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# Mass transfer performance of 2-amino-2-methyl-1-propanol and piperazine promoted 2-amino-2-methyl-1-propanol blended solvent in high pressure CO<sub>2</sub> absorption



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#### ABSTRACT

This paper reports the absorption performance study for 2-amino-2-methyl-1-propanol (AMP) and piperazine (PZ) promoted AMP (PZ+ AMP) blend for the removal of CO<sub>2</sub> from CO<sub>2</sub> rich-natural gas (NG) at high pressure conditions. The absorption experiments were conducted in a high pressure bench-scale absorption column packed with Sulzer metal gauze packing. The effects of PZ concentration (3–9 wt%), liquid flow rate (2.89 to 4.33 m<sup>3</sup>/m<sup>2</sup> h), and CO<sub>2</sub> concentration in NG (30–50%) on the mass transfer performance were evaluated in terms of CO<sub>2</sub> removal efficiency (%) and overall volumetric mass transfer coefficient based on partial pressure driving force ( $K_C a_v$ ). In addition, the effect of various operating pressures (0.1–5.0 MPa) has also been evaluated in terms of the overall volumetric mass transfer coefficient based on unit mol fraction ( $K_y a_v$ ). The results in this study have shown that, the mass transfer performance of PZ + AMP blended solvent is better than that of the single AMP solvent. The value of  $K_G a_v$  increases with increasing PZ concentration and liquid flow rate. However, this value decreases with increasing CO<sub>2</sub> concentration in NG. Other than that, the process performance can be increased by increasing the operating pressure, with a significant increase in the  $K_y a_v$  value which can be observed at operating pressures of higher than 2.0 MPa.

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#### 1. Introduction

Natural gas (NG) that consists primarily of methane (CH<sub>4</sub>) is one of the cleanest energy sources with high energy conversion efficiencies for power generation. Being a crucial commodity, NG is used for power (electricity), heating energy and fuel for fixed engines or motor transport and as chemical feedstock. The depletion of conventional NG resources has driven many oil and gas companies to explore CO<sub>2</sub>-rich NG reserves around the world. Some unexplored NG reserves such as the Natuna gas field in Indonesia, was reported to contain as high as 71% CO<sub>2</sub> (Suhartanto et al., 2001) while in Malaysia, CO<sub>2</sub> concentration may vary, up to 87% (Darman and Harun, 2006).

The presence of  $CO_2$  and moisture in gas pipelines that transport NG from offshore rigs to onshore purification plants pose high corrosion risks. Therefore,  $CO_2$  content in NG must be reduced before the NG can be transported through sub-sea gas pipelines to the gas purification plants. This step is important towards reducing the capital expenditure (CAPEX) for transporting less valuable gases through lengthy pipelines as well as the cost required for compressing these gases for onshore operations and end-users. Furthermore,  $CO_2$  must be removed from NG to increase its heating value as well as to meet the sales gas specifications. The removal of  $CO_2$  at the offshore platform capitalizes on the high pressure raw natural gas from gas reservoirs which could lead to high  $CO_2$  removal due to the high driving force for  $CO_2$ – $CH_4$  separation.

Commercial  $CO_2$  removal technologies include absorption, adsorption, membrane and cryogenic separation. The most widely used  $CO_2$  removal technology for offshore platforms is membrane separation (Echt and Meister, 2009). However, it has several limitations such as plasticization of the membrane especially at high pressure conditions and aggressive gas environment (Ahmad et al., 2015). Therefore, one of the established technologies which is chemical absorption using amines as the absorbent, could be an alternative for handling this sub-quality gas at offshore platforms due to its ability to handle  $CO_2$ -rich NG at high pressure condition.

