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## SO<sub>2</sub> Influence on MEA Degradation in CO<sub>2</sub> Capture Absorption-Regeneration Process: Corrosion Impact on Oxidative Degradation

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### Abstract

Carbon capture technologies have become essential in our climate mitigation strategy as atmospheric CO<sub>2</sub> concentrations continue to rise. The IPCC has identified carbon capture and storage (CCS) as indispensable for achieving the Paris Agreement's temperature stabilization targets, particularly considering the EU's 2050 carbon neutrality commitment.

Among carbon capture technologies, the absorption-regeneration one using amine-solvent is a widespread technology known for almost a century [1], even if the interest for this technology in the climate change context is above 30 years [2]. However, this technology still requires improvements, especially regarding solvent management related to the various degradation mechanisms that occur with the amine component of the solvent used. Among these solvents, one of the most well-known is monoethanolamine (MEA), typically dosed at 30% by mass in water. Numerous degradation products, as well as their mechanisms, have already been identified regarding aspects related to both thermal degradation and oxidative degradation of MEA [3], [4], [5]. In addition to this type of degradation, other pollutants present in most of the flue gases could have serious impacts on the solvent, which is the case for SO<sub>x</sub> and NO<sub>x</sub>. Indeed, NO<sub>x</sub>, reacting with amine (and especially DEA present as an impurity in MEA) will form nitrosamines and nitramines which are known carcinogens, even at very low concentrations. On the other way, SO<sub>2</sub> is known to form Heat Stable Salt (HSS) via reaction between protonated MEA and the dissolved sulfate anion SO<sub>4</sub><sup>2-</sup> [6]. Moreover, another specificity of SO<sub>x</sub> and NO<sub>x</sub> is their corrosion impact on steel equipment. Following the work of Lamprou et al. [7], NO<sub>x</sub> and SO<sub>x</sub> contaminants have particular effects on corrosion behaviors of stainless-steel in MEA solutions. This corrosion will lead to the release of metal cations in the solvent which can cause an acceleration of oxidative degradation kinetics due to the catalytic effect caused by these cations [8].

In this work, we evaluate both impacts of SO<sub>2</sub>, knowing the reaction with MEA but also its corrosion effect, to develop a kinetic model. Using the Degradation Test Rig (DTR) developed by Léonard et al. [9], different gas conditions are used. Indeed, based on a 72h test with accelerated degradation conditions, it is possible to find similar degradation

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products obtained after several months on pilot-scale structures. Accelerated conditions include temperature (120 °C), pressure (4 barg) and agitation speed (700 rpm) in a semi-batch reactor containing around 300 mL of MEA solvent. A diagram of the DTR is shown in Figure 1, where the 4 gas lines can be seen, as well as the main pieces of equipment, while pictures of the DTR are shown in Figure 2.

