV} D_{CO_2}} \ \frac{P_{CO_2}}{H_e} \tag{6}$$

This equation was used to find the overall reaction rate constant. Then, the apparent reaction rate constant can be obtained as follows:

$$k_{app} = k_{ov} - k_{OH^-}[OH^-] = k_{DETA} [DETA]^n \tag{7}$$

By plotting of log (k_{app}) vs. log (DETA), k_{DETA} and n can be determined.

3. Materials and Methods

3.1. Materials

The potassium carbonate and diethylenetriamine with purity of 99% were purchased from Acros in Italy. The N_2O, N_2 and CO_2 gases with purity of 99.9% were also supplied by Air Liquid (Italy). Four different mixed solutions with concentrations of 2 M K_2CO_3 + 0.2 M DETA, 2 M K_2CO_3 + 0.3 M DETA, 2 M K_2CO_3 + 0.4 M DETA and 2 M K_2CO_3 + 0.5 M DETA were selected in this study as absorbents.

3.2. Density and viscosity measurements

An Ubbelohde viscometer and Gay-Lussac pycnometer were used in this work to measure viscosity and density of the solutions, respectively. The procedure for density and viscosity measurements is the same as our previous publications [19, 20, 21]. The measurements were

performed at different temperatures of 303-323 K and concentrations of 2 M K_2CO_3 + (0.2-0.5) M DETA.

3.3. CO_2 absorption rate measurement

The absorption rate measurements were conducted by a stirred cell reactor. A schematic diagram of the equipment was given in **Fig. 1**. This experimental setup consisted of an equilibrium cell, water bath, gas storage tank, magnetic stirrer, temperature indicator, vacuum pump and pressure transmitter. The experimental method for CO_2 absorption rate measurement and the equipment used in this work are similar to that described in our previous publications [21-23]. The double jacketed stirred cell reactor with a plane interface which was purchased from Buchiglas (Switzerland) is main part of the experimental setup. The reactor temperature was kept constant by a jacket. The reactor was also equipped with external magnetic motor, pressure transmitter and temperature sensor. The solution in the reactor is stirred using impeller mounted on shaft of magnetic motor. The temperatures of the solution and gas storage tank were controlled within ±0.1 K using a water bath. The reactor was first purged with Nitrogen gas to remove air. Then, the solution of K_2CO_3 + DETA was charged in the reactor. CO_2 was injected to gas storage tank to be heated to the desired temperature and was transferred to the reactor. Once CO_2 was injected, the stirrer is switched on at a constant speed. The pressure inside the reactor because of reaction between CO_2 and K_2CO_3 + DETA solution decreases. The pressure decrease versus time was recorded by pressure transmitter and slope of pressure plot versus time ($\frac{dP_{CO_2}}{dt}$) was used to calculate CO_2 absorption rate. A knowledge of gas phase volume in the reactor (V), the gas-liquid interfacial area (A), temperature (T) and therefore enable CO_2 absorption rate to be calculated.

$$N_{CO_2} = -\frac{V}{R\,T\,A}\,\frac{dP_{CO_2}}{dt} \tag{8}$$

6

Fig. 1

4. Results and discussion

4.1. Density and viscosity

The density and viscosity of water at temperatures of 303 to 323 K were measured and compared with the literature [24, 25] to check the validity of our procedure. According to **Table 1**, a good agreement between our results and literature were observed. The obtained densities and viscosities of K_2CO_3 + DETA were given in **Fig. 2** and **Fig. 3**, respectively. As it can be seen, both of these parameters increase as the concentration of DETA increases and decreases as temperature increases.

Table 1

Fig. 2

Fig. 3

4.2. Reaction kinetics of CO_2 absorption in K_2CO_3+DETA

The values of solubility (H_{CO_2}) and diffusivity of CO_2 (D_{CO_2}) in K_2CO_3 + DETA solution are required to obtain the kinetics parameters and to analyze the results of CO_2 absorption rate. Since CO_2 undergoes a chemical reaction with the K_2CO_3 + DETA solution, H_{CO_2} and D_{CO_2} in solution cannot be directly measured [26]. It has been proved by many works that the N_2O analogy can be used to estimate the values of H_{CO_2} and D_{CO_2} in amines [26, 27]. Therefore, this method and modified Stokes-Einstein equation were applied in this work to calculate diffusivity and H_{CO_2} in K_2CO_3 + DETA solutions [27].

$$\left(\frac{D_{CO_2}}{D_{N_2O}}\right)_{solution} = \left(\frac{D_{CO_2}}{D_{N_2O}}\right)_{H_2O} \tag{9}$$

$$\left(\frac{H_{CO_2}}{H_{N_2O}}\right)_{solution} = \left(\frac{H_{CO_2}}{H_{N_2O}}\right)_{H_2O} \tag{10}$$

7

The solubility and diffusivity of CO_2 and N_2O in water were determined using Eqs. (11-14):

$$D_{CO_2,H_2O} = 2.35 \times 10^{-6} \exp\left(\frac{-2119}{T}\right) \tag{11}$$

$$D_{N_2O,H_2O} = 5.07 \times 10^{-6} \exp\left(\frac{-2371}{T}\right) \tag{12}$$

$$H_{CO_2,H_2O} = 2.8249 \times 10^6 \exp\left(\frac{-2044}{T}\right) \tag{13}$$

$$H_{N_2O,H_2O} = 8.547 \times 10^6 \exp\left(\frac{-2284}{T}\right) \tag{14}$$

Also, N_2O diffusion in solution was determined by Eq. (15):

$$D_{N_2O,solvent} = D_{N_2O,H_2O} \times \left(\frac{\mu_{H_2o}}{\mu_{solvent}}\right)^{0.6} \tag{15}$$

