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Graphical Abstract

Stefania Moioli*, **Laura A. Pellegrini**

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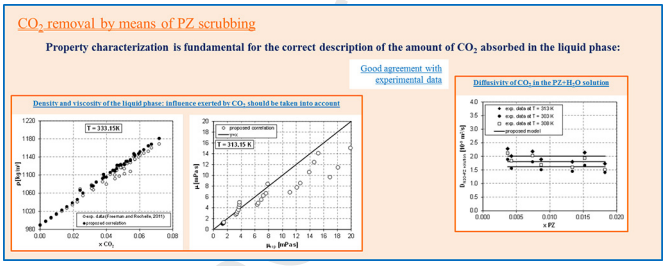
Physical properties of PZ solution used as a solvent for CO_2 removal



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Highlights

Stefania Moioli*, **Laura A. Pellegrini**

CHEMICAL ENGINEERING RESEARCH AND DESIGN XXX (2014) XXX–XXX

Physical properties of PZ solution used as a solvent for CO_2 removal

- Density and viscosity and CO_2 diffusivity of CO_2 loaded amine solutions are studied.
- A reliable description of physical properties is fundamental for a correct modeling.
- New correlations which takes into account the influence of CO_2 are proposed.
- An expression for diffusivity of CO_2 in the PZ solvent is proposed.
- The proposed expression allows a reliable representation of experimental data.

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Physical properties of PZ solution used as a solvent for CO_2 removal

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ABSTRACT

The climate change issues due to the emissions of carbon dioxide from various sources have become a challenge for many years because of the various negative effects on social, economic and environmental effects. The most commonly applied technique is absorption by chemical solvents. Piperazine, in particular, has been used in the past as activator when mixed to other amines, such as methyldiethanolamine. Nowadays in literature also aqueous solutions containing pure piperazine are considered for CO_2 capture. In this work properties such as density, viscosity and diffusivity of carbon dioxide related to this solvent have been carefully studied, taking into account the influence of carbon dioxide. New correlations have been proposed in order to best reproduce experimental data.

Keywords: Piperazine; Acid gas; Density; Viscosity; Diffusivity; Properties

1. Introduction

Carbon dioxide is emitted from various sources, as power plants, industries, transportation, agriculture, residential buildings and energy sector (Yang et al., 2008). Once emitted, CO_2 added to the atmosphere and oceans remains for thousands of years. Thus, climate changes forced by CO_2 depend primarily on cumulative emissions, making it progressively more and more difficult to avoid further substantial greenhouse effects (NOAA, 2013), and they have become a challenge for many years because of deleterious effects of climate change on social, economic and environmental issues. Therefore the interest in the development of CO_2 capture technology has increased due to the rising effects of global warming (IPCC, 2012) and many techniques have been developed, the most commonly applied being absorption by chemical solvents (Diamantonis et al., 2013; Khan et al., 2011; Lucquiaud and Gibbins, 2011; Mores et al., 2011, 2012; Posch and Haider, 2013; Sofia et al., 2014).

This technology is a mature technology, with monoethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA) (Khoo and Tan, 2006; Langé et al., 2013; Moioli et al., 2013; Pellegrini et al., 2011, 2013; Tuinier et al., 2010) being the most widely used solvents in industrial applications (Kohl and Nielsen, 1997). After the long time use of traditional amine-based solvent, various potential drawbacks, as inadequate lifetime due to amine oxidation degradation, losses of amines, low capacity of the solvent and high energy costs in the regeneration section (Giuffrida et al., 2013; Moioli and Pellegrini, 2013; Moioli et al., 2014; Shaikh et al., 2013) have been identified. Piperazine (PZ) has been found to be a very active promoter compared to other amines (Liu et al., 2012), because it leads to higher rates of absorption in the absorbing column while maintaining a low heat of regeneration in the stripper section. Nowadays also an aqueous solution containing only PZ is being considered (Kadiwala et al., 2010; Xia et al., 2003), since it can be very advantageous for CO_2 removal by means of a chemical absorption process.