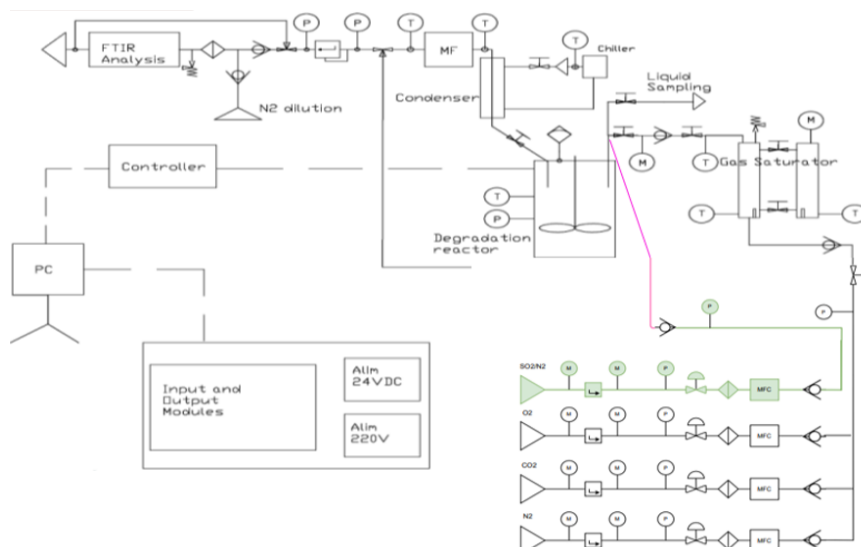


Figure 1: Diagram of the Degradation Test Rig

After 3 days of experiments, samples are analyzed through different analytical devices including HPLC-RID, GC-FID, and ICP-OES. HPLC enables the measurement of MEA concentration in different samples taken throughout the experiment, while GC enables to have the different degradation products present in the liquid sample. The ICP is used for analyzing the metal concentration of Fe, Cr, Cu, and Mn ions in the solvent, whatever their complexation state. The main identified oxidative degradation products are  $\text{NH}_3$  (gaseous emission), HEI, HEIA, HEPO and OZD (in the solvent). The impact of  $\text{SO}_2$  on those will be studied as a priority.



Figure 2: Degradation Test Rig

Based on those conditions, an initial CO<sub>2</sub>-loaded MEA solvent went from 30 %-mass. to 24 %-mass. showing the degradation of 20 % of its initial MEA content, when it is submitted to a gas mixture of 5 %-vol. O<sub>2</sub>, 15 %-vol. CO<sub>2</sub>, and 80 %-vol. N<sub>2</sub>. This concentration was measured 3 times by using HPLC and performing at the same time a calibration with precise MEA-H<sub>2</sub>O mixture with a concentration range from 0 to 30 %-mass. This degradation experiment was set as the base case for experimental planning describe below. Currently, GC-FID calibration is made for the main degradation products described above to compare with previous studies [3], [4] in order to have relevant data for studying the SO<sub>2</sub> impact. An example of GC-FID spectra completed during the PhD thesis of Léonard [4] is shown below for a base case at 400 rpm during two weeks of experimentation with the above-mentioned gas mixture.

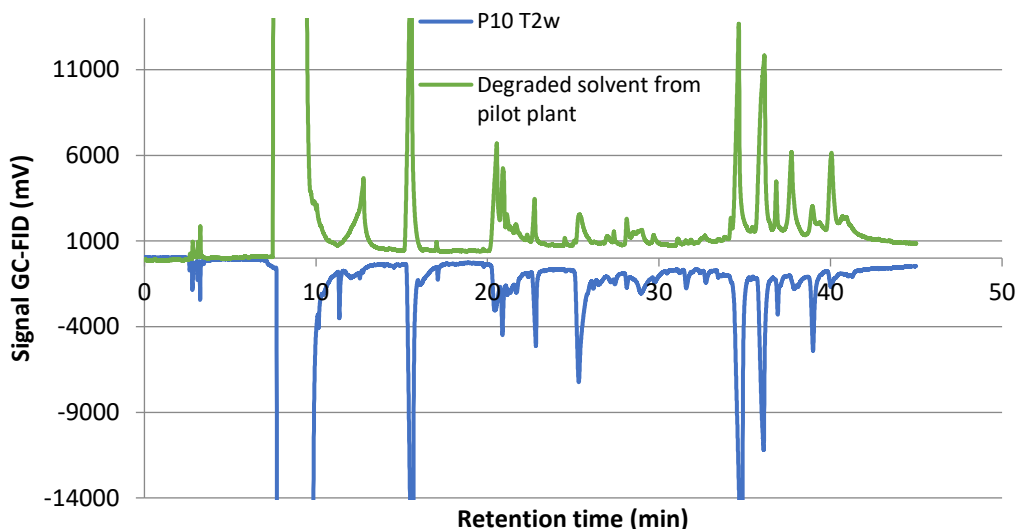


Figure 3: GC spectra from MEA degradation [4]

To analyze the corrosion effect, three similar experiments are performed first in a stainless-steel reactor (316L) and then in the same reactor with Teflon coating to lower the contact with metal. The SO<sub>2</sub> concentration is varied between 0 and 150 ppm, while the CO<sub>2</sub> and O<sub>2</sub> concentrations in the feed gas are maintained at 15% and 5% respectively, with N<sub>2</sub> used as a balance gas to reach 100%, knowing that each experiment follows the same procedure as described above.

Future work will expand these experiments across different temperatures and O<sub>2</sub> concentration to develop kinetic models that can predict MEA degradation due to SO<sub>2</sub> into simulation model for industrial CO<sub>2</sub> capture plants using amine-based absorption-regeneration processes.

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