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Structure and activity relationships for CO₂ regeneration from aqueous amine-based absorbents

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A B S T R A C T

A study to determine the relationships between structure and activity of various amine-based CO₂ solvents was performed. The desorption of CO₂ from saturated solvents at 80 °C and atmospheric pressure was measured to assess the initial desorption rate and desorption capacities at pseudo-equilibrium. Evaluation of the desorption capacity at lower temperature, 80 °C, will give a better understanding for more energy efficient and lower circulation rate absorbent for CO₂ absorption process. Results showed that an increase in chain length between the amine and different functional groups in the solvent structure up to four carbon, results in an increase in initial desorption rate and also an increase in the desorption capacity at pseudo-equilibrium was observed for most solvents. Steric hindrance effect was noticed when a side chain with an alkyl group was present at α -carbon position to amine group in the structure. Increase in the number of the amine group in solvent structure, results in higher desorption capacity up to 75% of CO₂ is desorbed. Aromatic amines substituted with an amine group by a side chain at the cyclic ring shows an increase in desorption capacity at pseudo-equilibrium compare to other group substitution.

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1. Introduction

Although the absorption of acid gases such as CO₂ in aqueous amine solutions, like e.g. monoethanolamine (MEA) from natural gas seems proven technology, removal of CO₂ from flue gases is not as straightforward as it could be expected. In these currently used systems a major part of the operational costs is caused by the solvent regeneration (up to 40%). In industrial processes, high temperatures (>100 °C) are used to regenerate MEA solutions. Usually high-pressure steam is applied, which provides the heat of reaction and enables the transport of CO₂ out of the reactor. The regeneration process is usually done at temperatures in excess of the boiling temperature, as the chemical kinetics of regeneration increases with temperature. The energy consumption in the stripper reboiler is estimated to be 15–30% of the net power production of a coal-fired power plant for about 90% CO₂ removed. The development of improved solvents with lower regeneration energy requirement can therefore be identified as the highest priority research and development objective for amine-based CO₂

capture systems. Such improvements are needed to reduce the too high-energy consumption of current systems, which is the major contributor to the relatively high cost of this technology.

A complete and rigorous (rate-based) simulation model of chemical absorption–desorption process requires a large number of physico-chemical data (solvent viscosity, density, diffusivities, kinetics data, and equilibrium model). Moreover to characterize a large number of solvent systems with all this information is a tedious task and could not be performed for a few solvents in a short time. Also studies devoted to desorption are not as numerous as those concerning absorption. More specifically, the relationship of structure and regeneration characteristics of various amines-based absorbent for CO₂ is hardly studied. Therefore, a screening method has been developed for the desorption of CO₂ from various loaded amine-based aqueous absorbents. The main aim in this investigation is to perform the screening of various solvents on their regeneration behaviour at low temperature and atmospheric pressure, with respect to rapid regeneration rate and low evaporation solvents losses especially. Desorption of CO₂

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from various amine-based absorbents was measured to assess the amount of CO₂ desorbed and the overall rate of desorption at 80 °C. In this study the effects which were investigated are the chain length, number of functional groups, different functional groups substitution at the α -carbon to the amines group, cyclic amine, substituted cyclic amines, etc. compared the performance of MEA which acts as a base case. Based on these results a better understanding of the structural effect on CO₂ desorption will be developed. This study will be advantageous in the development of an improved efficiency amine-based CO₂ absorbent.

2. Experiment

The various amine-based absorbents were tested in a simple screening apparatus (see Fig. 1), in which relative rates of the desorption and desorption capacity at pseudo-equilibrium can be measured and compared to the MEA default case. Due to the selection of the lower temperature of 80 °C for these experiments, compared to the commercial process regeneration temperatures of 120 °C, complete equilibrium for these desorption experiments was difficult to achieve. For these desorption experiments pseudo-equilibrium in this study work was defined for which most of the absorbents were close to their equilibrium composition. As desorption of CO₂ is faster compared to the absorption process, a desorption time of 20 min was found to be sufficient for the evaluation. The apparatus was designed to operate at atmospheric pressure and temperatures up to 80 °C. In a typical experiment first the solvent was degassed for sufficient time (approximately 2 h) and then saturated with pure CO₂ for approximately 1 h. This saturation of solvent was performed at 30 °C temperature and at atmospheric pressure. Once the solvent is completely saturated, a sample is taken to determine the total CO₂ loading in the solvent by a desorption/titration procedure as described by Blauwhoff et al. (1984). Once the total CO₂ loading is determined the known volume of a saturated amine sample of 40 ml is taken from the absorption column and is transferred into the desorption vessel. The temperature inside desorption vessel was maintained at 80 ± 0.5 °C. To ensure that the temperature in the absorbent solution during desorption remained constant, the solution is stirred continuously with a constant speed in every experiment. Saturated absorbent solution reached 80 °C within approximate 2.5 min.