However, while many published researches on the application of piperazine as activator can be found in literature (Alstom, 2009; Bishnoi and Rochelle, 2000, 2002; Sun et al., 2005; Xu et al., 1998), the use of this compound as single solvent has not been widely studied yet. For this reason, a reliable model based on thermodynamics (De Guido et al., 2014; Pellegrini et al., 2012a), mass transfer and physical properties

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is fundamental to well represent the absorption phenomenon occurring when dealing with PZ scrubbing. In particular, a correct description of properties of the absorbent solution, such as density and viscosity, and diffusivity of CO_2 in the used solvent are essential for modeling (Moioli and Pellegrini, 2014; Pellegrini et al., 2012b, 2014).

ASPEN Plus® uses generalized correlations for physical properties, in order to describe different mixtures by simply changing values of parameters. This generalization, however, affects the accuracy in describing the behavior of the amine system. Moreover, the influence exerted by the presence of the absorbed acid gases in the solution should be taken into account.

In this work a deep analysis of the properties involved in the calculation of mass transfer of absorbed species, i.e. CO_2, has been performed, in order to obtain reliable correlations for the description of density and viscosity of the PZ solution and of diffusivity of CO_2 in the solvent.

The obtained expressions have been implemented in the commercial simulator by means of an external Fortran user subroutine.

2. Theoretical basis

In literature several correlations for density of piperazine solutions are available, along with experimental data. Most of data (Derks et al., 2005; Liu et al., 2012; Muhammad et al., 2009; Samanta and Bandyopadhyay, 2006; Samanta et al., 2007; Sun et al., 2005) and of correlations (Freeman and Rochelle, 2011; Liu et al., 2012; Samanta and Bandyopadhyay, 2006) are related to temperature and PZ concentration; Freeman and Rochelle (2011) have studied the influence of the absorbed amount of CO_2, by determining experimental values for density and by proposing a correlation.

The model by Samanta and Bandyopadhyay (2006) well agrees with experimental data of density for a solution containing water and piperazine, covering all the range of temperatures for which experimental data are available, that is from 283.15 K to 338.15 K. This model (Samanta and Bandyopadhyay, 2006), however, does not take into account the influence of carbon dioxide, which should be considered for a complete modeling of the behavior of the rich amine solution. This is done by Freeman and Rochelle (2011), though the predicted densities are not very close to the experimental values for increasing values of CO_2 concentration in the solvent, so an improvement in the representation of available data (Freeman and Rochelle, 2011) can be obtained.

Also the viscosity of the liquid phase shows a strong dependence on temperature, piperazine concentration and the amount of absorbed carbon dioxide. At high loading, indeed, viscosity can assume values very different from the ones characteristic of a free PZ solution.

Correlations by Liu et al. (2012) and by Samanta and Bandyopadhyay (2006) are in good agreement with experimental data (Derks et al., 2005; Freeman and Rochelle, 2011; Liu et al., 2012; Muhammad et al., 2009; Samanta and Bandyopadhyay, 2006; Samanta et al., 2007; Sun et al., 2005). The former, however, shows a certain deviation from experimental values as the molar fraction of piperazine increases. In order to find a correlation suitable for a high range of amine concentration, the model by Samanta and Bandyopadhyay (2006) seems to be the most reliable among those already developed in literature. However, it does not take into account the influence exerted by the presence of carbon dioxide. As in the case of correlations for density, Freeman and Rochelle (2011) consider differences between the viscosity of a rich solution and the one of a lean solution, due to the contribution of CO_2. Dugas (2009) and Plaza (2012) have proposed two correlations, both of them based on the expression by Weiland, but characterized by different parameters one from the other. However, their models do not seem in very good agreement with experimental data.

For this reason a new correlation, based on the model by Samanta and Bandyopadhyay (2006), but with a contribution proportional to the amount of absorbed CO_2 has been proposed (see Section 2.1).

The diffusivity of carbon dioxide in the amine solution is fundamental in the determination of mass transfer rates. Despite the fact that a proper prediction is needed for a correct mass transfer modeling, $D_{CO_2,PZ\ solution}$ cannot be directly measured. Since CO_2 undergoes to reactions in PZ solution, indeed, the correct diffusivity is impossible to be obtained.

