



A kinetic and process modeling study of CO₂ capture with MEA-promoted potassium carbonate solutions

Hendy Thee, Yohanes A. Suryaputradinata, Kathryn A. Mumford, Kathryn H. Smith, Gabriel da Silva, Sandra E. Kentish, Geoffrey W. Stevens*

Cooperative Research Centre for Greenhouse Gas Technologies (CO2CRC), Department of Chemical and Biomolecular Engineering, The University of Melbourne, Victoria 3010, Australia

HIGHLIGHTS

- ▶ MEA significantly improves the overall absorption of CO₂ into a K₂CO₃ solvent.
- ▶ Reaction kinetics of CO₂ absorption into a MEA-promoted K₂CO₃ solution is reported.
- ▶ E-NRTL model suited for MEA-promoted K₂CO₃ system was developed using Aspen Plus™.
- ▶ MEA exhibits a larger rate constant at higher ionic strength conditions.

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ABSTRACT

Aqueous solutions of carbonate salts such as potassium carbonate (K₂CO₃) have gained widespread acceptance as viable solvents for pre and post combustion capture of carbon dioxide (CO₂). However, due to poor reaction kinetics a rate promoter is considered essential to improve the rate of CO₂ absorption and hydration to bicarbonate. Using a well characterized wetted-wall column, we have evaluated the reaction kinetics of CO₂ absorption into a K₂CO₃ solution promoted with monoethanolamine (MEA) under conditions resembling those found at industrial CO₂ capture plants. Results presented here show that at 63 °C the addition of MEA in small quantities, 1.1 M (5 wt.%) and 2.2 M (10 wt.%), accelerates the overall rate of absorption of CO₂ in a 30 wt.% potassium carbonate solvent by a factor of 16 and 45 respectively. The Arrhenius expression for the reaction between CO₂ and MEA is $k_{\text{MEA}} (\text{M}^{-1} \text{s}^{-1}) = 4.24 \times 10^9 \exp(-3825/T [\text{K}])$ where the activation energy is 31.8 kJ mol⁻¹. Incorporating our experimental results into Aspen Plus™, we have developed an E-NRTL model that can replicate pilot plant and simulate industrial capture processes employing K₂CO₃ promoted with MEA as the capture agent.

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1. Introduction

The removal and sequestration of carbon dioxide (CO₂) from the flue gas of coal-fired power stations by aqueous absorption is being actively investigated as a technology to help mitigate global warming due to human activities [1–3]. Aqueous solutions of both carbonate and amines, mainly monoethanolamine (MEA), are widely used in CO₂ removal processes [2,4–6]. Patents from Germany exploring hot carbonate absorption of CO₂ exist as early as 1904 [7]. In the 1950s Benson and Field developed the Benfield process which used hot potassium carbonate as a CO₂ absorption solvent, with an objective of reducing the costs of synthesis-gas purification in which high CO₂ partial pressures and high temperatures were employed [8–10]. This process was further developed during

the 1970s incorporating diethanolamine (DEA) as a rate promoter resulting in substantial lowering of capital and operating costs and higher treated gas purity [7].

In the carbonate system, CO₂ is hydrated to bicarbonate (HCO₃⁻) via the following overall reaction (NB: in this work all species are aqueous unless otherwise stated) [11]:



where the rate-limiting elementary reaction step is:



Under industrial CO₂ capture conditions where pH is greater than 9, the contribution of the direct reaction between CO₂ and H₂O can be deemed negligible [12]. The rate of reaction r_{CO_2} ($d[\text{CO}_2]/dt$) is thus given by:

$$r_{\text{CO}_2} = -k_{\text{OH}}[\text{CO}_2][\text{OH}^-] \quad (3)$$

* Corresponding author. Tel.: +61 3 8344 6621; fax: +61 3 8344 8824.

E-mail address: gstevens@unimelb.edu.au (G.W. Stevens).