INFLUENCE OF ETHYLENEDIAMINE CONTENT OVER PERFORMANCE OF CO₂ ABSORPTION INTO POTASSIUM CARBONATE SOLUTIONS

Maria Harja[1], Ramona Tataru-Farmus[1]*, Elisabeta Droniuc Hultuana[1], Consuelo Gómez de Castro[2], Gabriela Ciobanu[1], Liliana Lazar[1]

[1]*"Gheorghe Asachi" Technical University of Iaşi, Faculty Chemical Engineering and Environmental Protection,*
73 Prof. D. Mangeron Blvd., 700050, Iasi, Romania
[2]*Complutense University of Madrid, Faculty of Chemical, Department of Materials*
and Chemical Engineering, Av. Séneca, 2, 28040 Madrid, Spain

Abstract

In the last years, various experimental works investigated the efficiency of different type of promoters over CO₂ absorption. The absorption process is recommended for removal of acidic contaminants from flue gases and for obtaining new products. The absorption with chemical reaction (chemisorption) can be done in: potassium carbonate solution promoted with amines; aqueous solution of the amines; ammonia solution, hydroxide solution etc. Chemisorption in potassium carbonate solution with the addition of amines, as promoters, at 343-353 K has larger used due to the following advantages: lower cost, easy regeneration, nontoxic, noncorrosive, etc. In this paper the post-combustion CO₂ capture in potassium carbonate solution promoted by amines is presented. It is known that physico-chemical fluid property values, crucial in the modeling of the reactor, are necessary to be determined for particular systems. The aim of this study is to measure the densities and viscosities of potassium carbonate solution with the addition of ethylenediamine and to evaluate the performance of the promoter content and temperature over the enhancement factor into a bubbling reactor. Experimental results showed that the enhancement factor values between 1.8 and 3 could be achieved, demonstrating the possibility of using ethylenediamine in bubbling absorption reactor.

Key words: absorption, carbon dioxide, ethylenediamine, potassium carbonate, properties

1. Introduction

Among the atmospheric pollutants, carbon dioxide substantially contributes to climate change, respectively, to global warming (Mustafa et al., 2020). The problem of global warming caused by CO₂ emission has raised serious concerns since the CO₂ concentration in the atmosphere has increased rapidly due to the industrial revolution. The recovery of carbon dioxide (CO₂) from flue gas streams by absorption processes remains an attractive technology to reduce global warming due to human activities, especially the burning of fossil fuels. Fossil fuels will continue to be a major source of energy for the next few decades because the non-carbon energy sources are still limited and expensive.

However, although CO₂ capture is a global issue, there is no simple solution that can be applied on a global scale. Different technologies have been developed to reduce carbon dioxide emissions from flue gases. Depending on plant configurations, CO₂ emissions from flue gas can be reduced by different methods: pre-combustion capture; post-combustion capture or oxyfuel combustion (Carpenter and Long, 2017; Tataru-Farmus et al., 2019; Wang et al., 2017). From these methods, post-combustion capture is one

---

* Author to whom all correspondence should be addressed: e-mail: mharja@tuiasi.ro; rtataru@ch.tuiasi.ro

of the most studied options for reducing CO_2 emissions. Post combustion capture can be realized by absorption, adsorption, membrane separation, cryogenic, etc. (Guo et al., 2013; Hu et al., 2018; Younasa et al., 2020). The advantages and disadvantages of different methods are reported in the literature (Harja et al., 2018; Tataru-Farmus et al., 2019); a careful analysis of those demonstrated that absorption is the most recommended method for CO_2 capture from flue gases (Prentza et al., 2018; Wu et al., 2014). Absorption with chemical reaction (chemisorption) can be easy to implement on at the large scale in functional power plants. The absorption of CO_2 into chemical solvents is a well-developed technology applied to numerous commercial processes, including gas treatment and ammonia production. Recently, through the collaboration between researchers and large companies, chemical absorption installations have been designed with excellent results (CCST, 2021; ITC, 2021; Wang et al., 2011; https://sequestration.mit.edu/).

Many solvents have been reported as potential absorbents for CO_2 removal, such as amine solution, ammonia solution, potassium carbonate solution, potassium carbonate solution promoted (Koronaki et al., 2015; Prentza et al., 2018; Smith et al., 2015). The potassium carbonate solutions are recommended to be used for CO_2 retention because of their lower rate of degradation at high temperature and are cheap compared to amines; however, the carbonate solutions have limitations, especially slower rate of reaction (Cullinane and Rochelle, 2004). The absorption process using potassium carbonate solutions for CO_2 removal has been known for many years, being used in ammonia technology for synthesis gas purification (Akinola et al., 2019; Harja et al., 2019).

More efficient than potassium carbonate are amines solutions, such as primary amines monoethanolamine (MEA) and diglycolamine (DGA), the secondary amines diethanolamine (DEA) and diisopropanolamine (DIPA), and tertiary amines methyldiethanolamine (MDEA) and triethanolamine (TEA) (Choi et al., 2017; Hu et al., 2017; Lang et al., 2017). The tertiary amines react more slowly with CO_2 and are not often used in the absorption processes. Comparing the potassium carbonate solution with amine solutions, the last have significant limitations, including: high degradation rates, corrosion, salts unstable at high temperatures, the possibility of stable salts formation due to impurities from gases; high toxicity, solvent losses, etc. (Gupta et al., 2014; Koronaki et al., 2015; Rezazadeh et al., 2017; Sreenivasulu et al., 2015). The main limitation is the regeneration cost of solvent (about 70-80% of total operating cost of the system).

To improve the rate of chemical reaction, scientists have suggested the addition of promoters or activators to K_2CO_3 solutions. Table 1 lists the most used substances as promoters for the potassium carbonate solution. Among these, amines remain most efficient. By using a small quantity of amines in potassium carbonate solution, we obtain a synergistic effect, combining the advantages of the two categories of absorbents. Primary and secondary amines are mostly used as promoters for CO_2 absorption, while tertiary amines do not have a significant effect (Thee et al., 2012). Other amines, such as N-methyldiethanolamine (MDEA), 2- amino-2-methyl-1-propanol (AMP), piperazine (PZ), which is a cyclic diamine, diethanolamine (DEA) are also recommended as activators (Afkhamipour and Mofarahi, 2017; Hu et al., 2016).