As in the case for MDEA solution (Bergman and Yarborough, 1978), in literature the analogy to N_2O diffusion coefficient is widely used:

$$D_{CO_2,PZ\ solution} = \frac{D_{N_2O,PZ\ solution}}{D_{N_2O,water}} D_{CO_2,water} \quad (1)$$

Versteeg and van Swaaij (1988) proposed correlations for $D_{CO_2,water}$ and $D_{N_2O,water}$, which well match experimental values (Al-Ghawas et al., 1989; Bindwal et al., 2011; Li and Lee, 1996; Mandal et al., 2004; Samanta et al., 2007; Versteeg and van Swaaij, 1988). Experimental data (Samanta et al., 2007;

Table 1 – Parameters of the proposed expression for the calculation of density of the liquid solution according to Eqs. (2)–(5).

Parameter	Value
A_0	0.7550
B_0	1.88660E−03
C_0	−3.60560E−06
A_1	3.17160E−04
B_1	7.00060E−07
C_1	−6.13370E−10
A_2	3.54370E−05
B_2	−1.75480E−07
C_2	2.21150E−10
m_{CO_2}	2.51273E+03

Table 2 – Per cent absolute average deviations (AAD%) of the considered expressions for the calculation of density of the liquid solution.

Correlation	AAD%	
	$PZ + H_2O$ solution	$CO_2 + PZ + H_2O$ solution
Liu et al. (2012)	0.5646	N/A
Samanta and Bandyopadhyay (2006)	0.0331	N/A
Freeman and Rochelle (2011)	0.9635	1.6236
Dugas (2009)	8.3400	4.6379
Proposed correlation	0.0331	0.5441

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Sun et al., 2005) of diffusivity of N_2O in PZ solutions are available and have been used to compare literature models and to develop a new expression (see Section 2.2).

In this work different correlations have been proposed to properly describe the density and the viscosity of the PZ solution and the diffusivity of carbon dioxide in the solvent.

2.1. *Proposed correlations for the density and the viscosity of the liquid solution*

A new correlation for the density of the PZ solution has been proposed with the aim of taking into account the effects of the presence of carbon dioxide in the liquid phase. It is based on the expression proposed by Samanta and Bandyopadhyay (2006), with an addition of a contribution due to the influence of CO_2:

$$\rho\left[\frac{kg}{m^3}\right] = (\rho_0 + \rho_1 + \rho_2)\cdot 1000 + m_{CO_2}\cdot x_{CO_2} \quad (2)$$

with:

$$\rho_0 = A_0 + B_0\cdot T + C_0\cdot T^2 \quad (3)$$

$$\rho_1 = A_1\cdot W_{PZ} + B_1\cdot W_{PZ}\cdot T + C_1\cdot W_{PZ}\cdot T^2 \quad (4)$$

$$\rho_2 = A_2\cdot W_{PZ}^2 + B_2\cdot W_{PZ}^2\cdot T + C_2\cdot W_{PZ}^2\cdot T^2 \quad (5)$$

and T in [K]. Parameters of Eqs. (2)–(5) are reported in Table 1. In Table 2 the per cent absolute average deviations (AAD%) of all the considered expressions, including the proposed one, are reported. The proposed correlation is characterized by the lowest value of AAD% for the representation of the density of both the free piperazine solvent and of the CO_2 loaded solution.

The proposed expression for the viscosity of the liquid solution is:

$$\mu\ [Pa\,s] = \frac{\exp(\mu_0 + (\mu_1/T) + \mu_2\cdot T)\cdot \exp(m_{1,CO_2}\cdot x_{CO_2}\exp(m_{2,CO_2}\cdot x_{CO_2}))}{1000} \quad (6)$$

Table 3 – Parameters of the proposed expression for the calculation of viscosity of the liquid solution according to Eqs. (6)–(9).

Parameter	Value
A_0	−1.61E+01
B_0	0.1599
C_0	−2.11E−03
D_0	2.44E−04
A_1	3.41E+03
B_1	1.95E−03
C_1	1.33E−03
D_1	−3.14E−03
A_2	1.54E−02
B_2	−4.28E−04
C_2	8.69E−06
D_2	−6.69E−07
m_{1,CO_2}	4.82290
m_{2,CO_2}	21.478

Table 4 – Per cent absolute average deviations (AAD%) of the considered expressions for the calculation of viscosity of the liquid solution.

