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From MEA 30 wt.% to demixing solvents: investigation of an aqueous-alcoholic MEA-based solution

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Abstract

Since the beginning of the industrial era, the atmospheric carbon dioxide concentration has drastically grown, from around 285 ppm in 1850[1] up to 427 ppm in February 2025[2]. It approximatively corresponds to a 50% increase and 40% of this rise has been observed only in the last 25 years. Such a CO_2 level in the atmosphere is responsible, not only for a significant part of the current global warming, but also consequently for more intense and more frequent extreme weather phenomena. This CO_2 concentration increase and its consequences are mainly driven by the anthropogenic carbon dioxide emissions which have reached 41.6 $GtCO_2$ in 2024[3]. Reducing the atmospheric CO_2 concentration therefore inevitably involves a decline of the anthropogenic CO_2 emissions. This reduction can be partially achieved by changing the production methods for heat, electricity and various materials and the means of transport. Nevertheless, the emergency related to the impacts of CO_2 emissions requires to take actions, in the short term, on the already-developed emitting industrial processes and, in the long term, on processes emitting intrinsic and unavoidable carbon dioxide such as cement or limestone plants.

Among the available technologies, the absorption-regeneration carbon capture process using chemical solvents is the most mature technique for industrial level applications of post-combustion capture (for typical CO_2 concentrations between 5 and 30 mol.%). However, the major penalty of such a process is the thermal energy required for the solvent regeneration, once carbon dioxide is absorbed. For the most common benchmark in the CO_2 absorption sector (aqueous monoethanolamine MEA – 30 wt.%), it is between 3 and 4 GJ/tCO_2[4]. This energy demand represents the major part of the operative costs[5]. Its reduction is therefore a key element to ensure the affordability of the absorption-regeneration process. Various options can be implemented to reduce this regeneration energy such as alternative configurations or new packed beds in the columns. The solvent itself is of major importance to achieve this goal. Demixing (or biphasic) solvents are one category of emerging solvents, currently under research[6]. Under certain conditions of CO_2 concentration and temperature, such a solvent splits into a CO_2-lean phase and a CO_2-rich phase. Only a solvent fraction is directed to the regeneration column (see Figure 1) and has a higher CO_2 concentration than classical non-demixing solvents. Both features can significantly reduce the regeneration needed at the reboiler.

The present work was preceded by a selection of the liquid-liquid biphasic solvents already investigated in literature (see Figure 2). Some existing demixing solvents (such as DMX™ blend[7]) are proprietary solvents (without open access to their composition). The present study aims to focus on demixing systems with an open-access composition. The selection methodology includes both a techno-economic step and a health, safety and environment step[8]. This review and selection process highlights the promising

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nature of amine-based solvents, replacing a substantial part of the water content by an alcohol, especially propan-1-ol. The presence of an alcohol is used in these ternary mixtures (aqueous-alcoholic amine) as a phase-splitting agent which induces the demixing phenomenon. In addition to the benchmark (aqueous MEA 30 wt.%) used as a reference solvent, a demixing amine-based solvent, containing water and propan-1-ol (PROP) as non-reactants components, was selected for the other parts of the project. In this solvent, MEA (30 wt.%) is kept as the reactive chemical but a part of water is replaced by propan-1-ol (40 wt.%). Water finally weighs only 30 wt.%. The following parts of the research project involve various experiments and simulations at different scales (see Figure 2).