The study of Cullinane and Rochelle (Cullinane and Rochelle, 2004) showed that when an activator is used, the potassium carbonate solution becomes an energy-efficient solvent and has several advantages over amino solvents, such as MEA. There are also experimental kinetic studies that have shown that absorption systems based on potassium carbonate promoted with piperazine (PZ) have a higher rate of absorption than amino solvents (Bishnoi and Rochelle, 2000).

For a better absorption process, it is essential to find more efficient solvents (Mangalapally et al., 2009; Quyn et al., 2013). The challenge is to search for the most appropriate promoter to overcome the drawback of amines.

Table 1. The promoters for the absorption into K_2CO_3 solution

Promoters	References
MEA –monoethanolamine	Lang et al. (2017); Grimekis et al. (2019); Thee et al. (2012)
DEA – diethanolamine	Hu et al. (2017); Pashaei et al. (2017)
DETA – diethylenetriamine	Ramezani and Di Felice (2019)
MDEA – methyl diethanolamine	Grimekis et al. (2019); Lang et al. (2017)
DGA–diglycolamine; DIPA – diisopropanol-amine;	Borhani et al. (2015a)
PZEA – 2-(1-piperazinyl)-ethylamine; TEPA – tetraethylenepentamine	Ramazani et al. (2016)
PZ - piperazine	Pashaei et al. (2020)
Aminoacids	Hu et al. (2018)
GL - Glycine	Guo et al. (2013); Smith et al. (2015)

For a high absorption rate, a concentrated aqueous solution of 20-40 % wt. potassium carbonate, as close to saturation, can be used. A higher concentration must be avoided due to the low bicarbonate ($KHCO_3$) solubility, which can cause precipitation and fouling problems. Recently, there are recommendation to use for carbon dioxide absorption a precipitating potassium carbonate solvent (Smith et al., 2015; Wu et al., 2017). Promoter concentrations for amines are usually in the 1–10 wt. % range (Borhani et al., 2015a; Hu et al., 2016; Grimekis et al., 2019).

The addition of promoters in potassium carbonate solutions provides additional characteristics, which would improve the CO_2 absorption performance. This work proposes a new solvent containing concentrated aqueous potassium carbonate and ethylenediamine (EDA, CAS 107-15-3) for CO_2 capture from waste gas streams. The present paper reports on the achieved progress in the solution properties for CO_2 absorption process. The solvent holds an advantage over other amines: because EDA is a diamine, the solvent can react with two moles of CO_2 per mole of amine. Coupled with the potassium carbonate solution, the solvent has the potential for a higher CO_2 capacity than other amines. Moreover, the two amine functional groups will favorably affect the rate of absorption.

To study the absorption process, the properties of the phases must be determined. Physical properties, such as density, viscosity of the potassium carbonate – bicarbonate solution without and with promoters, CO_2 diffusion coefficient, Henry constant for reactive absorption etc., are crucial for the practical development, design and implementation of a solvent. Knowledge of the density and viscosity are crucial to establish rate modeling and to determine the constants of the reaction rate (Hultuana et al., 2018). There are numerous types of reactors that can be used for chemisorption processes: bubbling jet reactor, combined packed and spray tower absorber, cable wet scrubber, packed column, etc. (Harja, 2019; Tataru-Farmus et al., 2017, Tataru-Farmus et al., 2019). In the last period, the stirrer bubble columns are applied in the potassium carbonate-amine CO_2 absorption processes (Pashaei et al., 2017; 2020). The bubble column is recommended for flue gas purification owing to the simple design, low investment costs, high heat and mass-transfer performance etc. Due to the low investment and operating costs this study uses a bubbling reactor, for scale-up the results.

The study aims to evaluate the properties and the performance of the new promoters' quantities, since there is no data in the literature. In order to design and implement an efficient and low-cost process a carbon dioxide capture absorbent was developed and laboratory studies were conducted to establish optimal operational parameters for obtaining high value for the enhancement factor, and data about the physico-chemical absorbent properties, necessary in absorption simulation.

2. Materials and method

2.1. Materials

Chemicals were used as received unless otherwise stated. Potassium carbonate 99 % and potassium bicarbonate 99.7 % were provided from Fluka Analytical. Ethylenediamine (EDA, CAS 107-15-3), with chemical formula $C_2H_8N_2$, was purchased from Sigma Aldrich with a certified GC purity greater than 99%. The distilled water was obtained from an apparatus delivering ultrapure water (Millipore). An electronic analytical balance (Kern ABS 220-4N) with the measurement accuracy of $\pm 1 \times 10^{-4}$ g was used to weight the chemicals. Each solution contains 25 wt.% of potassium carbonate, 4% wt. potassium bicarbonate and amine between 0 and 7% wt. The maximum concentration of the amine in the solutions was kept at 0.07, similar with the commercial formula of the solvent; the uncertainty in the mass fraction was determined to be $\pm 1 \times 10^{-3}$.
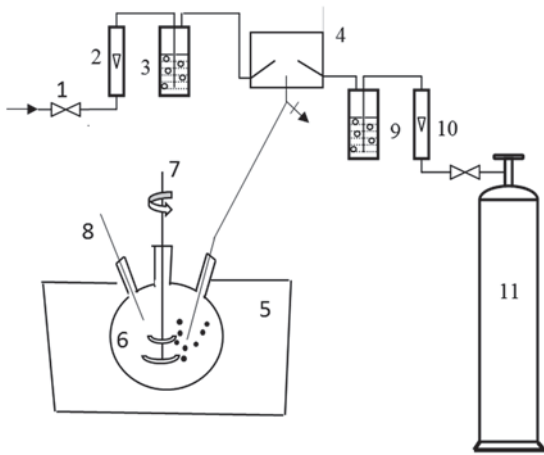
2.2. Measurements of physico-chemical properties

The densities of the potassium carbonate – bicarbonate solution, promoted with amines were determined at atmospheric pressure, temperature range between 298-348 K, using a DMA 4500 Anton Paar vibrating glass tube. The U-tube of densitometer was cleaned with acetone and distilled water and then air-dried with a built-in air pump to remove any moisture or contamination. The apparatus was calibrated before use and after the measurement, to obtain accurate results. Standard water of Millipore quality was used in the calibration process at atmospheric conditions, to minimize the measurement errors. The reported densities were measured after achieving thermal equilibrium, and the equipment was set to slow mode for better accuracy. Each experiment was performed in triplicate, and the data reported are the average values.