Correlation	AAD%	
	$PZ + H_2O$ solution	$CO_2 + PZ + H_2O$ solution
Liu et al. (2012)	10.2766	N/A
Samanta and Bandyopadhyay (2006)	2.4309	N/A
Freeman and Rochelle (2011)	411.6352	228.3669
Dugas (2009)	14.7195	81.8026
Plaza (2012)	7.3040	19.5442
Proposed correlation	2.4309	14.3151

Table 5 – Parameters of the proposed expression for the calculation of diffusivity of carbon dioxide in the liquid solution according to Eqs. (10)–(12).

Parameter	Value
$A_{CO_2,water}$	2.350E−06
$B_{CO_2,water}$	−2119
$A_{N_2O,water}$	5.070E−06
$B_{N_2O,water}$	−2371
$A_{N_2O,PZ\ solution}$	0
$B_{N_2O,PZ\ solution}$	1.44218E+03
$C_{N_2O,PZ\ solution}$	2.05857E+03

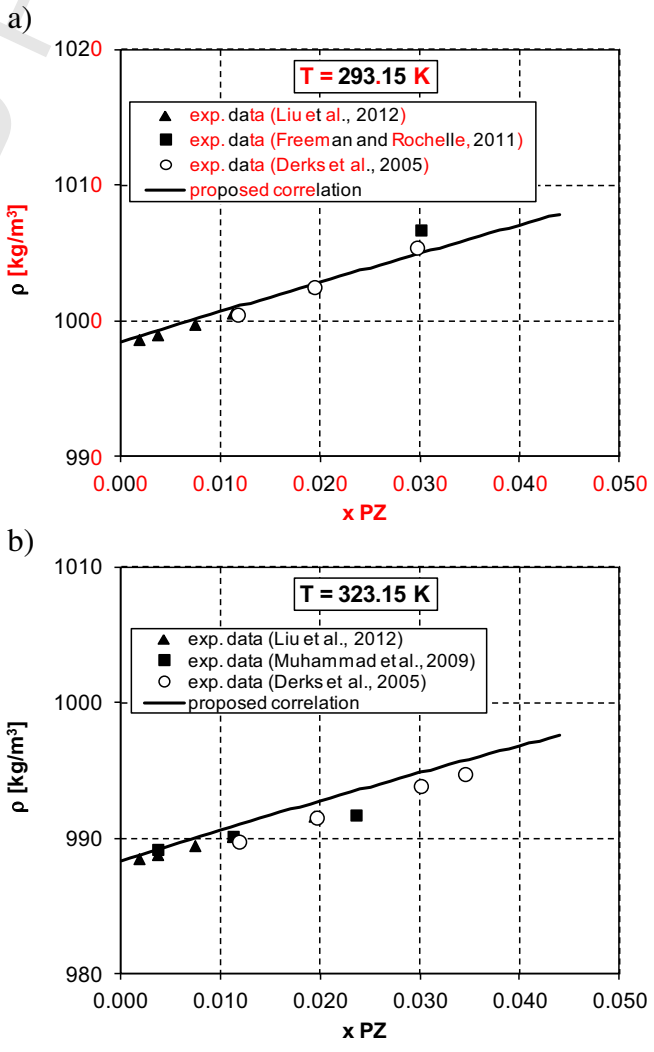

Fig. 1 – Results obtained with the proposed expression and experimental data of density of PZ solution vs. amine mass fraction at (a) 293.15 K and (b) 323.15 K.

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with T in [K] and:

$$\mu_0 = A_0 + B_0 \cdot W_{PZ} + C_0 \cdot W_{PZ}^2 + D_0 \cdot W_{PZ}^3 \quad (7)$$

$$\mu_1 = A_1 + B_1 \cdot W_{PZ} + C_1 \cdot W_{PZ}^2 + D_1 \cdot W_{PZ}^3 \quad (8)$$

$$\mu_2 = A_2 + B_2 \cdot W_{PZ} + C_2 \cdot W_{PZ}^2 + D_2 \cdot W_{PZ}^3 \quad (9)$$

and with parameters of Eqs. (6)–(9) reported in Table 3. AADs% reported in Table 4 show that the proposed correlation allows a good representation of experimental data of viscosity of pure PZ solvent. Values of per cent absolute average deviation are higher when considering the presence of carbon dioxide, however Eq. (6) is characterized by a value lower than the one of the other literature sources.