Major industrial amine-based absorbents include monoethanolamine (MEA), diethanolamine (DEA), *N*-methyldiethanolamine

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(MDEA) and di-2-propanolamine (DIPA). Primary amine such as MEA is the most widely used alkanolamines absorbent due to its high reactivity with  $CO_2$  (Mandal et al., 2003). However, based on stoichiometry, the maximum  $CO_2$  loading capacity of alkanolamines are up to 0.5 mol  $CO_2$ /mol amine and the solvent would require high energy for regeneration. Another commercially attractive absorbent is the 2-amino-2-methyl-1-propanol (AMP) which has better properties such as high  $CO_2$  absorption capacity of up to 1.0 mol  $CO_2$ /mol amine, low regeneration energy (Mimura et al., 1997; Pei et al., 2008; Weiland et al., 2010) and more resistant to thermal degradation (Weiland et al., 2010). However, the AMP was reported to exhibit slow reactivity and has lower  $CO_2$  mass transfer rate compared to the MEA (Sartori and Savage, 1983). This issue could be overcome by adding promoters or activators in order to increase its reactivity.

The use of promoter blend with AMP increases the reactivity of the solvent. For example, piperazine (PZ) promoted AMP was used as an alternative absorbent for  $CO_2$  absorption at atmospheric pressure. It was proven that it has the capability of overcoming the limitations faced by the MEA. PZ has a cyclic and diamine structure which theoretically, can absorb two moles of  $CO_2$  for every mole of amine. Apart from its high reactivity, PZ is also highly resistant to thermal and oxidative degradations (Freeman et al., 2010). Numerous research developments in solubility, kinetic and mass transfer studies of PZ + AMP blended solvents have been reported at atmospheric condition (Artanto et al., 2014; Dash et al., 2011, 2012; Samanta and Bandyopadhyay, 2009; Sun et al., 2005; Yang et al., 2010). However, the performance of the PZ + AMP blended solvent at high pressure operation for  $CO_2$ -rich NG purification process has yet to be reported.

The removal of CO<sub>2</sub> from feed gas containing less than 15% of CO<sub>2</sub> concentration has been discussed in details for various operating parameters, conducted at atmospheric pressure (Aroonwilas and Tontiwachwuthikul, 1997, 1998; Aroonwilas et al., 2001; Artanto et al., 2014; Dey and Aroonwilas, 2009; Fu et al., 2012; Naami et al., 2012; Sema et al., 2013; Setameteekul et al., 2008; Zeng et al., 2011). However, literature on the process performance of CO<sub>2</sub> absorption studies at high pressure operation is still limited. Godini and Mowla (2008) had conducted an absorption study at operating pressures of up to 1.0 MPa. They discovered that by increasing the pressure and the ratio of the liquid/molar flow rate of gas (L/G), they were able to increase the absolute efficiency of H<sub>2</sub>S and CO<sub>2</sub> absorptions. In addition, Dash et al. (2014) reported a simulation study of 8 wt% PZ + 22 wt% AMP at column pressures of up to 2.0 MPa and found that the CO<sub>2</sub> capture (%) and rich loading had increased while keeping the column pressure at the same L/G.

Tan et al. (2012) reported the removal of 75 vol.% CO<sub>2</sub> in NG at 1.0 MPa using a mixed amines solvent, known as Stonvent-II in a pilot scale absorption column, packed with IMTP No. 25 packing. They discovered that the performance of liquid temperature at 35 °C was better than at 45 °C. Recently, Tan et al. (2015) had further investigated the CO<sub>2</sub> absorption performance of 50% CO<sub>2</sub> in NG gas stream at pressure conditions of up to 5 MPa for 20 wt% monoethanolamine (MEA) aqueous solution and MEA hybrid solution consisting of 20 wt% MEA with 40 wt% N-methyl-2-pyrrolidone (NMP)+40 wt% water in a counter current packed column. They found that the increased pressure had positive impacts on CO<sub>2</sub> removal performance for the MEA aqueous solution as well as for the MEA hybrid solution. In addition, Abdul Halim et al. (2015) studied the effects of process parameters on CO<sub>2</sub> absorption for 20%  $CO_2$  in NG using MEA at 5.0 MPa. Their study reported that the  $K_G a_v$ values for the effects of liquid flow rate, gas flow rate, MEA concentration and liquid temperature had followed the same trends of  $K_G a_v$  at atmospheric condition. However, the effect of various operating pressures at CO<sub>2</sub> content of higher than 20% in NG still requires further researches.