At the laboratory scale, various physico-chemical properties are measured and some comparisons are made on CO_2 absorption performances to the benchmark solvent. These data are a first step to confirm the potential interest of such solvents to be used in an absorption-regeneration process at larger scale. Different CO_2 absorption tests were performed to assess the demixing behavior of the investigated biphasic solvent. For this biphasic blend, the demixing phenomenon occurs above 1.3 molCO_2/L, at around half the absorption capacity. At the CO_2 saturation, 97.9% of the total CO_2-content in the solvent is contained in only 60% of the total volume. Dynamic viscosity and density were also measured at different CO_2 concentrations. These results show that the highest viscosity value was 19.5 mPa.s at 25 °C which is completely suitable for the absorption-regeneration process (taking place at higher temperatures). Absorption performances were evaluated through a cable-bundle contactor. On this experimental device, continuous tests (with a single pass of the liquid solvent) show similar absorption ratio for the blend MEA (30 wt.%) – PROP (40 wt.%) and the reference solvent (aqueous MEA – 30 wt.%), with a slight decreasing trend with the increasing of CO_2 gas fractions for the demixing solvent (see Figure 3). Semi-continuous tests (with recirculation of the solvent) have shown a material loss for the demixing solvent. It highlights the volatility of propan-1-ol and the need to adapt the process design (e.g. addition of a water-wash section and/or a condenser), considering this solvent feature.

The investigation on the demixing blends also includes experiments at an intermediate level in a micro-pilot unit (see Figure 4), composed of an 1m-packed bed (designed to treat around 3 Nm^3/h of flue gas) and a liquid-liquid phase separator unit. This pilot unit is also designed to perform regeneration experiments, under pressure (up to 3 barg). Indeed, the regeneration of aqueous MEA (30 wt.%) is commonly optimized at a pressure of 2 bar.

The final part of this work is the simulation of the absorption regeneration process at industrial level using AspenPlus®. For the liquid-liquid phase separation modeling, the thermodynamic model (eNRTL with the Helgeson model[9]) was selected by a comparison with small-scale experimental data (especially, CO_2 concentration volume proportion in each phase). The modeling was performed for a flue gas (32 000 m^3/h, $yCO_2 = 24$ mol.%) coming from a limestone plant. For the demixing blend (MEA 30 wt.% – PROP 40 wt.%), the flowsheet includes a condenser at the top of the absorption column to tackle potential propan-1-ol (see Figure 5). The estimated specific regeneration energy is 2.68 GJ/tCO_2. For the same parameters, the modeling of the absorption-regeneration process with the benchmark (aqueous MEA 30%) assesses a value of 3.32 GJ/tCO_2. This 20% reduction already demonstrates the interest of replacing a part of water (from the benchmark solvent composition) by propan-1-ol to decrease the operative costs of such a process. Following the selection methodology developed in the first part of the work, other amine-based aqueous-alcoholic solvents are also promising to further reduce the regeneration energy of a carbon capture process by absorption.

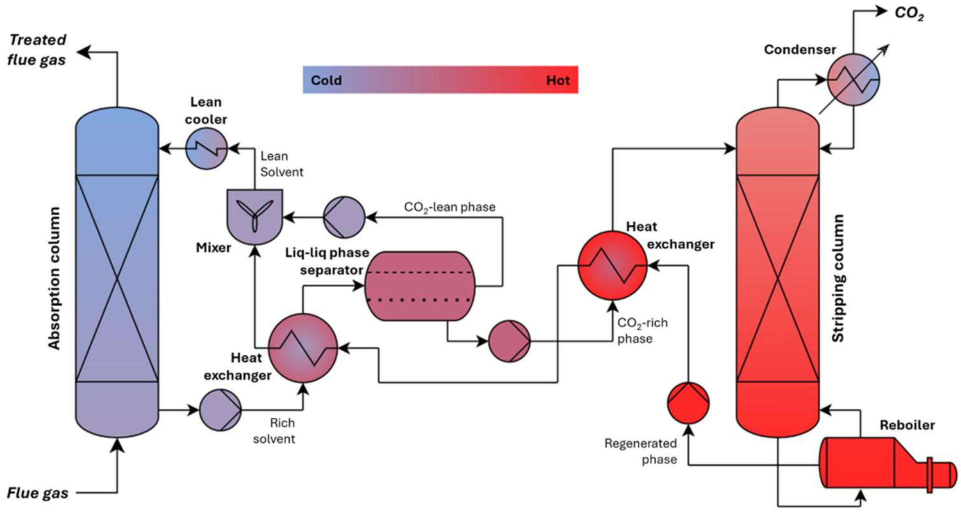

Figure 1. Typical flowsheet of absorption-regeneration carbon capture process using demixing solvents