The viscosity was determined by Ostwald viscometer (Harja et al., 2020; Tataru-Farmus et al., 2007), which works on the basis of the Hagen-Poiseuille law. For each determination, the temperature was accurately controlled using the Viscosimeter Thermostat 655. The viscometer was cleaned with water and acetone and further dried with air to ensure reliable measurements. Water was used as calibrating liquid. A standard chronometer was employed for the time flowing measuring (12 times for each probe) with an accuracy of 0.01 s. The maximum error of viscosity measures was estimated at $\pm 1, 00$ %. The details of the methods and techniques used to determine the viscosity have been previously described (Tataru-Farmus et al., 2007). The viscosity was measured in the temperature range of 298 - 348 K. A SP-870 plus METERTECH UV–Vis spectrophotometer was used to monitor the K_2CO_3-$KHCO_3$-EDA systems absorbance (optical path =1cm).

2.3. Absorption procedure

The absorption efficiency was determined using the experimental set up presented in Fig. 1. The reactor used was a bubbling type, with heating system, having a volume of 1 L. The gas phase was a CO_2 steam, obtained by mixing CO_2 from the tank with air, having known flow and concentration (a CO_2 analyzer was used). The gas phase was bubbled in reactor, using distributor with one pipeline, at constant temperature. The mixed gas flow rate was 2.1 L/min. The gas temperature was 293 K. The unabsorbed CO_2 was continuously measured using a gas analyzer.

Fig. 1. Experimental plant
1– ventil air, 2, 10– rotameters, 3,9 – saturated vessel, 4 – mixture vessel, 5 – thermostat vessel; 6 –reactor, 7– stirrer, 8– CO_2 analyzer and thermometer; 11– CO_2 gas tank

The CO_2 concentration in the gas phase at the inlet and outlet of the reactor was measured using AWM5000 Series Microbridge Mass Airflow Sensor. The compositions of both phases (gas and liquid) were analyzed in order to evaluate the absorption performance.

The liquid temperature was constant, determined by thermometers placed inside of the reactor. At the preset time periods, samples of 5 mL were taken and analyzed for determining the liquid concentration. For the liquid phase, the titration method was used to establish the concentration of the species: CO_3^{2-} and HCO_3^{-}. The samples were treated with HCl and two indicators: phenolphthalein and methyl red. A two-step reaction occurs, consuming two moles of HCl to neutralize one mole of carbonate (V_1), and a volume V_2 of HCl is used to neutralize all the bicarbonates, the titration finishes when the color becomes totally pink (Salmón et al., 2018).

In the industrial process, the liquid phase has a complex composition. It also contains $KHCO_3$ because a semi-regenerated solution is introduced in the absorption column. By completely regenerating only a part of the carbonated solution, significant energy savings are made. Increasing the temperature and concentration of the solutions increases the absorption rate. Also, the solubility of carbonates increases with temperature. The temperature and concentration of the solution are chosen based on the liquid-solid equilibrium data in the ternary system K_2CO_3 - $KHCO_3$ - H_2O, so that during the absorption process no solid phase appears. There is little data on the properties of ternary solutions in the conditions that occur in industrial processes. According to previous studies in the literature, the composition of the liquid phase is 25% K_2CO_3, 4% $KHCO_3$ and 1-7% EDA. The solution temperature was maintained at constant temperature by electrical heating. All tests were performed at atmospheric pressure.

The absorbent efficiently was established by enhancement factor (Hagiu and Ivaniciuc, 1997; Harja et al., 2018; Petrescu et al., 1998), considering the two films theory. The enhancement factor (E) may be defined by Eq. (1), as the ratio between the flux transferred in presence of chemical reaction ($N`_A$) and the flux transferred for physical absorption (N_A):

$$E = \frac{N`_{CO_2}}{N_{CO_2}} = \frac{Ha}{tanh(Ha)} \left[1 - \frac{\gamma}{cosh(Ha)} \right] \frac{1}{1-\gamma} \quad (1)$$

where expression for γ is presented in Eq. (2):

$$\gamma = \frac{C_{CO_2}^v}{C_{CO_2}^i} \quad (2)$$

and for $C_{CO_2}^v \ll C_{CO_2}^i$, $y \to 0$ and Eq. (1) becomes Eq. (3) (Harja et al., 2019):

$$E = \frac{Ha}{\tanh(Ha)} \quad (3)$$

Hatta modulus (Ha) was calculated with Eq. (4):

$$Ha = \frac{\sqrt{k_1 D_{CO_2}}}{k_L^0} \quad (4)$$

where: k^0_L, mass transfer coefficient of CO_2 from interface to liquid bulk (m/s); k_1, rate coefficient of the reaction (1/s); D_{CO_2} - molecular diffusivities of CO_2 (m²/s);

The experiments were conducted in potassium carbonate/bicarbonate solution, potassium carbonate/bicarbonate solutions promoted with amines and in water, for establishing the transferred flux without chemical reaction. Experimentally, we determined concentration gradient in time, by monitoring the liquid concentration. Initially, were realized experiments for establishing reaction time (time for which is realized maximum absorption). Experimental results demonstrated that after 45 min the equilibrium was reached, therefore the time considered for enhancement factor was 60 min.

Because all conditions were maintained unmodified, the enhancement factor may be calculated by Eq. (5):

$$E = \frac{C_{K_2CO_3}^0 - C_{K_2CO_3}}{C_{CO_2}^*} \quad (5)$$

where: $C_{CO_2}^*$ is the CO_2 concentration in the water, calculated from the Henry law (or experimental determination by titration with $Ba(OH)_2$, mol/L; $C_{K_2CO_3}^0, C_{K_2CO_3}$ the carbonate concentration at the initial and final time, mol/L.