The H_{N_2O} in the solution was calculated from the correlations proposed by Schumpe [28] and Wiesenberger [29]. Then, using these results and Eqs. (9-15) H_{CO_2} and D_{CO_2} in K_2CO_3 + DETA solution can be estimated. To confirm the results and also measurement method in this work, H_{CO_2} and D_{CO_2} in 35wt% K_2CO_3 solution at 303 K were calculated and compared to with those published previously. For example, the obtained data in this work for H_{CO_2} and D_{CO_2} in K_2CO_3 are 2.68×10^4 (kPa.m^3/kmol) and 0.74×10^{-9} (m^2/s), respectively. These data show a good agreement with 2.55×10^4 (kPa.m^3/kmol) and 0.83×10^{-9} (m^2/s) which were reported by Shen et al. [10]. The H_{CO_2} and D_{CO_2} in K_2CO_3 + DETA solution at 303, 313 and 323 K calculated in this study were plotted in **Fig.4** and **Fig. 5**. As can be observed in **Fig. 4**, D_{CO_2} increases with temperature and decreases with increasing DETA concentration. The reason for this is that viscosity of the solution is higher at higher concentrations and lower temperatures. The effect of temperature and concentration on the solubility of CO_2 in the solution were investigate in **Fig. 5**. It can be seen that H_{CO_2} increases from 1.45×10^4 to 2.16×10^4 (kPa.m^3/kmol) when temperature and concentration increases.

Fig. 4

Fig. 5

To verify the reliability of the results and also measurement method for kinetics study, initial runs were made for the CO_2 absorption rate in single K_2CO_3 solution and compared with the literature. The measured CO_2 absorption rate in K_2CO_3 solution at 303 K by stirred cell reactor in this study was found to be 0.64×10^{-6} kmol/m^2.s, which is in excellent agreement with the data given by Bhosale et al. (0.50×10^{-6} kmol/m^2.s) [8]. The absorption rate of CO_2 in K_2CO_3 solution promoted by DETA was then measured at different temperatures, concentrations and agitation speeds, and the results were shown in **Figs. 6-8**. The value of CO_2 absorption rate as a function of agitation speed at 313 K and at two different concentration of DETA were illustrated in **Fig. 6**. It is clear from this figure that the absorption rate of CO_2 is independent of the speed of agitation which shows reaction is in the pseudo-first-order regime [8]. **Fig. 7** shows the effect of addition of DETA to 2 kmol/m^3 K_2CO_3 solution on absorption rate of CO_2 at 313.15 K. The CO_2 absorption rate increases as DETA concentration increases from 0.2 to 0.5 kmol/m^3. In addition, the results indicated that all the promoted solutions has higher absorption rate than pure K_2CO_3. This can be due to the structure of DETA which has three amino groups. The amino groups make DETA a very reactive amine [14]. The effect of temperature on CO_2 absorption rate of K_2CO_3 + DETA solution was also shown in **Fig. 8**. It is observed that, temperature actually has a positive effect on absorption rate. The decrease in viscosity with temperature and increase in reaction rate constant are two reasons for this positive effect.

Fig. 6

Fig. 7

Fig. 8

9

The values of overall reaction rate constant (k_{ov}) for $CO_2+K_2CO_3+DETA+H_2O$ system were determined from Eq. (17) and the results were presented in **Fig. 9** and listed in **Table 2**. As can be observed in this figure, k_{ov} increases with increasing of DETA concentration.

Fig. 9

Table 2

Using these values of k_{ov} and Eq. (18), apparent reaction rate constant (k_{app}) were determined at different temperatures. The reaction order and reaction rate constant can be calculated from the plot of log (k_{app}) versus log (DETA) as shown in **Fig. 10**. The value of the reaction order with respect to DETA determined in this study at 313.15 K was found to be 1.61 which is in good agreement with the 1.7 value reported by Hartono et al. [14]. In addition, values of reaction rate constant at temperatures 303, 313 and 323 K were calculated and the results at 313 k were depicted in **Fig. 11.** The activation energy which was calculated using Arrhenius regression of reaction rate constant to be 35.6 kJ/mol, and reaction rate constant was correlated as follows:

$$k_{DETA} = 3.99 \times 10^{10} \exp\left(\frac{-4285.2}{T}\right) \tag{16}$$

Fig. 10

Fig. 11

The liquid mass transfer coefficient was calculated using correlation given by Littel et al. [30] and the results were presented in **Table 2**. This parameter is necessary to calculate Hatta number and enhancement factor.

$$\frac{K_L d_s}{D_{CO_2}} = 0.3929 \left(\frac{n_s d_s^2 \rho}{\mu}\right)^{0.6632} \left(\frac{\mu}{\rho D_{CO_2}}\right)^{0.33} \tag{17}$$

Fig. 12 shows the calculated Hatta number and enhancement factor for CO_2 absorption in K_2CO_3 + DETA solution at 313.15 K. It is clear that, Hatta numbers are greater than 3 and lower than enhancement factor which satisfies condition given in Eq. (1).

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Fig. 12

A comparison between predicted and measured absorption rates of CO_2 was illustrated in **Fig. 13**. As can be seen, experimental results are in excellent agreement with model calculations with the average absolute deviation percent (AAD) about 3%.

$$\%AAD = 100 \times \frac{1}{n} \sum_{i=1}^{i=n} \left| \frac{\left|(N_{CO_2})_{exp} - (N_{CO_2})_{cor}\right|}{(N_{CO_2})_{exp}} \right| \tag{18}$$

The performance of different promoters in terms of enhancement reaction rate of K_2CO_3 with CO_2 was investigated at 313.15 K and shown in **Fig. 14**. DETA shows a higher absorption rate in comparison with proline [31], EAE [8], arginine [10], MEA [10], taurine [32], histidine [33], alanine [34] and AMP [10]. It can be concluded that the addition of DETA to K_2CO_3 results in significant enhancement in CO_2 absorption rate compared to the investigated promoters. Therefore, DETA can be considered as a promising amine in order to improve reaction kinetics of potassium carbonate with CO_2.

