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Investigation of catalysts used for the regeneration step of an amine-based absorption-regeneration CO_2 capture process

Célia Pasté, Lionel Dubois, Diane Thomas*

Chemical and Biochemical Process Engineering Unit, University of Mons (UMONS), Place du Parc 20, 7000 Mons, Belgium

Abstract

Global warming, primarily caused by CO_2 emissions from fossil fuel combustion, is a major environmental challenge. Over the last decades, the atmospheric CO_2 concentration has significantly increased, reaching 427 ppm in February 2025 compared to 316 in 1960 [1]. In response to this alarming rise, nearly two hundred countries signed the Paris Agreement in 2015, pledging to limit global temperature increases to below 2°C [2]. However, the growing demand for energy by industries is outstripping the capacity of renewable alternatives. Consequently, Carbon Capture, Utilization, and Storage (CCUS) is a promising solution according to the Intergovernmental Panel on Climate Change (IPCC) [3]. Among CO_2 capture techniques, absorption-regeneration process using amine-based solvents remains the most developed to date. Nevertheless, its high energy consumption poses a major challenge, which is driving research toward innovative strategies to reduce this energy requirement [4], [5].

Recent studies by Idem et al. [6] demonstrate that using a solid acid catalyst for the solvent regeneration can reduce the energy demand. These catalysts act in two distinct points during CO_2 desorption: a proton from a Brønsted acid site attacks the carbamate ($RNHCOO^-$) (equation (1)), and Lewis acid sites weaken the nitrogen-carbon bond which is essential for CO_2 release. Additionally, Brønsted acid sites regenerate protons through the deprotonation of the protonated amine (equation (2)). Therefore, this catalytic effect accelerates desorption kinetics, enhancing the overall efficiency of the process [6], [7].

$$RNHCOO^- + H^+ - B\ (Brønsted\ acid\ site) \rightleftharpoons RNH_2 + B\ (empty\ site) + CO_2 \quad (1)$$

$$RN^+H_3 + B \rightleftharpoons RNH_2 + BH^+ \quad (2)$$

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* Corresponding author. Tel.: +32 65/37.44.04
E-mail address: diane.thomas@umons.ac.be

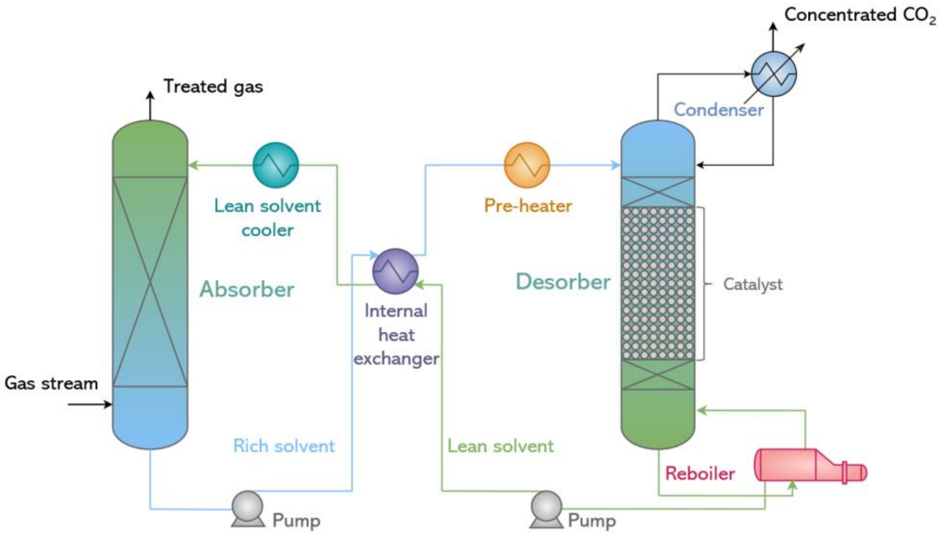

Figure 1: Schematic representation of the absorption- regeneration process including catalyst in the desorber (based on [8]).

This study assesses the potential of catalytic regeneration through a multi-stage approach (Figure 2). The first part includes a broad literature review conducted to identify the most promising catalyst candidates. Laboratory-scale and micro-pilot experiments are then carried out to evaluate the performance of catalytic regeneration with a MEA 30 wt% aqueous solvent. Finally, a comprehensive model (including thermodynamic, kinetic and hydrodynamic parameters) is developed on AspenPlus® to simulate a large-scale absorption-regeneration process incorporating catalysts, providing insights into their industrial viability.

