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Investigation of a MEA-based CO_2 capture process applied to fluctuating flue gases through dynamic simulations

Lionel Dubois*, Antoine Verhaeghe, Guy De Weireld and Diane Thomas

Chemical & Biochemical Process Engineering and Thermodynamics Units, Faculty of Engineering, University of Mons, 20, Place du Parc, 7000 Mons, Belgium.

Abstract

Implementing CCUS (Carbon Capture Utilization and/or Storage) appears to be an important part of the solution in order to significantly decrease the carbon dioxide emissions from several industrial sectors (e.g. steel, cement, lime, glass, etc.). For example, in its 2025 Net Zero Roadmap [1], the Belgian Cement Industry Federation (FEBELCEM) highlighted that CCUS should contribute to 62% in all the cement decarbonation levers by 2050. Focusing on the end-of-pipe post-combustion CO_2 capture technology, especially the absorption-regeneration process using amine-based solvent, it requires limited adaptations for the upstream industrial process but the thermal energy needed for the solvent regeneration is still very high. It is therefore mandatory to work for the process improvement (e.g. innovative solvents, process configurations, …) but also to efficiently manage the process, especially when it is applied to fluctuating flue gases, such as in the lime industry.

Indeed, the thermal decomposition of limestone in a kiln ($CaCO_{3(s)} \rightarrow CaO_{(s)} + CO_{2(g)}$) to produce lime, known as calcination, occurs typically at temperatures above 900°C and Parallel Flow Regenerative Kilns (PFRK) are the most popular type of kiln for this application in Europe. Such process involves alternating between heating one shaft and cooling the other, which helps in recovering heat and improving fuel efficiency. Nevertheless, this inversion phenomenon between two operating modes leads to a fluctuating flue gas, both in terms of flow rate and composition (CO_2, O_2, H_2O, …). For example, a typical cycle of 15 min, could include ~11 min at nominal conditions and ~4 min at low inlet CO_2 gas flow (e.g. 25% of the nominal value). Such fluctuations could also be potentially observed in other industrial processes with different fluctuation parameters (see Table 1).

In that context of the application of the amine-based CO_2 capture process to fluctuating flue gases, the present work investigated the dynamic answer of the CO_2 capture process to these fluctuations, including the investigation of technical solutions to keep a captured CO_2 flow as stable as possible. For such purpose, different simulations were performed using Aspen Plus® and Aspen Dynamics® (V14.0) software considering monoethanolamine (MEA) 30wt.% as solvent and a conventional process configuration.

As detailed in [2], the developments were divided in two steps: (i) Aspen Plus® steady state simulations were performed in order to validate the model used for the dynamic simulations based on micro-pilot results; (ii) the Aspen Plus® steady state model was converted to Aspen Dynamics® and combined to Matlab-Simulink® interface in order to manage the temporal evolution of all the simulation inputs and outputs (see Fig. 1).

The Aspen Plus® simulation model was developed in equilibrium mode (mandatory for the conversion to Aspen Dynamics®), the thermodynamic models used being Redlich Kwong (RK) and Electrolyte Non-Random Two-Liquid (ELEC-NRTL) for the gas and liquid phases respectively. The reaction set implemented in the columns (equilibrium reactions only allowed), corresponding to the CO_2-MEA-H_2O chemistries, includes the different dissociation equilibrium reactions [2].

* Corresponding author. Tel.: +32(0)65/37.50.54, *E-mail address:* lionel.dubois@umons.ac.be