3. Results and discussion

3.1. Density

Experimentally measured data for density were converted into graphical form with respect to EDA content, in the temperature range 298 - 348 K and are presented in Fig. 2. This density trend is similar to previous papers (Valtz et al., 2006; Tataru- Farmus et al., 2006; Amann, 2007; Harja et al., 2008). The density of the K_2CO_3-$KHCO_3$-water system decreases when a small amount of EDA is added. Organic molecules, especially their hydrocarbon parts, are surrounded by water molecules, arranged structurally to form cages. In the presence of EDA, water has a new structure until, quantitatively, EDA becomes sufficient to disorganize the system, destroy cages and form complexes.

At a constant temperature (298 K) and the lowest mass fraction for the promoter (1% EDA), the density value of the aqueous solution was 1.274 g cm⁻³. In addition, the density data showed a decreasing trend with temperature increasing. This may be due to large spaces between molecules at higher temperatures, which leads to the expansion of the volume of the solution and thus to the reduced density.

Equation (6) was used to correlate the density experimental data:

$$\rho = aT + b \qquad (6)$$

where: ρ is the density of K_2CO_3 + $KHCO_3$ + EDA solution, and T is the temperature, K. The optimized parameters of the correlation equation are reported in Table 2, with the standard deviation calculated using (Eq. 7):

$$SD = \left[\frac{\sum \left(\rho_{exp} - \rho_{calc} \right)^2}{n} \right]^{0.5}$$

$$(7)$$

3.2. Viscosity

The experimental results for K_2CO_3- $KHCO_3$-EDA systems are presented in the Fig. 3 for 25% K_2CO_3, 4% $KHCO_3$ and different mass concentrations of EDA (1 to 7% wt.).

The viscosity continuously increases with concentration and decreases with increasing temperature, for each system, as expected. This is due to the decrease in the internal resistance of the molecules with increasing temperature, which allows the molecules of the solution to flow easily, thus decreasing the viscosity.

However, increasing the concentration of amine in aqueous solutions increases the viscosity of the mixture. Higher concentration solutions have higher viscosity than lower ones, due to the fact that molecular strength increases in higher concentration solutions.
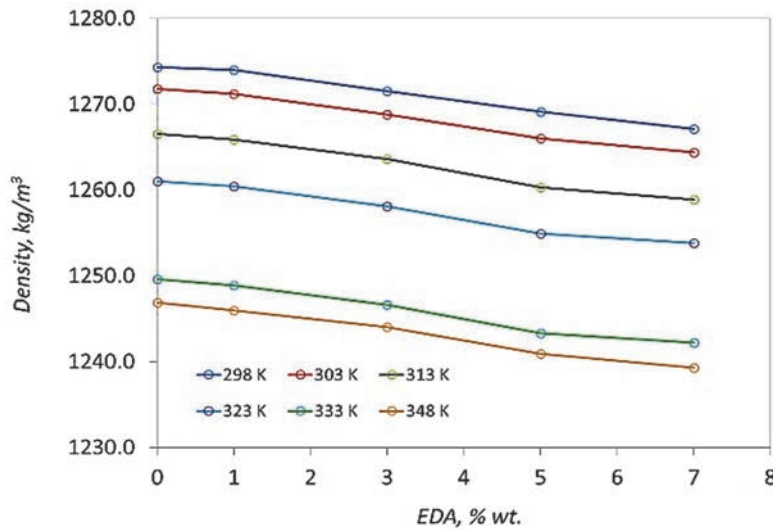

Fig. 2. Plot of density of K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA (0, 1, 3, 5, 7 %) solution vs. EDA concentration, at constant temperature

Table 2. Correlation (Eq. 6) parameters and SD for the density of aqueous blends of K_2CO_3+ $KHCO_3$ – EDA

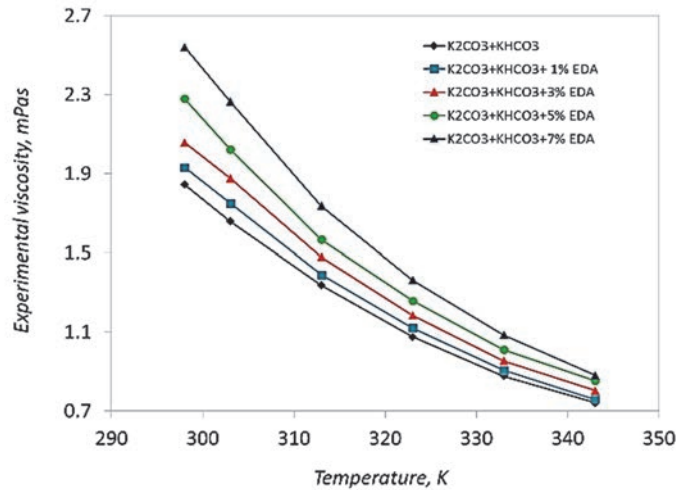| w_{K2CO3}+ w_{KHCO3} +w_{EDA},	$\rho = a\,T + b$			
	a	*b*	R^2	*SD*
0.25 + 0.04 + 0	-0.5511	1438.8	0.9997	0.1876
0.25 + 0.04 + 0.01	-0.5582	1440.5	0.9996	0.2094
0.25 + 0.04 + 0.03	-0.5527	1436.4	0.9997	0.1821
0.25 + 0.04 + 0.05	-0.5649	1437.3	0.9998	0.1712
0.25 + 0.04 + 0.07	-0.5551	1432.7	0.9996	0.2111

Fig. 3. Plot of experimental viscosity of K_2CO_3+ $KHCO_3$ – EDA function of temperature

As seen in Fig. 3 there is a non-linear decrease in viscosity with increasing temperature. At a constant temperature of 298 K and at the lowest mass fraction of 1 % EDA, the value of viscosity of aqueous solution was 1.931 mPa s. The correlation of the viscosity values of potassium carbonate – potassium bicarbonate with activator solutions is difficult because the properties of the liquid vary greatly in composition and temperature. Equation (8) was used to correlate the viscosity data:

$$\eta = a + bw + cw^2 + dw^3 \tag{8}$$

where: η is the viscosities of K_2CO_3 + $KHCO_3$ + EDA solution, and w is the mass fraction of EDA in the mixture.

The optimized parameters of the correlation equation are reported in Table 3, with the standard deviation calculated using an equation similar with (Eq. 7). After the analysis of the predicted density and viscosity data obtained from the correlation equations,

it was observed that the predicted data agreed well with the experimental data (Figs. 4-5).