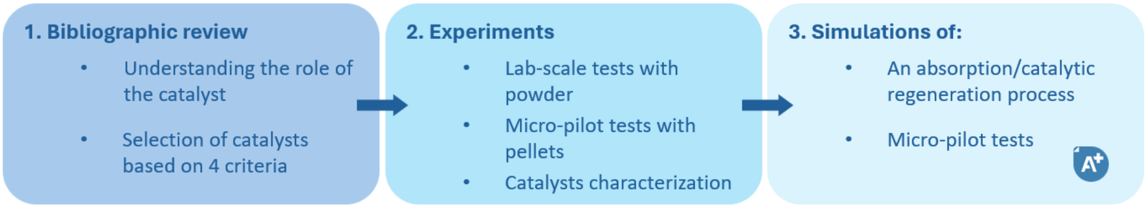

Figure 2: Overview of the work.

The selection of catalysts aims to identify the best candidates among those already studied in the literature. A total of 23 catalysts, directly available on the market (i.e., catalysts that have not undergone any special treatment or modification) were considered, including metal oxides, zeolites, mesoporous silicas, and natural clays. As completely described in [9], the selection is based on four key criteria: physicochemical properties, performance based on literature data, an economic aspect and a safety criterion. Through this screening, five metal oxides (γ-Al_2O_3, Nb_2O_5, MoO_3, Cr_2O_3 and ZrO_2) and three zeolites (H-ZSM5, H-Mordenite and H-Beta) emerged as the most promising candidates for further investigation.

Laboratory-scale experiments were conducted to evaluate the catalytic effect and assess the performance of the selected catalysts. The experimental setup used for the regeneration experiments of a 30% wt. MEA solvent with and without catalyst is illustrated in Figure 3. These tests allow for the determination of the CO_2 desorption rate over time for the different cases.

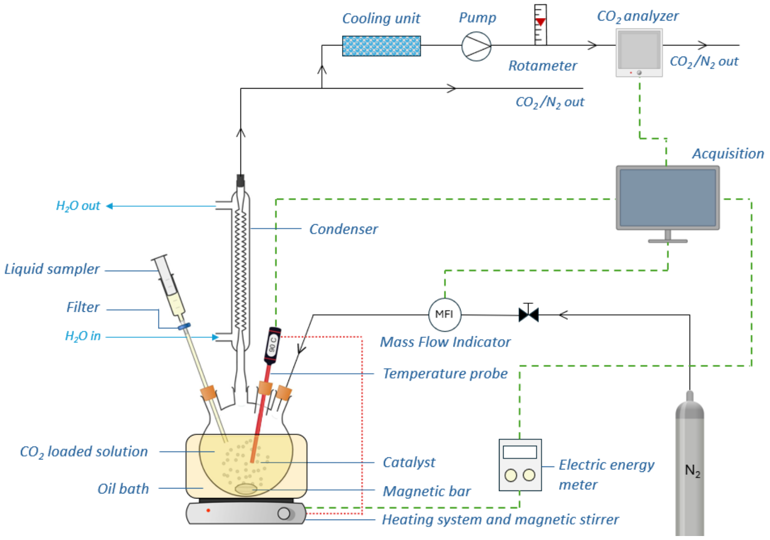

Figure 3: Lab-scale device for catalytic regeneration experiments.

Figure 4 (left) shows the evolution of the CO_2 desorption rate for a 30 wt% MEA solution with a loading of 0.55 ± 0.01 mol CO_2/mol MEA, comparing the cases without catalyst to those with 25 g/L of powdered γ-Al_2O_3, Cr_2O_3, and H-ZSM5 at a desorption temperature of 80°C. The presence of a catalyst increases the maximum desorption rate compared to the non-catalyzed case, with a particularly pronounced effect for γ-Al_2O_3 and H-ZSM5, which exhibit 24.6% and 21.0% higher peak desorption rates, respectively. Additionally, the catalysts induce an earlier desorption peak, indicating a faster CO_2 release. This trend is observed for all three catalytic cases. Notably, all desorption curves tend to converge toward the end of the experiment. This behaviour aligns with thermodynamic principles: as time progresses, the system approaches equilibrium, where the catalyst influences desorption kinetics but does not alter the final equilibrium state.

The amount of CO_2 desorbed is calculated by integrating the area under the desorption curve. In this study, the amount of CO_2 desorbed was calculated after 25 minutes. This parameter is relevant since the same amount of energy was supplied to the system over the 25-minute period. Figure 4 (right) shows the increased amount of CO_2 desorbed in the presence of catalysts compared to the 30 wt% MEA solution alone. γ-Al_2O_3 and H-ZSM5 cases stand out with an increase of 57 and 75%, respectively.