The investigations were performed considering the following conditions: a nominal flue gas flow rate of 34 000 m³/h (at 110°C and 1 atm), a nominal CO_2 concentration in the flue gas of 25 vol.%, a nominal absorption rate (amount of CO_2 captured relatively to the inlet CO_2 content) of 90% and a recovered CO_2 purity of 99 wt.%. All the operating and dimensioning data are based on the same process investigated in [2]. The liquid-to-gas optimum volumetric ratio (minimizing the regeneration energy) in the absorber, corresponding to a liquid flow rate of 192 m³/h, was determined as performed in [3] using the steady state Aspen Plus® simulations considering the nominal operating data. This ratio varied between 4.65 10^{-3} m³/m³ and 1.08 10^{-2} m³/m³ due to the temporal variation of the inlet gas flow rate. It has to be noted that other gaseous components (e.g. SO_x, NO_x, …) were not considered in the present study. The simulated Aspen Dynamics® flow sheet is provided in Fig. 1. Two hours of operations (corresponding to Fig. 2 (left) temporal profiles) were simulated considering different regulation loops comprising PID (Proportional-Integral-Derivative) controllers. The PID parameter values (gain, integral and derivative times) were optimized (starting from Aspen Dynamics® default values) in order to keep an efficient regulation all along the process. As illustrated on Fig. 2 (left) for three cases, different flue gas parameters were varied, namely the fluctuation frequency, the fluctuation time and the fluctuation amplitude (see Table 1).

It can be seen on Fig. 2 (right) that due to the flue gas variations, the produced CO_2 flow (at the stripper's outlet) is varying (e.g. for flue gas – 1, the produced CO_2 flow is decreasing during ~4 min, reaching a minimum value corresponding to a decrease of around 7% of the nominal value). Concerning the effect of the fluctuation parameters, it was shown that quite logically, reducing the fluctuation amplitude (e.g. ~50% instead of ~25%) leads to a reduction of the produced CO_2 variation amplitude (e.g. see results with flue gas – 2 and flue gas – 3 on Fig. 2). Moreover, the reduction of the fluctuation time (e.g. ~2 min instead of ~4 min) allows to keep the nominal CO_2 production flow during a larger period of time. Concerning the fluctuation frequency (e.g. ~9 min instead of ~15 min), it could be highlighted that a more frequent perturbation (together with an intermediate value of fluctuation amplitude) leads to a more fluctuating produced CO_2 flow but with a smaller variation amplitude (e.g. only max 3% of produced CO_2 flow variation with flue gas – 3 on Fig. 2). These effects on the CO_2 production flow can be related to the effect on the solvent CO_2 loading value, especially the rich solution one (see Fig. 3). Such observations are interesting in view of the application of this CO_2 capture technology to fluctuating flue gases (e.g. if technical measures can be taken to reduce the fluctuation time and amplitude, even with a slightly higher fluctuation frequency, that could be potentially beneficial in view of smoothing the CO_2 production flow). It has to be noted that technical ways were also investigated to smooth the temporal variations of the CO_2 production flows, namely the regulation of the reboiler heat duty, the implementation of a liquid tank on the rich solvent line, the recirculation of a part of the treated flue gas and produced CO_2 flow, and also the combination of some of these solutions. These pathways showed their efficiency in smoothing the CO_2 production with a limited CO_2 capture cost increase (<5%).

As a conclusion, this study pointed out that even if an industrial plant generates a flue gas with some temporal fluctuations on its inlet CO_2 content, implementing an amine-based absorption-regeneration CO_2 capture process is feasible despite the fluctuations on the recovered CO_2 flow. Obtaining a more "stable" captured CO_2 production flow is possible by acting on the upstream process characteristics (e.g. modifying fluctuation time, frequency and/or amplitude) or by implementing technical solutions like reboiler duty regulation and/or the addition of a liquid tank.