2.2. *Proposed correlation for the diffusivity of CO_2 in the liquid solution*

The calculation of $D_{CO_2,PZ\ solution}$ is based on Eq. (1), with $D_{CO_2,water}$ and $D_{N_2O,water}$ calculated according to Versteeg and van Swaaij (1988). As for $D_{N_2O,PZ\ solution}$, a regression of experimental data (Sun et al., 2005) has been performed in order to obtain a new correlation, though without considering the dependence on the amine concentration. A dependence had been considered firstly, but, due to the low number of experimental data, an extension to higher values of molar fraction of piperazine causes a negative value of diffusivity. This problem is due to the limited range of piperazine concentration

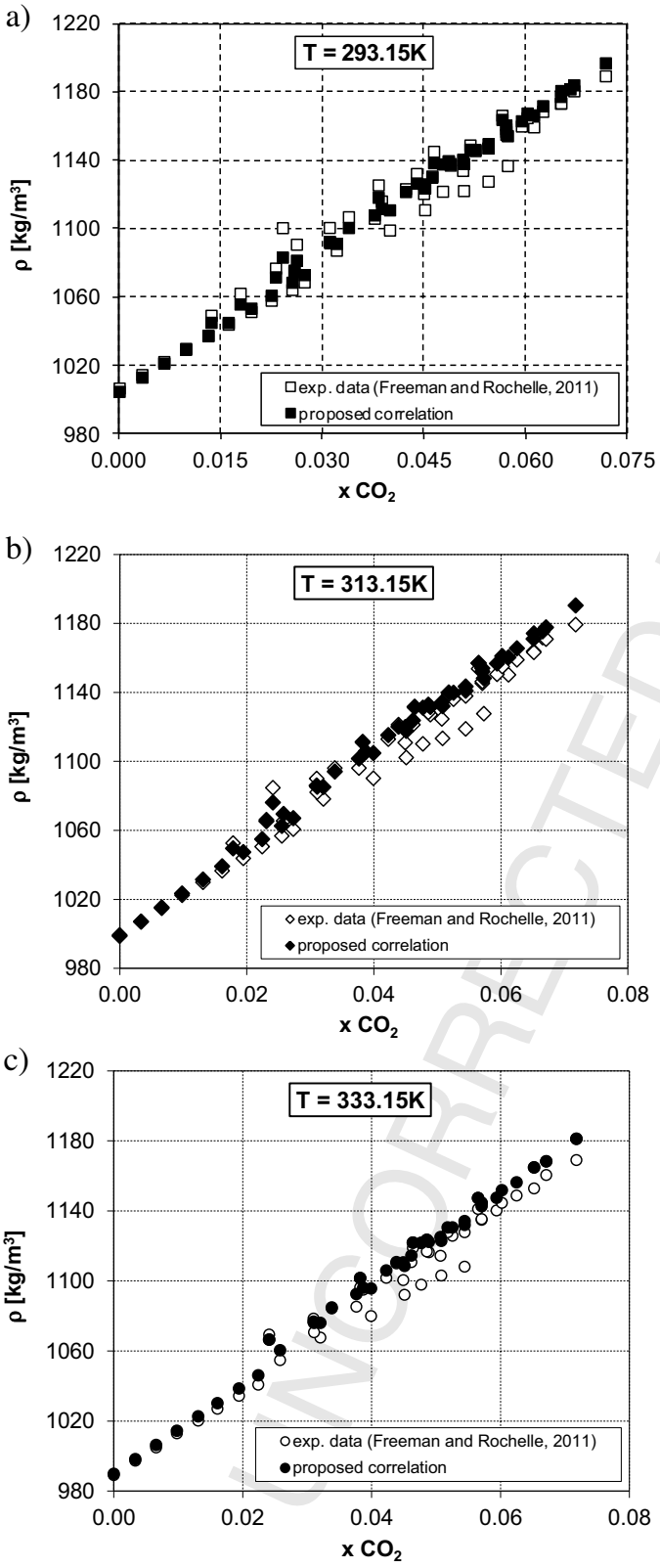

Fig. 2 – Results obtained with the proposed expression and experimental data of density of PZ solution vs. CO_2 mole fraction at (a) 293.15 K, (b) 313.15 K and (c) 333.15 K.