Fig. 13

Fig. 14

5. Conclusion

In this work, reaction kinetics between K_2CO_3 + DETA solution and CO_2 was studied using a stirred cell reactor. The solubility and diffusivity of CO_2 were estimated using N_2O analogy. It was found that both solubility and diffusivity increase as temperature increases. The CO_2 absorption rate of the solution was measured and the results were compared to pure K_2CO_3. The obtained results showed that the absorption rate increases with increasing temperature and concentration. In addition, the slower reaction kinetics of K_2CO_3 can be promoted by addition of DETA, indicating DETA acts as an effective promoter. The order of reaction, activation energy

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and reaction rate constant were obtained to be 1.61, 35.6 kJ/mol and $3.99 \times 10^{10} \exp(-\frac{4285.2}{T})$, respectively. A comparison between reaction rate constant determined in this study and other promoters indicated that reaction kinetics between CO_2 and K_2CO_3 + DETA solution was faster than other blended solutions.

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Table Captions:

Table 1: Density and viscosity of water at 303, 313 and 323 K.

Table 2: Kinetic data for the CO_2 absorption in solution of K_2CO_3+DETA.

Figure Captions:

Fig. 1: Schematic diagram of the experimental setup

Fig. 2: The density of K_2CO_3+DETA solution at different temperatures and concentrations

Fig. 3: The viscosity of K_2CO_3+DETA solution at different temperatures and concentrations

Fig. 4: The diffusivity of CO_2 in K_2CO_3+DETA solution

Fig. 5: The solubility of CO_2 in K_2CO_3+DETA solution

Fig. 6: The absorption rate of CO_2 in K_2CO_3+DETA solution as a function of speed of agitation.

Fig. 7: The absorption rate of CO_2 in K_2CO_3+DETA solution at 313 K.

Fig. 8: The absorption rate of CO_2 in K_2CO_3+DETA solution as a function of temperature.

Fig. 9: The effect of DETA concentration on overall reaction rate constant

Fig. 10: Plot of $log(k_{app})$ versus log of DETA concentration

Fig. 11: Arrhenius plot for the K_2CO_3+DETA+H_2O system

Fig. 12: The Hatta number and enhancement factor for K_2CO_3+DETA+H_2O system

Fig. 13: Parity plot for the predicted absorption rate and the experimental data.

Fig. 14: Comparison of reaction rate constant for different absorbents at 313.15 K.

Table 1

T (K)	ρ (g.cm^{-3})		μ (mPa.s^{-1})	
	Exp.	Lit. [24]	Exp.	Lit. [25]
303	0.9951	0.9957	0.7903	0.7975
313	0.9919	0.9922	0.6501	0.6532
323	0.9872	0.9880	0.5504	0.5471

standard uncertainties: u(T) = 0.01 K, u(ρ) = 0.0001 g.cm^{-3}, u(μ) = 0.0001 mPa.s^{-1}

Table 2

T (K)	[K$_2$CO$_3$] kmol/m^3	[DETA] kmol/m^3	N$_{CO_2}$ × 10^6 (kmol/m^2.s)	k$_{OV}$ (1/s)	K$_L$ × 10^5(m/s)	Ha	Ei
313	2	0.2	2.97	3899	2.76	104	782
313	2	0.3	3.89	7054	2.68	142	822
313	2	0.4	4.74	11061	2.63	179	872
313	2	0.5	5.81	17425	2.57	228	918
303	2	0.5	4.25	8942	1.96	183	860
323	2	0.5	6.77	33760	3.04	275	973