The carbon dioxide released during this heating time is measured by a gas burette connected to the desorption vessel. When the solvent temperature is 80 °C the valve of the gas burette is closed and the CO₂ is monitored by online CO₂ IR detector. At the same time pure N₂ gas is bubbled through the saturated absorbent to increase the interfacial area for CO₂ desorption. The vapour leaving the desorption vessel containing the CO₂ and N₂ is fed into a vertical condenser. The condenser was kept at a temperature of 20 °C. The gas leaving the top of the condenser was at 25 ± 1 °C and thus contained only about 3 vol.% of water vapour. Further in this outlet stream, N₂ gas is added for dilution. This diluted stream is passed through an ice trap with a temperature of approximately 2 °C to remove the remaining amount of water vapour from the stream.

Next this stream is sent to CO₂ IR detector where the amount of CO₂ in the stream is measured. To ensure that there was no significant depletion on the partial pressure of CO₂ during desorption experiment, the flow of N₂ gas is kept very low in the saturated absorbent. Hence the effect on CO₂ partial pressure was negligible in the desorption experiment. Therefore, the complete desorption experiment was at prevailing atmospheric pressure, as the pressure-drop during the experiment was very small. The N₂ gas flow in solvent and for dilution was kept constant in every experiment. After a certain time in this desorption experiment, when pseudo-equilibrium is reached, the total CO₂ loading in the desorbed solvent is determined by desorption/titration procedure as described by Blauwhoff et al. (1984). The time for each desorption experiment is kept constant. The amine concentration in the solution could vary with the type of compound only to, e.g. molecular weight and solubility. As a base case MEA absorbent 2.5 moles/l concentration was chosen for comparison.

3. Results and discussion

Examination of the desorption behaviour from a saturated amine-based solvent with pure CO₂, allows for a comparison of the initial desorption rates and desorption capacity defined as the amount of CO₂ that remains in the solvent at the experimental conditions at the end of the experiment. From the initial desorption rate, an indication can be obtained about the reactivity of the various amine-based absorbents investigated. It must be noted, however, that due to mass transfer effects,

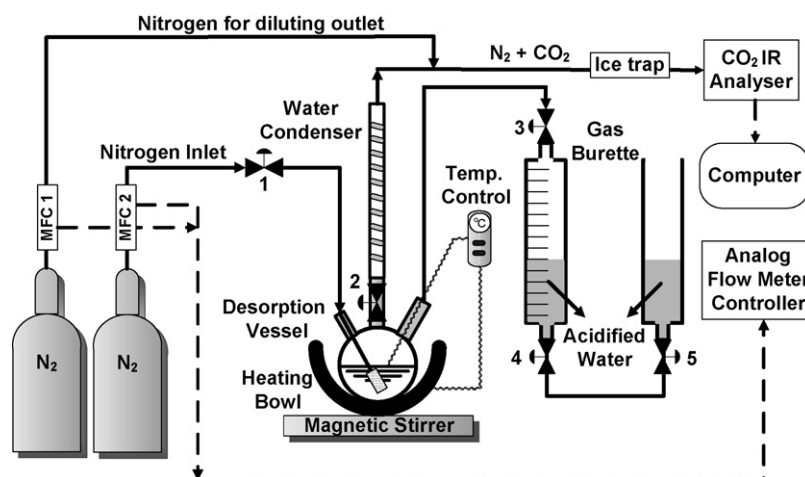


Fig. 1 – Schematic diagram of the experimental set-up for determining the desorption capacity of various amine-based absorbents.

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