This research focuses mainly on evaluating the performance of single AMP and PZ+AMP blended solvent for the removal of CO<sub>2</sub> from CO<sub>2</sub>-rich NG at high pressure conditions. The mass transfer performance of this process was determined by conducting experiments in a high pressure bench-scale packed absorption column at various process parameters such as PZ concentration, liquid flow rate, and CO<sub>2</sub> concentration in NG. CO<sub>2</sub> concentration in the range of 30-50% in NG was chosen because it is within the range and slightly above practical applications in the membrane technology used at offshore platforms (Echt and Meister, 2009). The effect of various operating pressures of up to 5.0 MPa was also studied to match the offshore platforms' normal operating pressures of higher than 3.7 MPa (Echt and Meister, 2009). In this study, the mass transfer performances for CO<sub>2</sub> absorption were evaluated in terms of the efficiency (%) of CO<sub>2</sub> removal and  $K_G a_v$  for PZ concentration, liquid flow rate, and CO<sub>2</sub> concentration while the effect of operating pressure was evaluated in terms of  $K_{v}a_{v}$ . Both  $K_{G}a_{v}$  and  $K_{v}a_{v}$  are the overall mass transfer coefficients with differing units. The effect of operating pressure is more appropriate to be calculated in terms of  $K_{v}a_{v}$  where pressure unit does not interfere with the calculation.

### 2. Determination of overall volumetric mass transfer coefficient (*K<sub>G</sub>a<sub>v</sub>*)

The rate of absorption per unit volume of packed column is given in the following equation:

$$r = K_y a_v (y - y^*) \tag{1}$$

where  $K_y a_v$  is the overall volumetric mass transfer coefficient in the gas phase, while  $(y - y^*)$  is the mol fraction driving force for the gas phase.

The two-film theory is often used as a basis for diffusion plus chemical reactions (McCabe et al., 2005). Based on this theory, equilibrium is assumed at the interface and the overall resistance to mass transfer is obtained by adding the resistance of the individual phase, as shown in the following equation (McCabe et al., 2005):

$$\frac{1}{K_{y}a_{\nu}} = \frac{1}{k_{y}a_{\nu}} + \frac{1}{k_{x}a_{\nu}}$$
(2)

The term  $1/K_y a_v$  can be considered as the overall resistance to mass transfer, while the terms  $1/k_y a_v$  and  $m/k_x a_v$  are resistances in the gas and liquid films, respectively. The gas-film coefficients are often reported based on a partial pressure driving force instead of mole-fraction differences (as shown in Eq. (2)) and written as  $K_G a_v$  or  $k_G a_v$ , as shown below:

$$\frac{1}{K_G a_\nu} = \frac{1}{k_G a_\nu} + \frac{1}{E k_L^\circ a_\nu}$$
(3)

In Eq. (3),  $k_G a_v$  is the individual volumetric mass transfer coefficient for gas phase, based on partial pressure driving force; H is the coefficient for Henry's law; and  $k_L^{\circ}a_v$  is the mass transfer coefficient for the liquid phase without chemical reactions. Meanwhile, E is defined as the mass transfer flux with chemical reaction, divided by the mass transfer flux without chemical reaction.

For a rich gas containing higher than 10% of component A (solute) while component B is the inert/carrier gas, the decrease in total gas flow and increase in liquid flow must be accounted for in the material balance (McCabe et al., 2005). In this case, the total gas flow rate changes significantly along the height of the column because the inert gas concentration was varied between 10–90%. Therefore, the overall mass transfer coefficient should be written as

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