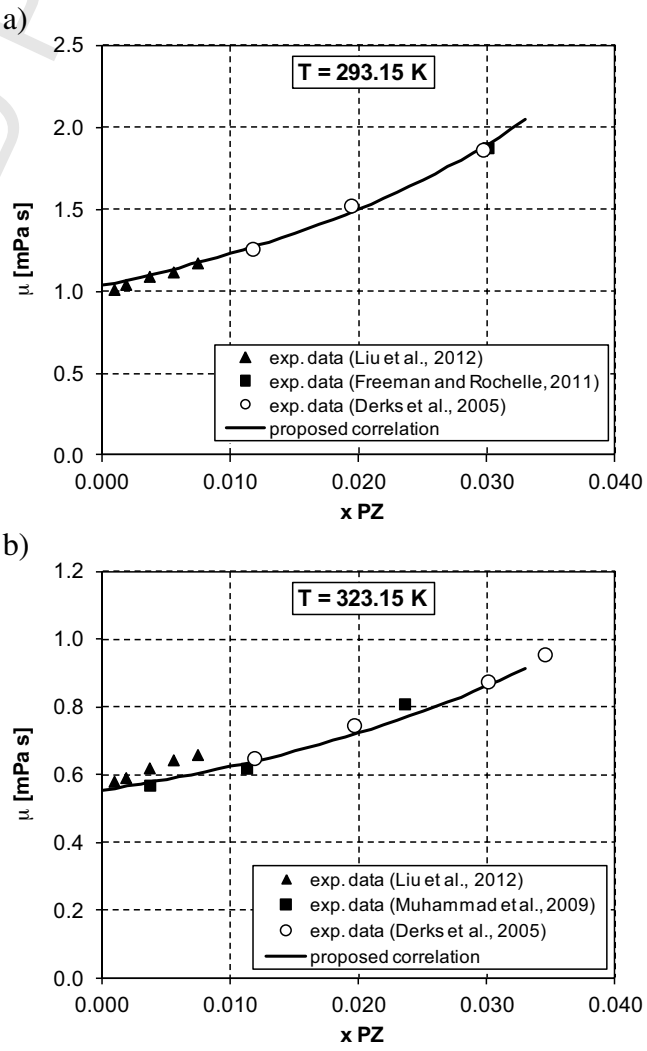

Fig. 3 – Results obtained with the proposed expression and experimental data of viscosity of PZ solution vs. amine mass fraction at (a) 293.15 K and (b) 323.15 K.

experimentally available, too low if compared to the amount of amine usually used to run plants (Plaza, 2012).

The proposed expression is then Eq. (1) with:

$$D_{CO_2,water}\left[\frac{m^2}{s}\right] = A_{CO_2,water}\cdot\exp\left(\frac{B_{CO_2,water}}{T[K]}\right) \quad (10)$$

$$D_{N_2O,water}\left[\frac{m^2}{s}\right] = A_{N_2O,water}\cdot\exp\left(\frac{B_{N_2O,water}}{T[K]}\right) \quad (11)$$

$$D_{N_2O,PZ\ solution}\left[\frac{m^2}{s}\right] = \left(-A_{N_2O,PZ\ solution}\cdot x_{PZ} + B_{N_2O,PZ\ solution}\cdot\exp\left(\frac{-B_{N_2O,PZ\ solution}}{T[K]}\right)\right)\times 10^{-9} \quad (12)$$

with parameters reported in Table 5.

3. Results and discussion

Fig. 1 shows that Eq. (2) well reproduces experimental data of density of free PZ solutions as well as the expression proposed by Samanta and Bandyopadhyay (2006) while in Fig. 2 the density of PZ solution vs. the amount of absorbed CO_2 at the temperature conditions for which experimental data are available is reported. Results obtained with the proposed expression show a good agreement with experimental density data in the presence of carbon dioxide.