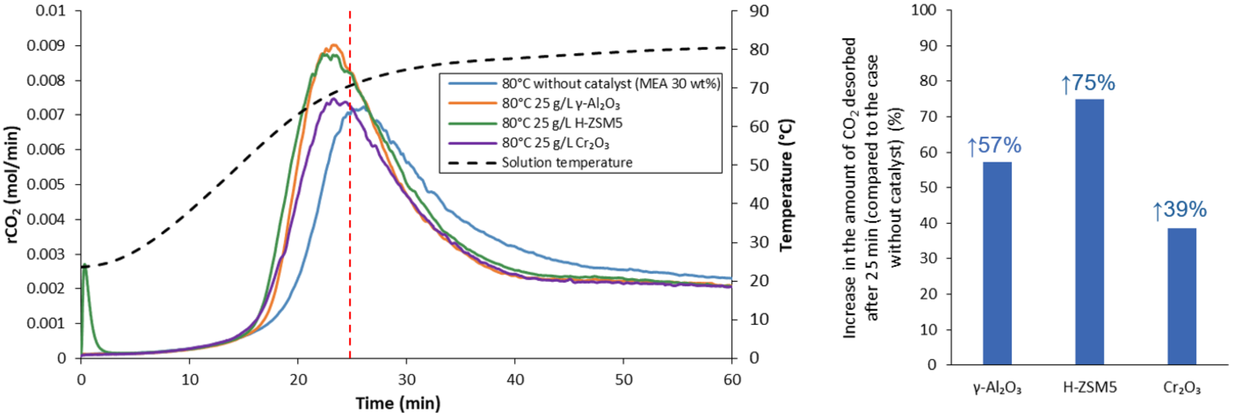

Figure 4: CO_2 desorption rate curves without and with catalysts (left) and Increase in the amount of desorbed CO_2 after 25 min (right) without and with catalysts (γ-Al_2O_3, H-ZSM5 and Cr_2O_3).

Alongside the experimental tests, the catalysts are characterized using various techniques. Nitrogen adsorption-desorption measurements determine the specific surface area and pore size distribution of the material. X-ray diffraction (XRD) analysis identifies their crystalline phases. Furthermore, the quantity and strength of acidic sites are assessed through NH_3-TPD measurements. Finally, pyridine adsorption coupled with infrared spectroscopy distinguishes and quantifies Lewis and Brønsted acid sites. The physicochemical properties obtained from these analyses are then correlated with the experimental results.

Finally, this study demonstrated that the tested catalysts γ-Al_2O_3, Cr_2O_3, and H-ZSM5 enhance amine solvent regeneration by accelerating desorption. Beyond laboratory-scale experiments and catalyst characterization, micro-pilot tests and larger-scale simulations will be conducted to further evaluate the energy-saving potential associated to the catalysts. These evaluations aim to improve the industrial absorption-regeneration CO_2 capture applications.

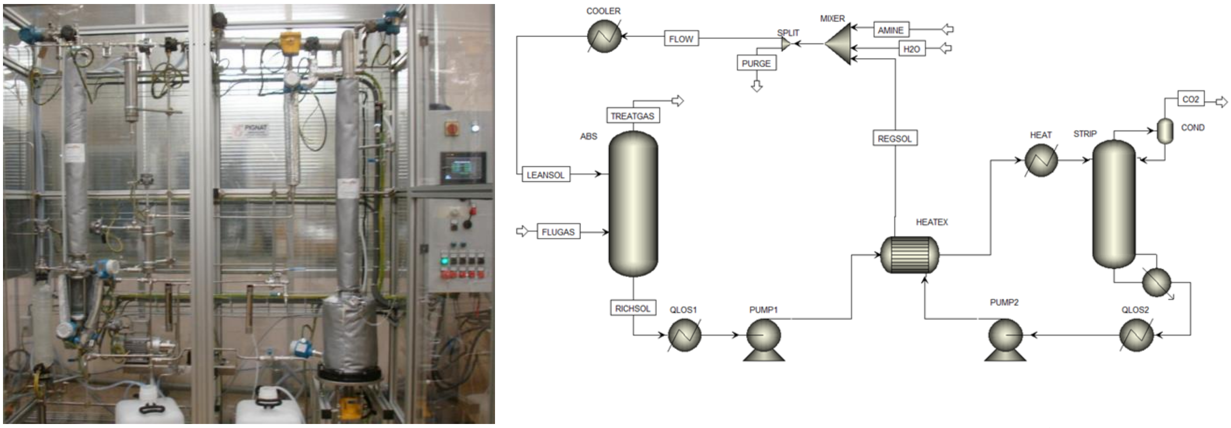

Figure 5: Micro-pilot unit for absorption-regeneration experiments (UMONS) and the corresponding AspenPlus® flowsheet.

Keywords: Absorption-regeneration carbon capture process; Amine-based solvents; Solid acid catalysts; Zeolites; Metal oxides;

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