Table 3. Fitting parameters of Eq. 8 and SD for viscosity of aqueous blends of K_2CO_3+ $KHCO_3$ – EDA

T, K	$\eta = a + b\,w + c\,w^2 + d\,w^3$					
	a	*b*	*c*	*d*	R^2	*SD*
298	504.92	13.39	6.47	1.85	0.9985	9.83E-03
303	1848.10	-151.05	10.17	1.66	0.9999	1.79E-03
313	1352.40	-108.98	6.70	1.34	0.9997	2.36E-03
323	624.14	-51.05	4.60	1.08	0.9998	1.34E-03
333	298.48	-20.31	2.93	0.88	0.9998	9.65E-04
343	-441.53	40.99	1.28	0.74	0.9999	6.35E-04

3.3. Absorbance

The UV-Vis spectra were recorded for K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA system, for different EDA concentrations (0, 1, 3, 5, 7 %). All the spectra are similar, exhibiting a maximum absorption in the UV-A range (320-370 nm) (Fig. 6).
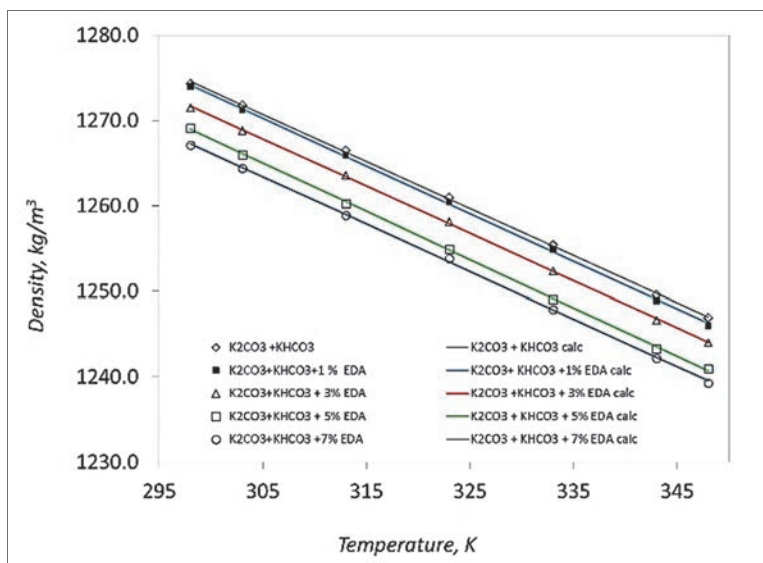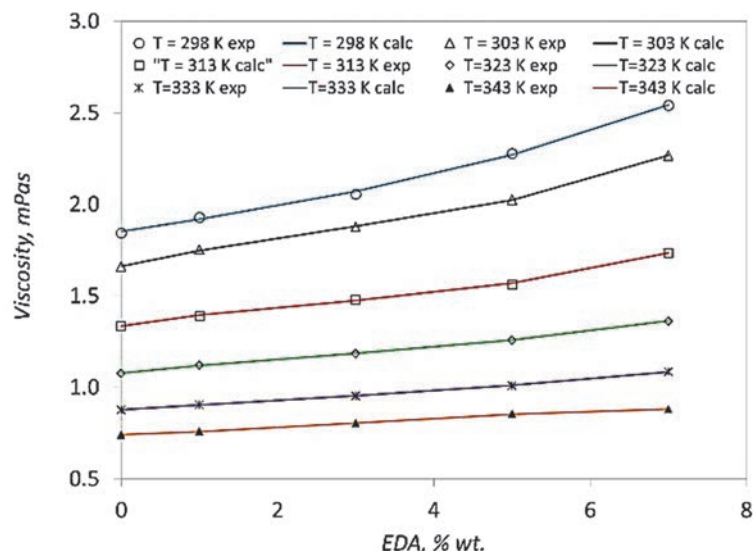

Fig. 4. Comparison between experimental values versus predicted values of density
for aqueous blends of K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA (0, 1, 3, 5, 7 %), at different temperatures

Fig. 5. Comparison between experimental values versus predicted values of viscosity
for aqueous blends of K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA (0, 1, 3, 5, 7 %), at different temperatures
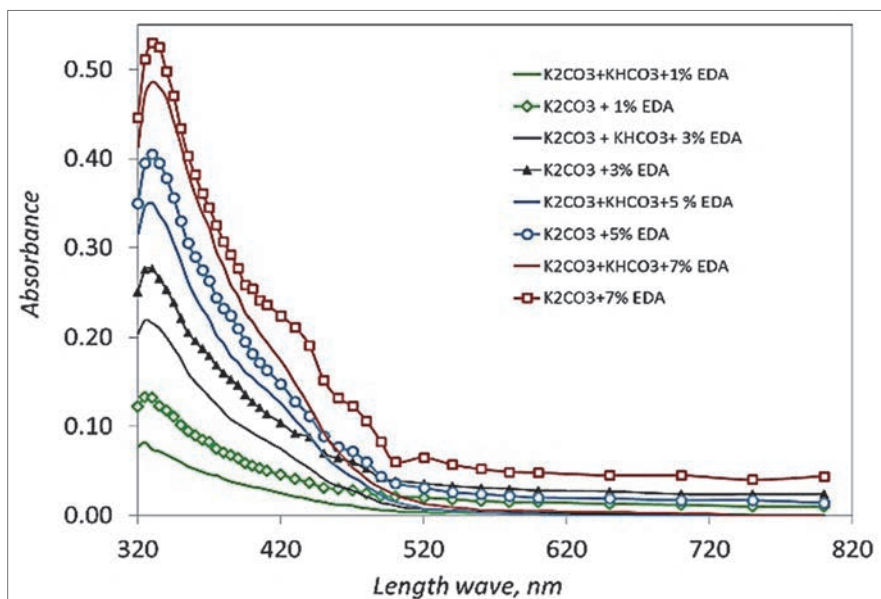
Spectral absorption increases with increasing amine concentration, for all blends. The same effect was reported in a study conducted for the characterization of solvents based on potassium carbonate - EDA (Bouteau et al., 2019; Harja et al., 2020; Ünveren et al., 2017). Compared to the potassium carbonate and potassium carbonate – EDA spectra, the addition of potassium bicarbonate in aqueous solution generates a hypochromic effect (Fig. 7).