Fig. 1

Fig. 2

Fig. 3

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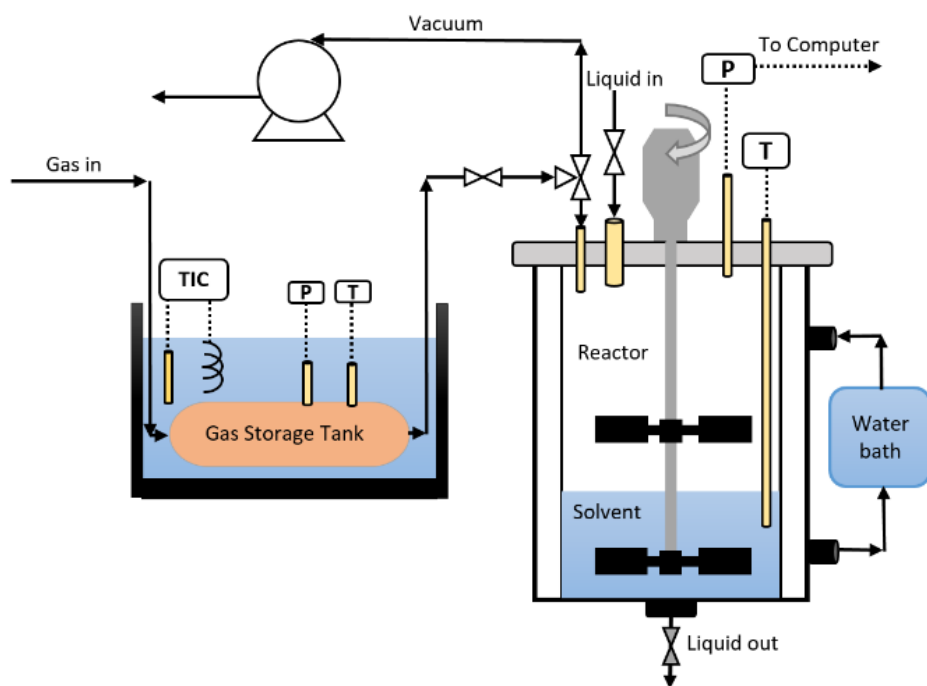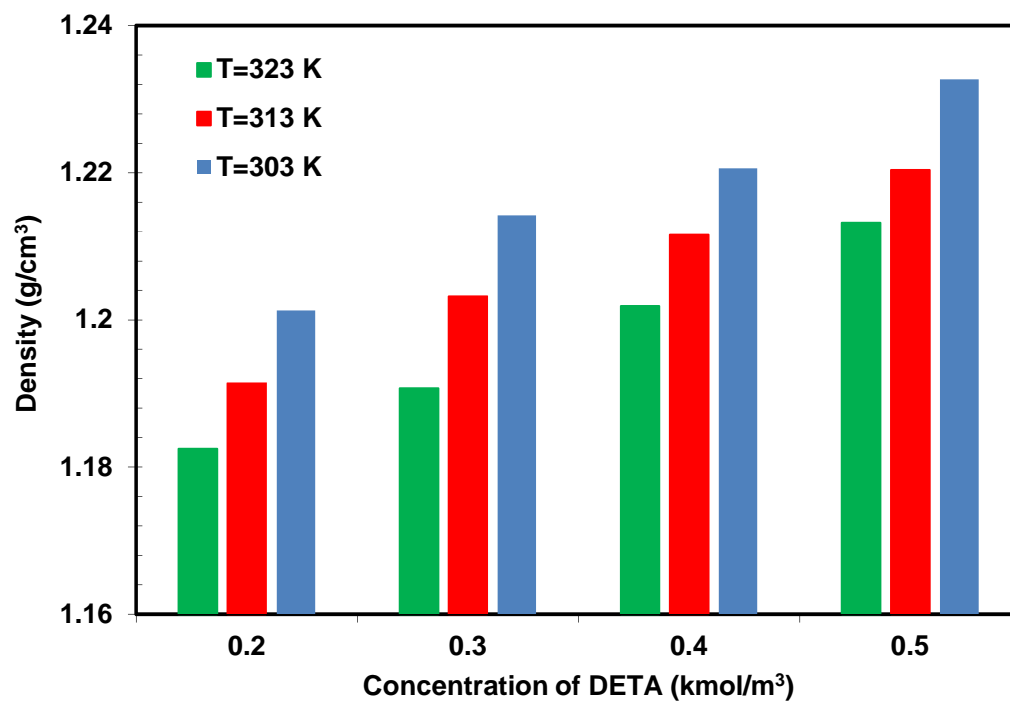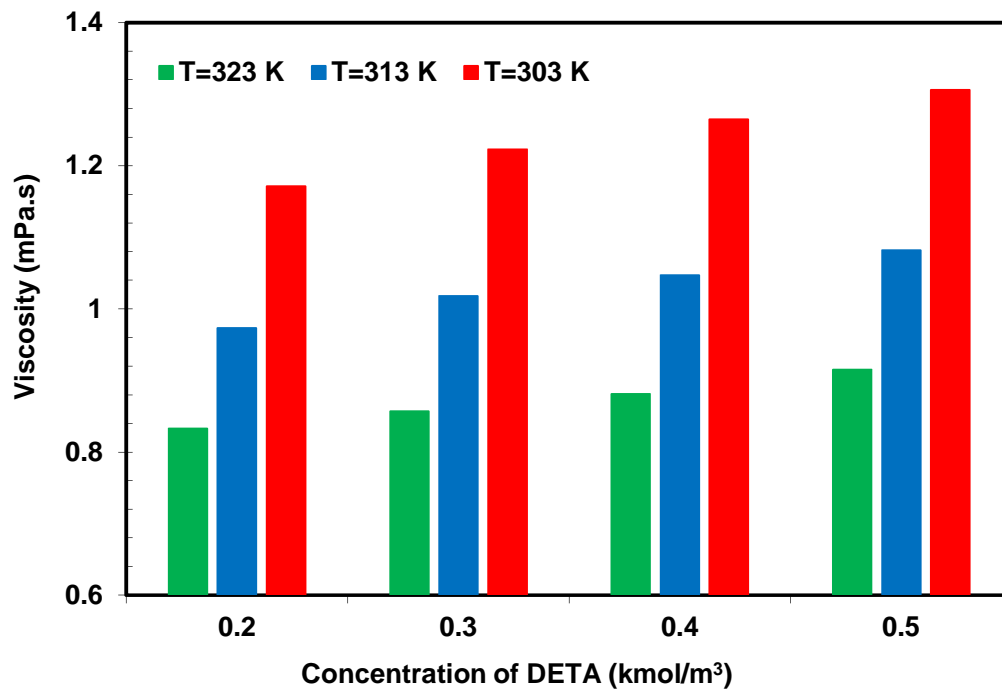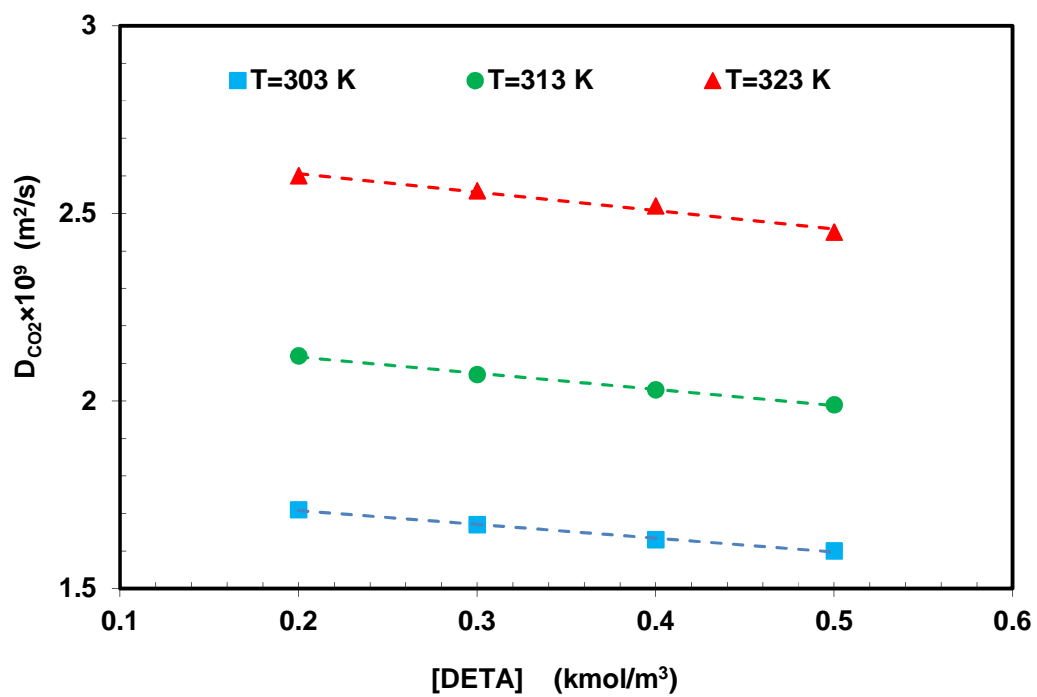
Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