Figures and Table

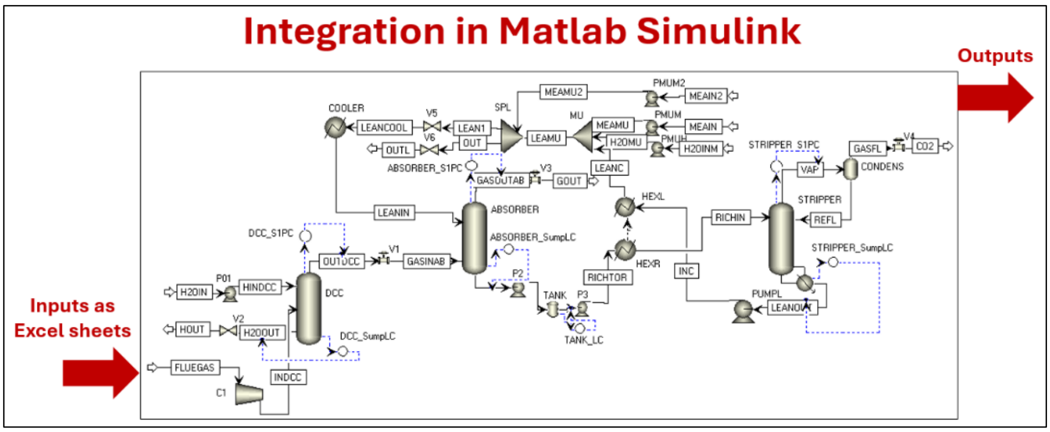

Fig. 1. Aspen Dynamics® simulation flow sheet using MEA 30wt.% as solvent included in a Matlab-Simulink® interface.

Table 1. Flue gas parameters considered for the different dynamic simulations, namely FF: Fluctuation Frequency (time between two minima), FT: Fluctuation Time (time below nominal value for one cycle) and FA: Fluctuation Amplitude (minimum CO_2 content/nominal CO_2 content (%)).

Flue gas n°	FF	FT	FA
1	~15 min	~4 min	~25%
2	~15 min	~2 min	~50%
3	~9 min	~2 min	~50%

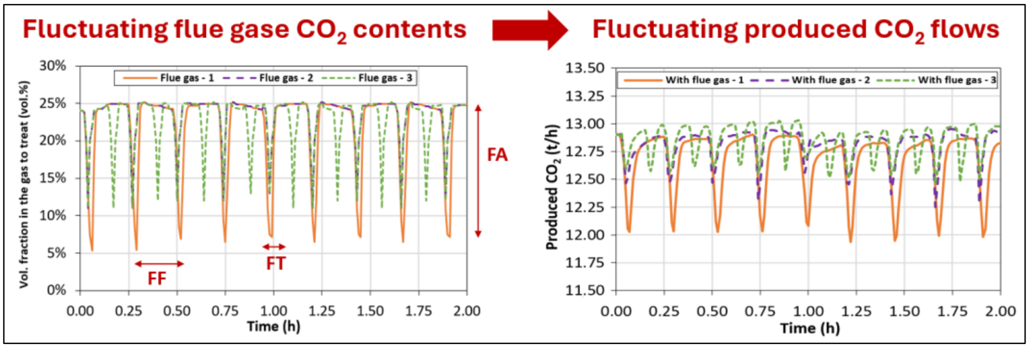

Fig. 2. Example of a fluctuating flue gas CO_2 contents (left) and the associated produced CO_2 flows (right) from the dynamic simulation

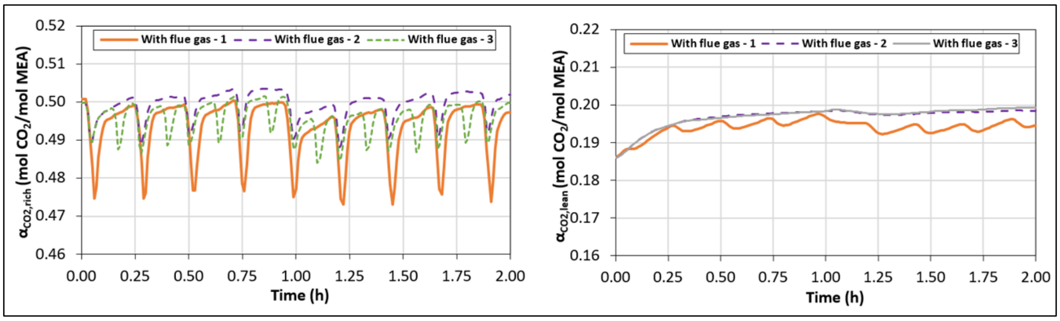

Fig. 3. Rich (left) and lean (right) CO_2 loading values when treating different fluctuating flue gases.

References

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Keywords: Post-combustion CO_2 capture; Absorption-regeneration amine-based process ; Aspen Dynamics® simulation; Fluctuating flue gases.

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