3.4. Enhancement factor

The effect of the EDA percentage in the mixture on the enhancement factor is presented in Fig.

8, where from it can be observed that increases in the temperature improve the enhancement factor values. Alternatively, by increasing EDA content in the blend the enhancement factor increases; however, over 5% EDA, the influence is insignificant, especially for low temperatures. Thus, our studies recommended as promoters' content to be 5 %, in accord with the literature (Borhani et al., 2015b; Hu et al., 2016; Liu et al., 2020).

Experimental data demonstrated that EDA could replace DEA in industrial chemisorption processes, being more efficient at lower content. A chemical solvent with a high absorption capacity and high speed (enhancement factor E) considerably reduces the cost of the absorption process.

Fig. 6. The UV-Vis spectra for K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA (0, 1, 3, 5, 7 %) systems
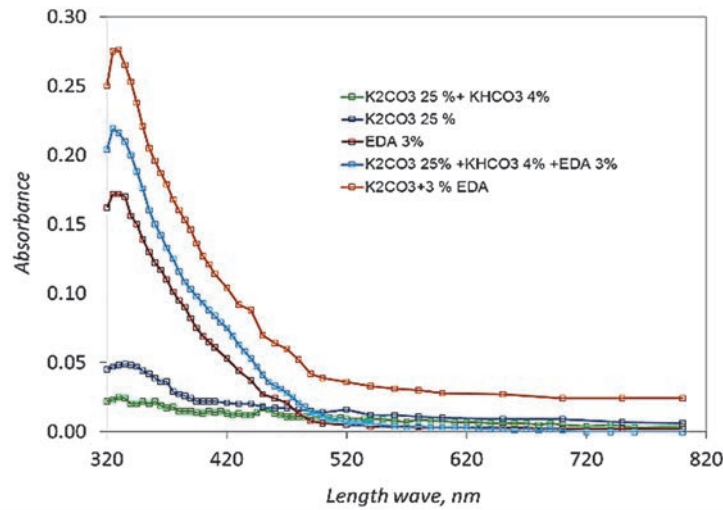
Fig. 7. The UV-Vis spectra for K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA 3 % system
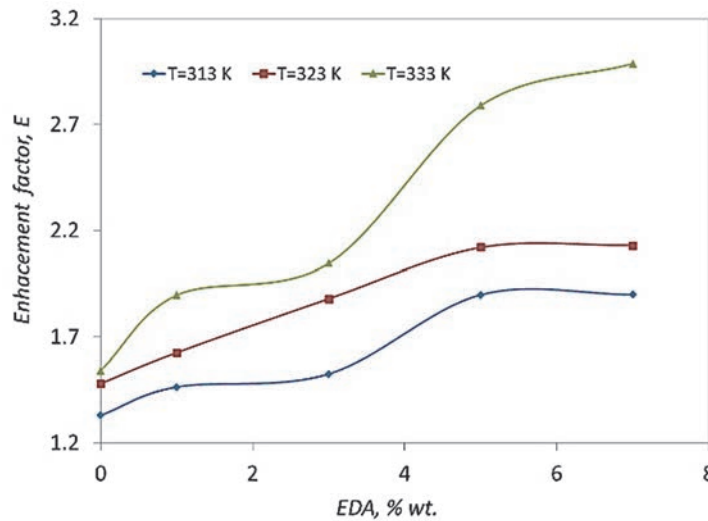

Fig. 8. The Enhancement factor K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA (0, 1, 3, 5, 7 %) systems

4. Conclusions

Absorption is a recognized technology for post combustion CO_2 capture, but the downfalls of the process, such as toxicity, solvent losses, corrosion, etc., imposed more studies for establishing a better solvent. All existing technologies have advantages and limitations, but their reliability, stability and removal efficiency are mainly experimental. There is a great need for understanding the existing technology to improve performance, and increase the capability for CO_2 separation. The aqueous potassium carbonate is a good solvent for carbon dioxide capture because of its low regeneration energy, low degradation and reduced corrosivity; the major challenge of the potassium carbonate absorption system is its slow reaction rate with CO_2 in comparison to amines absorption systems, especially under post-combustion conditions. Researchers have focused on studying different rate promoters to improve the kinetics of the absorption process and therefore reduce the size and cost of the absorption process.

To the best of our knowledge, no data for the physical properties of aqueous solution of potassium carbonate – bicarbonate – ethylendiamine are available in the literature. Therefore, in this study, the physical properties, i.e., density and viscosity of aqueous K_2CO_3 – $KHCO_3$ - EDA solutions were determined, studied and reported. Additionally, all physical properties were correlated using empirical correlations, such as a function of temperature and concentration. All properties tended to decrease with increasing temperature. The same trend was reported in the available literature.

After the analysis of the predicted density and viscosity data obtained from the correlation equations, it was observed that the correlated densities and viscosities are in excellent agreement with the experimental data over the temperature and relative composition ranges studied. Equation (6) proposed in this paper for calculating the density of aqueous blends is valid in the temperature range of 298– 348 K and for the composition K_2CO_3 (25%) + $KHCO_3$ (4 %) + EDA (0, 1, 3, 5, 7 %). The maximum standard

deviation is 0.21. Eq. (8) can be used to predict the viscosity of K_2CO_3 - $KHCO_3$ - EDA solutions (standard deviation of 9.83E-03) at temperatures in the 298-343K range.

The achieved enhancement factor demonstrates that EDA can be used as a promising promoter for carbon dioxide retention, the optimum concentration is 5 %. However, to make better judgment about this solvent, more investigation needs to be performed to determine the corrosion rate and absorption heat of $K_2CO_3+KHCO_3$- EDA solution.

Acknowledgements

References

Afkhamipour M., Mofarahi M., (2017), Review on the mass transfer performance of CO_2 absorption by amine-based solvents in low-and high-pressure absorption packed columns, *RSC Advances*, **7**, 17857-17872.

Akinola T.E., Oko E., Wang M., (2019), Study of CO_2 removal in natural gas process using mixture of ionic liquid and MEA through process simulation, *Fuel*, **236**, 135-146.