$$y = 1.6124x + 4.7053$$

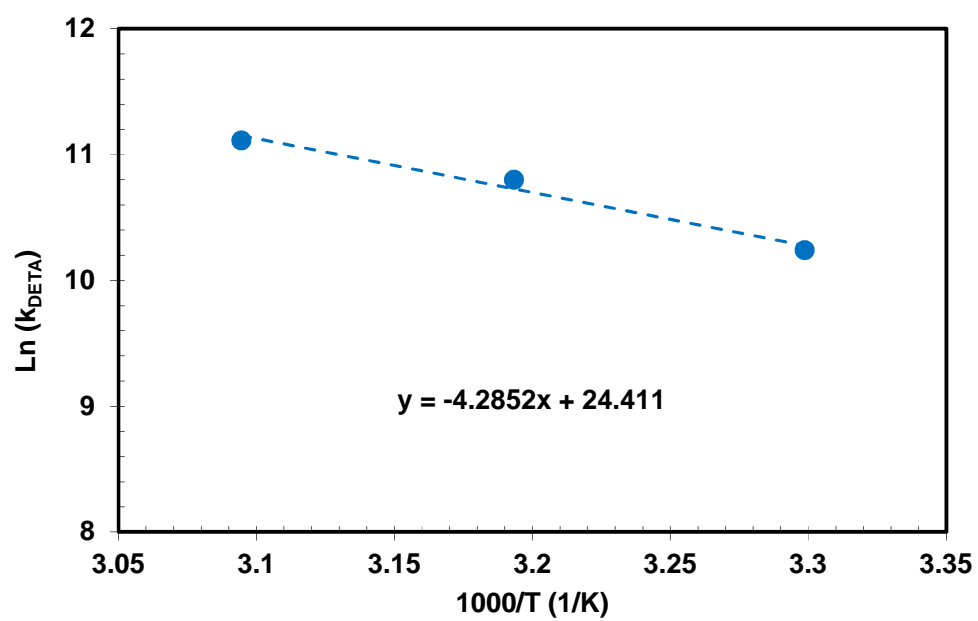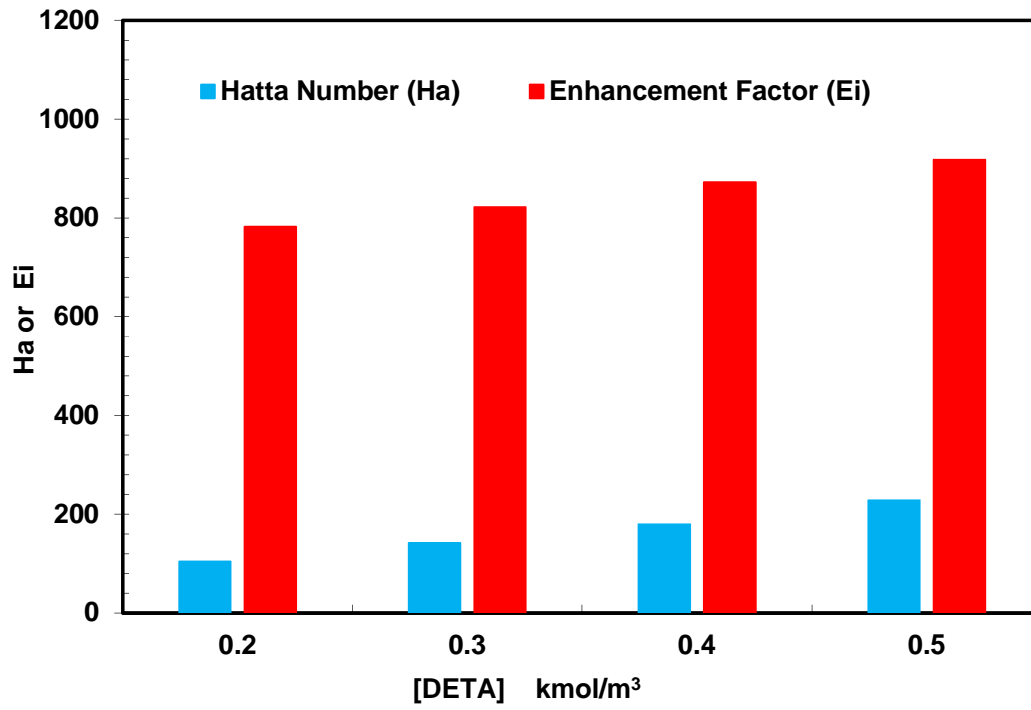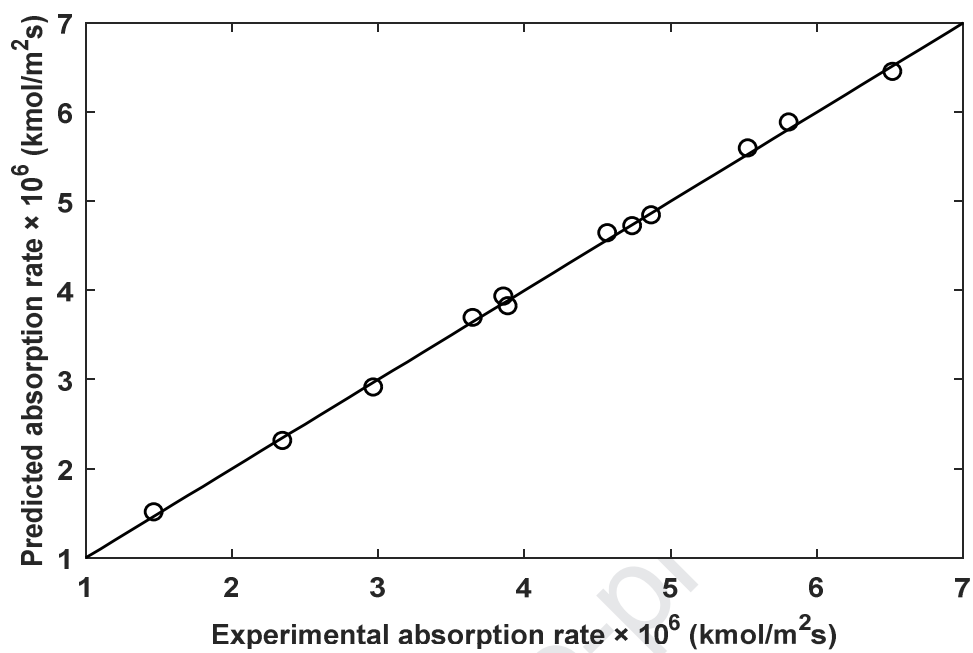**Fig. 10**