Results obtained with the proposed model for the prediction of viscosity of the PZ solvent are shown in Figs. 3 and 4: the proposed expression for the viscosity of the PZ solution, properly modified starting from the correlation by Samanta

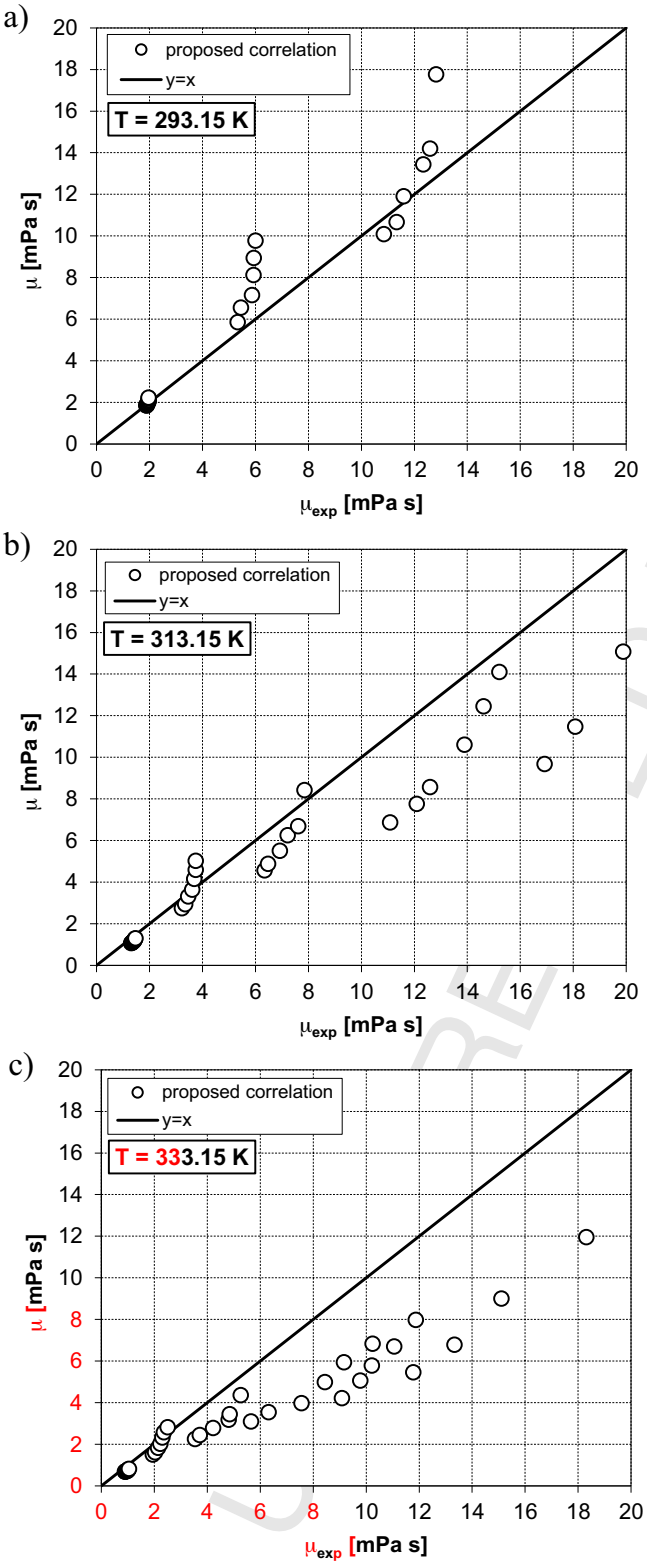

Fig. 4 – Parity plot of results obtained with the proposed expression and experimental data of viscosity of PZ solution vs. CO_2 mole fraction in the liquid solution at (a) 293.15 K, (b) 313.15 K and (c) 333.15 K.