Amann J.M., (2007), *Study of CO_2 capture processes in thermal power plants* (in French), PhD Thesis,, Chemistry National School of Mines, Paris.

Bishnoi S., Rochelle G.T., (2000), Absorption of carbon dioxide into aqueous piperazine: reaction kinetics, mass transfer and solubility, *Chemical Engineering Science*, **55**, 5531-5543.

Borhani T.N.G., Akbari V., Hamid M.K.A., Manan Z.A., (2015b), Rate-based simulation and comparison of various promoters for CO_2 capture in industrial DEA-promoted potassium carbonate absorption unit, *Jounral of Industrial Engineering. Chemistry*, **22,** 306-316.

Borhani T.N.G., Azarpour A., Akbari V., Alwi, S.R.W., Manan Z.A., (2015a), CO_2 capture with potassium carbonate solutions: a state-of-the-art review, *International Journal of Greenhouse Gas Control*, **41**, 142–162.

Bouteau G., Van-Nhien A.N., Sliwa M., Sergent N., Lepretre J.C., Gachot G., Sagaidak I., Sauvage F., (2019), Effect of standard light illumination on electrolyte's stability of lithium-ion batteries based on ethylene and di-methyl carbonates, *Scientific Reports*, **9**, 1-9.

Carpenter S.M., Long H.A., (2017), Integration of carbon capture in IGCC systems, *Integrated Gasification Combined Cycle (IGCC) Technologies*, **2017**, 445-463

CCST, (2021), Carbon Capture & Sequestrations Technologies, On line at: https://sequestration.mit.edu.

Choi J.H., Kim Y.E., Nam S.C., Park S.Y., Chun I.S., Yoon Y.I., Lee J.H., (2017), Promoter characteristic study on the K_2CO_3 absorbents for CO_2 capture: mass transfer according to functional group and chain length of promoter, *Energy Procedia*, **114**, 898-905.

Cullinane J.T., Rochelle G.T., (2004), Carbon dioxide absorption with aqueous potassium carbonate promoted by piperazine, *Chemical Engineering Science*, **59**, 3619-3630.

Grimekis D., Giannoulidis S., Manou K., Panopoulos K.D., Karellas S., (2019), Experimental investigation of CO_2 solubility and its absorption rate into promoted aqueous potassium carbonate solutions at elevated temperatures,

International Journal of Greenhouse Gas Control, **81**, 83-92.

Guo D., Thee H., Tan C.Y., Chen J., Fei, W., Kentish S., Stevens G., da Silva G., (2013), Amino acids as carbon capture solvents: chemical kinetics and mechanism of the glycine + CO_2 reaction, *Energy Fuels*, **277**, 3898-3904.

Gupta A.K., Verma Y.L., Singh R.K., Chandra S., (2014), Studies on an ionic liquid confined in silica nanopores: change in T_g and evidence of organic-inorganic linkage at the pore wall surface, *Journal of Physical. Chemistry C*, **118**, 1530-1539.

Hagiu C., Ivaniciuc M., (1997), Determination of the enhancement factor for CO_2 absorption in activated potassium carbonate solution, *Revista de Chimie*, **48**, 856-862.

Harja M., (2019), *Modeling and Simulation of Chemical Reactors. Gas-Liquid reactors*, Ecozone Publisher, Iasi, 2019.

Harja M., Ciobanu G., Juzsakova T., Cretescu I., (2019), New approaches in modeling and simulation of CO_2 absorption reactor by activated potassium carbonate solution, *Processes*, **7**, 78-97.

Harja M., Ciobanu G., Rusu L., Lazar L., (2018), The enhancement factor approach for chemisorption process, *Environmental Engineering and Management Journal*, **17**, 813-820.

Harja M., Hultuană E., Apostolescu N., Apostolescu G.A., Tătaru – Fărmuş R.-E., (2020), Viscosity of potassium carbonate solutions promoted by new amines. *Journal of Engineering Sciences and Innovation*, **5**, 239-246.

Harja M., Rusu L., Barbuta M., (2008), The viscosity and density of the aqueous solutions used for the absorption accompanied by chemical reaction, *The Bulletin of the Polytechnic Institute of Iaşi, Chemistry and Chemical Engineering Section*, **9**, 55-62.

Hu G., Nicholas N.J., Smith K.H., Mumford K.A., Kentish S.E., Stevens G.W., (2016), Carbon dioxide absorption into promoted potassium carbonate solutions: A review, *International Journal of Greenhouse Gas Control*, **53**, 28-40.

Hu G., Smith K., Wu Y., Kentish S., Stevens G., (2017), Recent progress on the performance of different rate promoters in potassium carbonate solvents for CO_2 capture, *Energy Procedia*, **114**, 2279-2286.

Hu G., Smith K.H., Wu Y., Mumford K.A., Kentish S.E., Stevens G., (2018), Carbon dioxide capture by solvent absorption using amino acids: A review, *Chinese Journal of Chemical Engineering*, **26**, 2229-2237.

Hultuana E., Ciobanu G., Cretescu I., Lazar L., Tataru-Farmus R.E., Harja M., (2018), Modeling of CO_2 absorption process in activated solutions, European exhibition of creativity and innovation - EUROINVENT, Iasi, Romania, 53-54.

ITC, (2021), International test centre for CO_2 capture (ITC), On line at: http://co2-research.ca.

Koronaki I.P., Prentza L., Papaefthimiou V., (2015), Modeling of CO_2 capture via chemical absorption processes - an extensive literature review, *Renewable and Sustainable Energy Reviews*, **50**, 547-566.

Lang P., Denes F., Hegely L., (2017), Comparison of different amine solvents for the absorption of CO_2, *Chemical Engineering Transactions*, **61**, 1105-1110.

Liu S., Ling, H., Gao H., Tontiwachwuthikul P., Liang Z., Zhang H., (2020), Kinetics and new Brønsted correlations study of CO_2 absorption into primary and secondary alkanolamine with and without steric-hindrance, *Separation and Purification Technology*, **233**, 115998, http://doi.org/j.seppur.2019.115998.