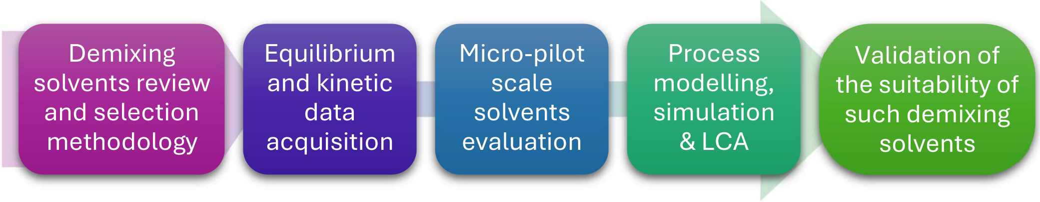

Figure 2. Main parts of the research project on demixing solvents

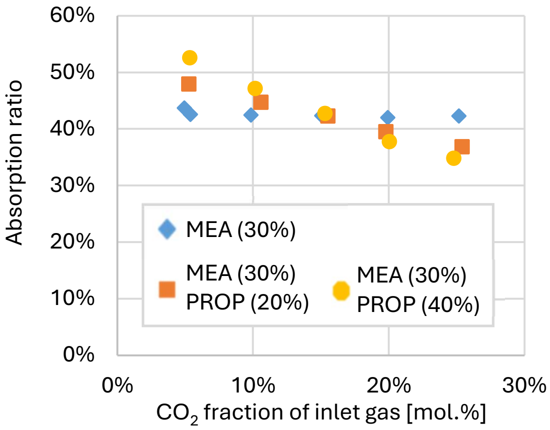

Figure 3. Absorption ratio for MEA-based blends

Figure 4. Micro-pilot unit for demixing solvents

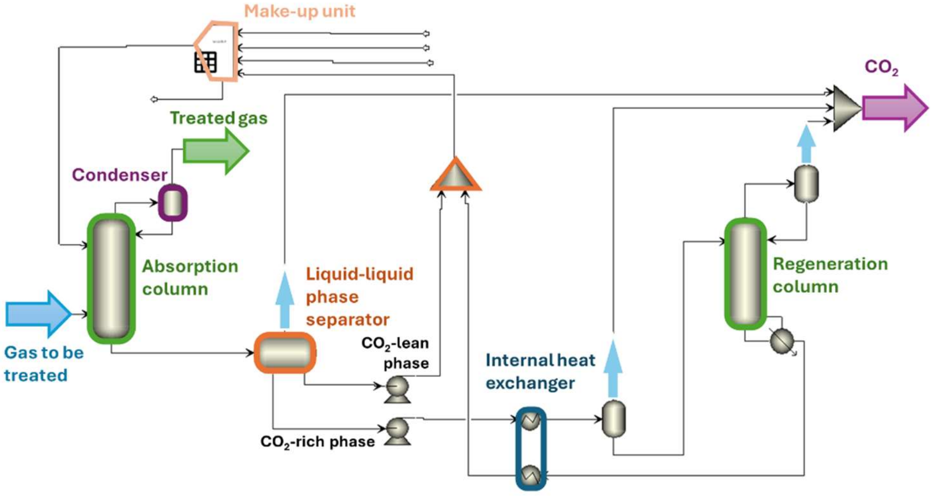

Figure 5. AspenPlus® flowsheet for MEA (30 wt.%) - PROP (40 wt.%) blend

Keywords: Carbon capture, Absorption-regeneration process, Biphasic amine-based solvents, Aqueous-alcoholic solution

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