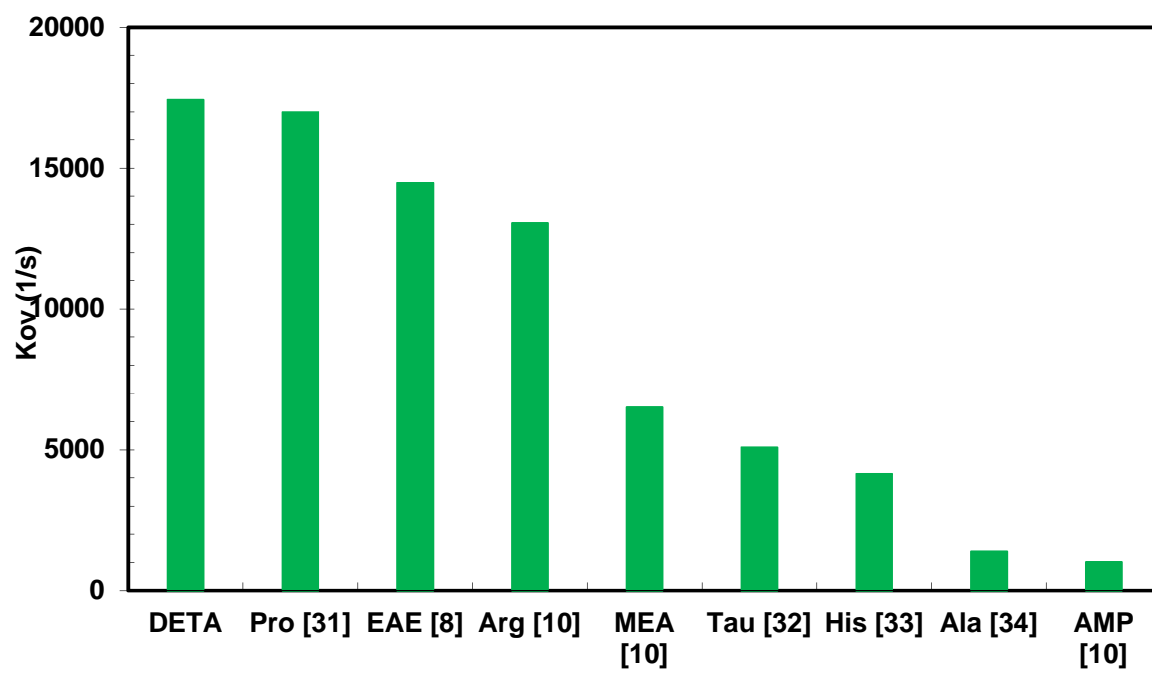$$y = -4.2852x + 24.411$$
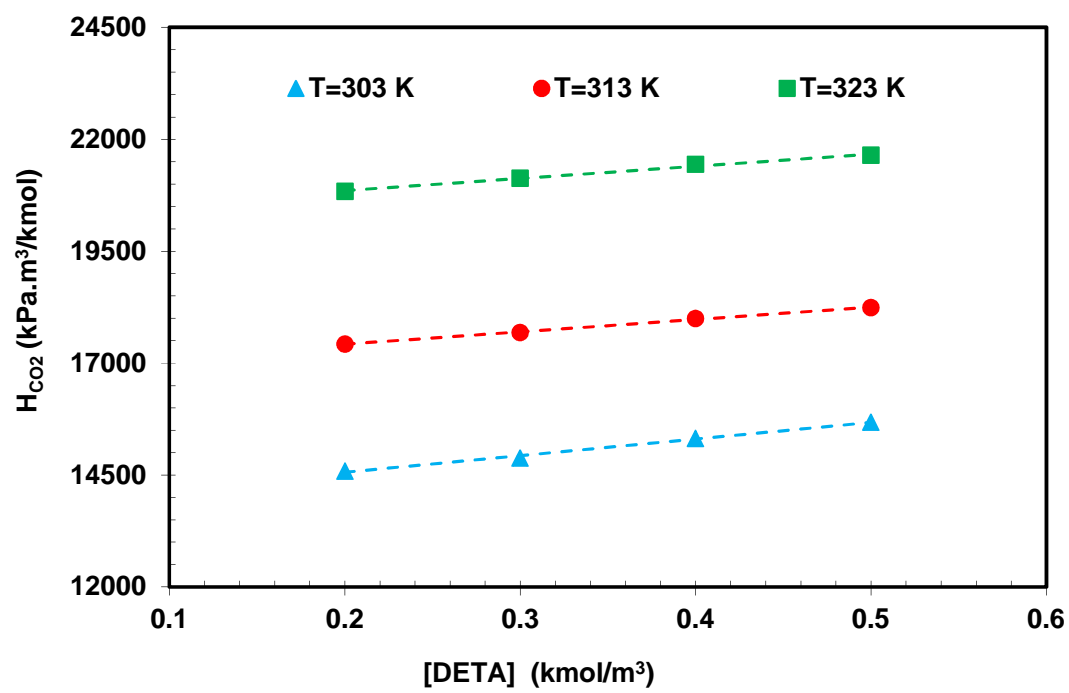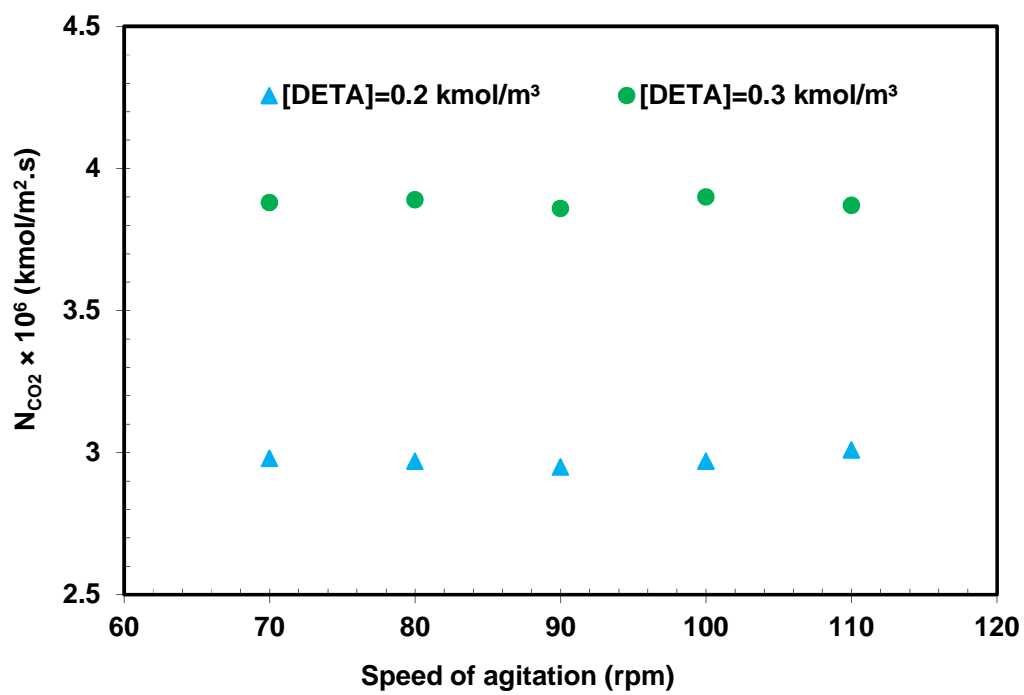