Mangalapally H. P., Notz R., Hoch S., Asprion N., Sieder G., Garcia H., Hasse H., (2009), Pilot plant experimental studies of post combustion CO_2 capture by reactive absorption with MEA and new solvents, *Energy Procedia*, **1**, 963-970.

Mustafa N.F.A., Mohd Shariff A., Tay W.H., Abdul Halim H.N., Mhd Yusof S.M., (2020), Mass transfer performance study for CO_2 absorption into non-precipitated potassium carbonate promoted with glycine using packed absorption column, *Sustainability*, **12**, 3873, http://doi.org/10.3390/su12093873.

Pashaei H., Ghaemi A., Nasiri M., Karami B., (2020), Experimental Modeling and Optimization of CO_2 Absorption into Piperazine Solutions Using RSM-CCD Methodology, *ACS Omega*, **5**, 8432-8448.

Pashaei H., Zarandi M.N., Ghaemi A., (2017), Experimental study and modeling of CO_2 absorption into diethanolamine solutions using stirrer bubble column, *Chemical Engineering Research and Design*, **121**, 32-43.

Petrescu S., Mămăligă I., Ivaniciuc M., (1998), Enhancement factor of the mass transfer for chemisorption process, *Revista de Chimie*, **49**, 64-67.

Prentza L., Koronaki I.P., Nitsas M.T., (2018), Investigating the performance and thermodynamic efficiency of CO_2 reactive absorption – A solvent comparison study, *Thermal Science and Engineering Progress*, **7**, 33-44.

Quyn D., Rayer A.V., Gouw J., Indrawan I., Mumford K.A., Anderson C.J., Stevens G.W., (2013), Results from a pilot plant using un-promoted potassium carbonate for carbon capture, *Energy Procedia*, **37**, 448-454.

Ramazani R., Mazinani S., Jahanmiri A., Van der Bruggen B., (2016), Experimental investigation of the effect of addition of different activators to aqueous solution of potassium carbonate: Absorption rate and solubility, *International Journal of Greenhouse Gas Control*, **45**, 27-33.

Ramezani R., Di Felice R., (2019), Kinetics study of CO_2 absorption in potassium carbonate solution promoted by diethylenetriamine, *Green Energy & Environment*, , doi: 10.1016/j.gee.2019.11.004.

Rezazadeh F., Gale W.F., Rochelle G.T., Sachde D., (2017), Effectiveness of absorber intercooling for CO_2 absorption from natural gas fired flue gases using monoethanolamine solvent, *International Journal of Greenhouse Gas Control*, **58**, 246-255.

Salmón I.R., Cambier N., Luis P., (2018), CO_2 capture by alkaline solution for carbonate production: a comparison between a packed column and a membrane contactor, *Applied Sciences*, **8**, 996-1011.

Smith K., Lee A., Mumford K., Li S., Thanumurthy N., Temple N., Stevens G. (2015), Pilot plant results for a precipitating potassium carbonate solvent absorption process promoted with glycine for enhanced CO_2 capture, *Fuel Processing Technology*, **135**, 60-65.

Sreenivasulu B., Gayatri D.V., Sreedhar I., Raghavan K.V., (2015), A journey into the process and engineering aspects of carbon capture technologies, *Renewable and Sustainable Energy Reviews*, **41**, 1324-1350.

Tataru-Farmus R.E., Siminiceanu I., Bouallou Ch., (2007), Carbon dioxide absorption into new formulated amine solutions (I), *Chemical Engineering Transactions*, **12**, 175-181.

Tataru-Farmus R., Siminiceanu I., Bouallou Ch., (2006), Density of three amine aqueous solution: measuring and correlation, *Annals of the Suceava University*, **V**, 16-20.

Tataru-Farmus R.E., Harja M., (2017), Experimental equipments used in the study of carbon dioxide absorption (I), *Bulletin of the Polytechnic Institute of Iaşi, Chemistry and Chemical Engineering Section*, **63**, 23-33.

Tataru-Fărmuș R.E., Moldovanu, R.E., Harja M., (2019), Experimental equipments used in the study of carbon dioxide absorption (II), *Bulletin of the Polytechnic Institute of Iaşi, Chemistry and Chemical Engineering Section*, **65**, 51-58.

Thee H., Smith K.H., da Silva G., Kentish S.E., Stevens G.W., (2012), Carbon dioxide absorption into unpromoted and borate-catalyzed potassium carbonate solutions, *Chemical Engineering Journal*, **181-182**, 694-701.

Ünveren E.E., Monkul B.O., Sarıoğlan Ş., Karademir N., Alper E., (2017) Solid amine sorbents for CO_2 capture by chemical adsorption: A review, *Petroleum*, **3**, 37-50.

Valtz A., Coquelet C., Nikitine C., Richon D., (2006), Volumetric properties of the water + ethylenediamine mixture at atmospheric pressure from 288.15 to 353.15 K, *Thermochimica Acta*, **443**, 251-255.

Wang M., Lawal A., Stephenson P., Sidders J., Ramshaw C., (2011), Post-combustion CO_2 capture with chemical absorption: A state-of-the-art review, *Chemical Engineering Research and Design*, **89**, 1609-1624

Wang Y., Zhao L., Otto A., Robinius M., Stolten D., (2017), A review of post-combustion CO_2 capture technologies from coal-fired power plants, *Energy Procedia*, **114**, 650-665.

Wu X., Yu Y., Qin Z., Zhang Z., (2014), The Advances of Post-Combustion CO_2 Capture with Chemical Solvents: Review and Guidelines, *Energy Procedia*, **63**, 1339-1346

Wu Y., Mirza N.R., Hu G., Smith K.H., Stevens G.W., Mumford K.A., (2017), Precipitating characteristics of potassium bicarbonate using concentrated potassium carbonate solvent for carbon dioxide capture. Part I: Nucleation, *Industrial & Engineering Chemistry Research*, **56**, 6764-6774.

Younasa M., Rezakazemi M., Daud M., Wazir M.B., Ahmad S., Ullah N., Inamuddin, Ramakrishna S., (2020), Recent progress and remaining challenges in post-combustion CO_2 capture using metal-organic frameworks (MOFs), *Progress in Energy and Combustion Science*, **80**, 100849, doi 10.1016/j.pecs.2020.100849.