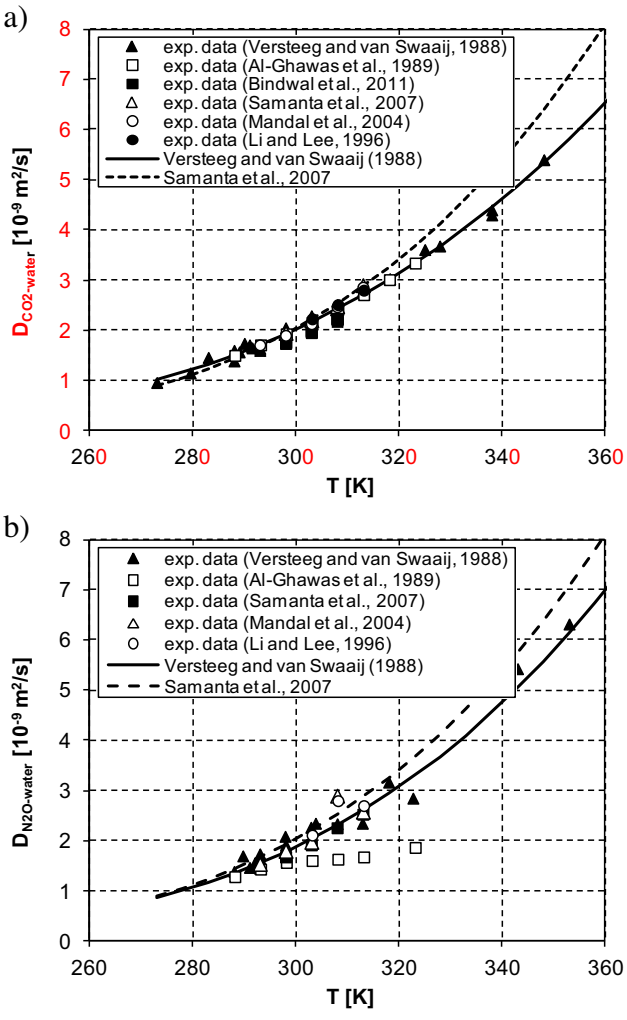

Fig. 5 – Diffusivity of (a) carbon dioxide and (b) nitrous oxide in water vs. temperature.

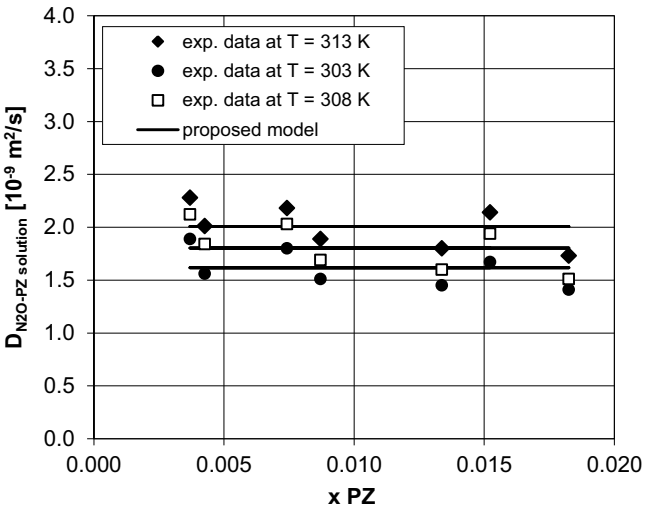

Fig. 6 – Experimental data (Samanta et al., 2007; Sun et al., 2005) and results obtained with the proposed expression for diffusivity of nitrous oxide in the PZ solution.

and Bandyopadhyay (2006), allows a good match with experimental data.

In Fig. 5 diffusivities of carbon dioxide and of nitrogen oxide in water are shown, while in Fig. 6 experimental data and results obtained with the proposed expression for diffusivity of nitrous oxide in the PZ solution are reported. The developed correlation for diffusivity allows a good estimation of $D_{N_2O,PZ\ solution}$, and so a reliable estimation of $D_{CO_2,PZ\ solution}$ according to Eq. (1) can be obtained. The correlation for diffusivity of N_2O in the PZ solution by Samanta et al. (2007) has not been considered because, in the form it is published, it results out of the range of Fig. 6.

The proposed expressions, then, can be employed to obtain a correct description of the physical properties involved in the modeling of the mass transfer of carbon dioxide from the vapor phase to the liquid phase when absorbed into the PZ solution.

4. Conclusions

In this work properties of the piperazine solution used as a solvent for CO_2 removal have been taken into account, by properly analyzing the ones which most influence mass transfer modeling.

In literature there are some correlations for density, viscosity and diffusivity of CO_2 in the PZ + H_2O mixture, along with experimental data. However, they are not in very good agreement with experimental data in most of the cases because they do not take into account the influence exerted by the presence of absorbed carbon dioxide.

In this paper a deep analysis of these properties has been performed, in order to obtain a reliable calculation for the CO_2 absorption process by PZ scrubbing, whose description needs to be as close as possible to the physical phenomenon.

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