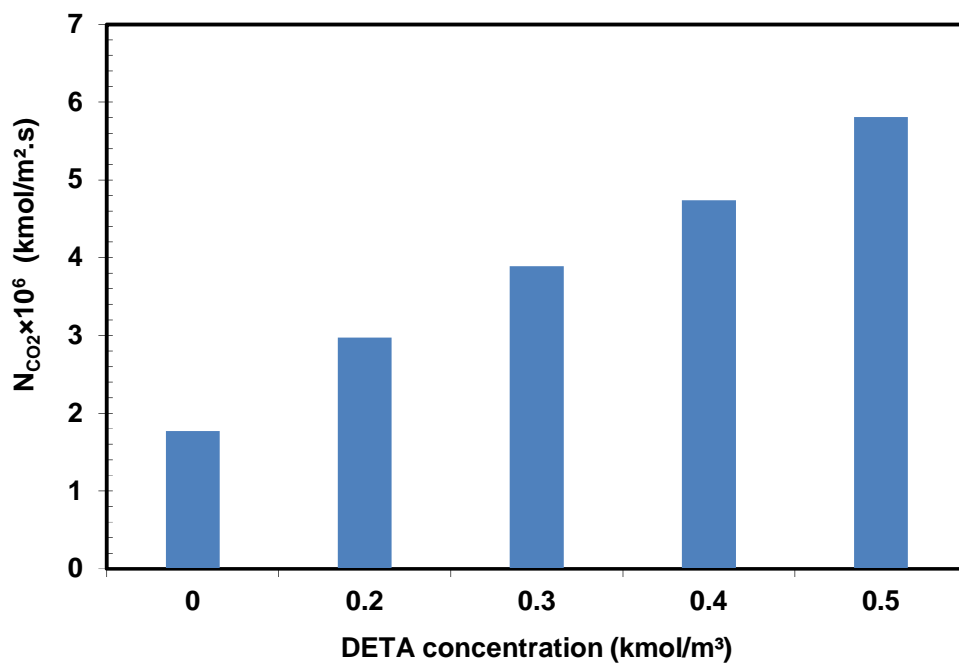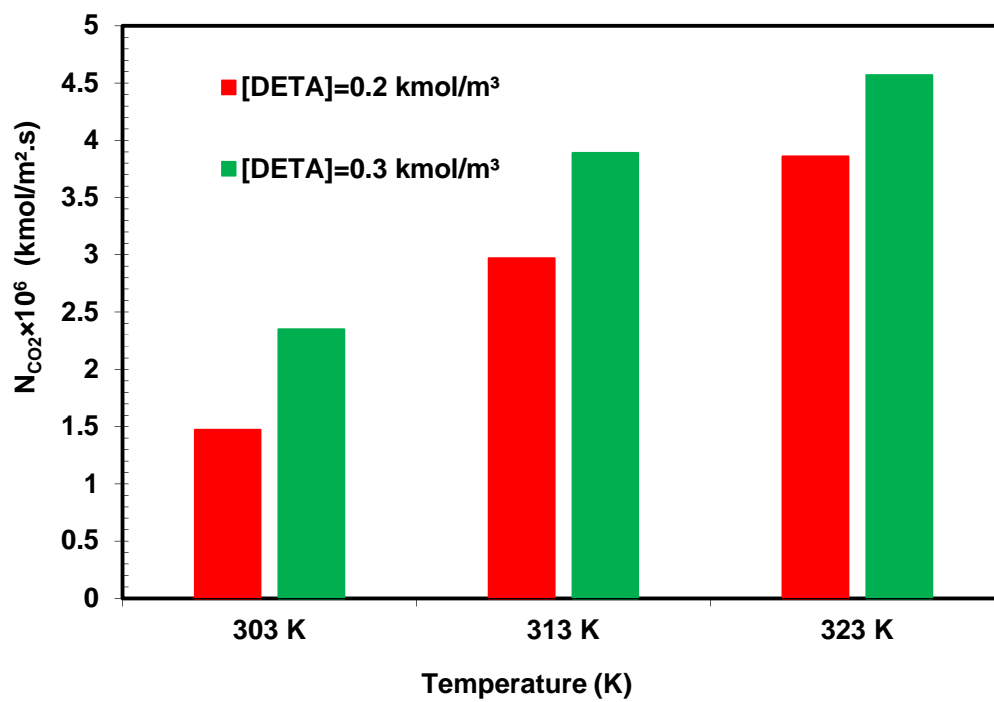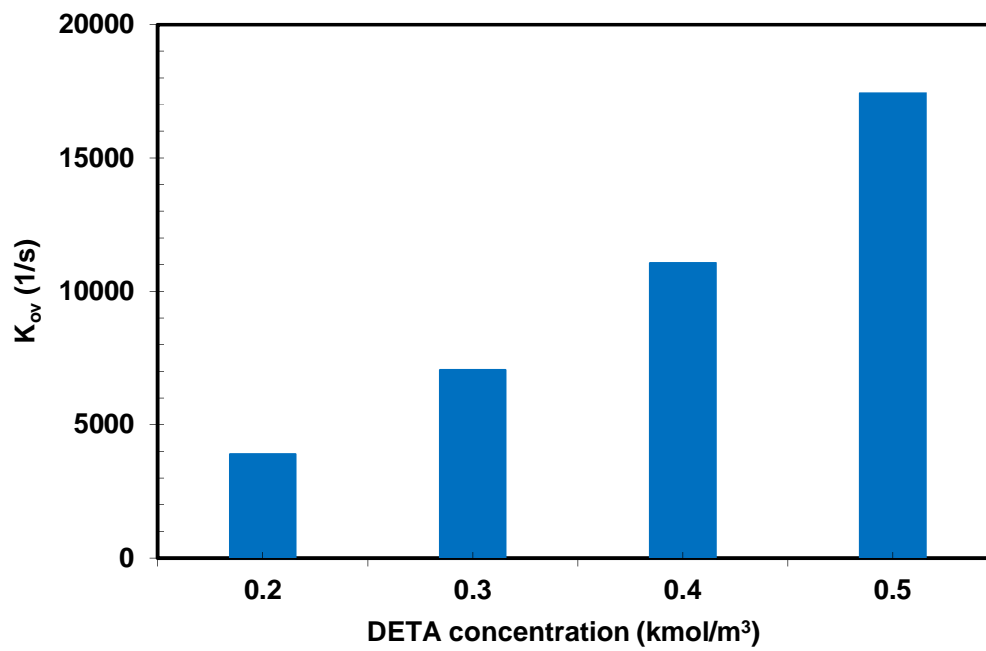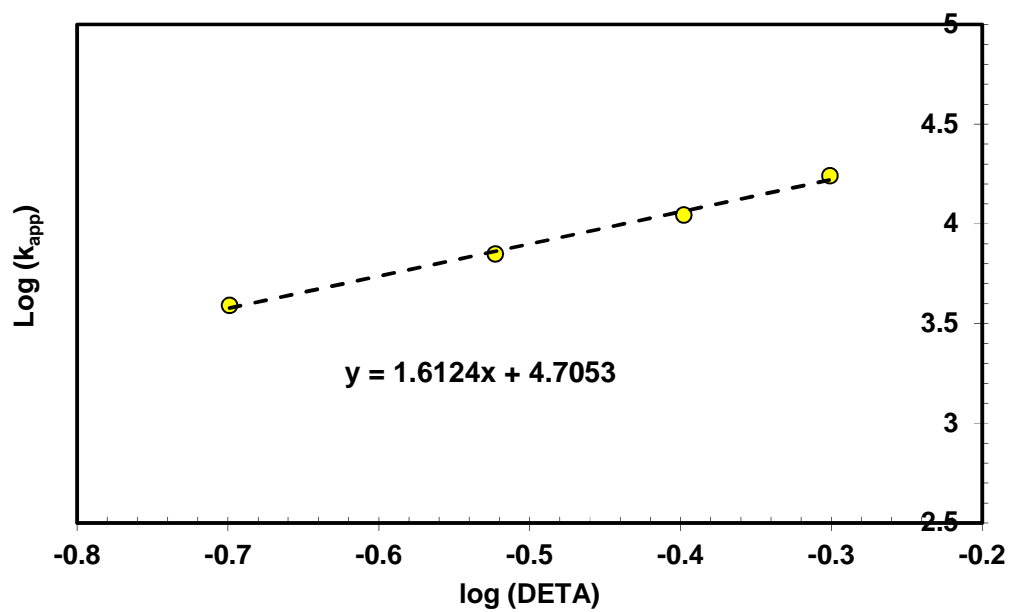
Fig. 11

Fig. 12

Fig. 13

Fig. 14

CONFLICT OF INTEREST

The author (editor) declares no conflict of interest, financial or otherwise.