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Application of Absorption by Potassium Taurate Solutions to Post-Combustion CO2 Removal from Flue Gases with Different Compositions and Flowrates

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Climate change issues have led many countries to actively participate to international conventions, with the aim of reducing the impact of human activities on the environment. In this framework the Paris Agreement has established to keep the temperature rise below 2°C above the pre-industrial level.

One suitable mitigation option is to remove carbon dioxide, before it is emitted into the atmosphere. Several technologies are available, including chemical processes where acid gases are absorbed by aqueous alkanolamines solutions. Although this process is already widely used in industry, these solvents are characterized by high energy requirements, in addition to other disadvantages such as corrosion and degradation.

Recently, alternative solvents are being studied as possible substitutes for traditional amine solutions. Precipitating solvents are considered in chemical absorption processes because of their characteristic of forming a solid phase, which allows the removal of one reaction product from the liquid solution, therefore shifting the equilibrium of the reaction and so enhancing the mass transfer of the CO2 from the vapor phase to the liquid phase. One class of precipitating solvents are amino acids, with several different types which can be used in aqueous solution. Previous studies have identified the most suitable types for the capture of carbon dioxide and have highlighted the additional advantages of low corrosion and high stability towards degradation, in particular for the amino acid species containing a sulfonic group, such as taurine.

This work focuses on the use of potassium taurate aqueous solutions for the removal of carbon dioxide from flue gases from natural gas and coal-fired power plants. The aim is to evaluate changes in the solvent performance arising from changes in the flue gases composition and flowrates.

The obtained results confirm the possible application of the potassium taurate aqueous solution for CO2 removal both in coal-fired and in NGCC power plants, with different energy requirements.

* 1. Introduction

The atmospheric concentration of greenhouse gases as carbon dioxide, methane, nitrous oxide and chlorofluorocarbons has increased over the last century due to human activity. It has been estimated by the Intergovernmental Panel on Climate Change (IPCC, 2005) that since industrial revolution their concentrations in the atmosphere have increased, with carbon dioxide being responsible for 50% of this increase.

One of the most relevant sources of carbon dioxide emissions is from the combustion of fossil fuels in power plants. Carbon Capture and Storage (CCS) has been identified as one of the most effective methods to mitigate from this source, because of the possibility of retrofitting already existing plants (Alie et al., 2005).

Technologies based on absorption, adsorption, membranes and other methods have been developed for separating carbon dioxide from flue gases, with amine processes being a technology commercially available from decades and widely employed (Kohl and Nielsen, 1997). However, when dealing with amine systems a high energy requirement is needed to run the plant, and issues related to corrosion and toxicity must be taken into account.

In the last years, alternative solvents based on phase transition have started to be considered as possible substitutes to traditional systems. Precipitating solvents as amino acids can be employed for chemical absorption of carbon dioxide because of the advantages associated with chemical reactions in the liquid phase. Moreover, by forming precipitate at high loadings (mol CO2 / mol amino acid), they allow a further enhancement of the mass transfer of carbon dioxide from the vapour to the liquid phases. Low corrosion (Ahn et al., 2010) and low enthalpy of reaction (Sanchez Fernandez et al., 2013) are general characteristics of this class of solvents. In addition, the separation of the slurry rich in solid phase from the liquid solution can also allow for further reduction of the energy requirement in the regeneration column.

Studies (Lerche, 2012; Majchrowicz, 2014; Majchrowicz et al., 2009) have focused on analysing on which are the most suitable amino acid-based systems for CO2 capture, determining the advantages of those containing a sulphonic group, as taurine, related also to low corrosion and high stability towards degradation (Ahn et al., 2010). An aqueous solution of taurine and potassium hydroxide (Lerche, 2012) has been determined as one of the most suitable ones, and experimental research on vapour-liquid-solid equilibrium (Kumar et al., 2003a; Kumar et al., 2003b; Sanchez Fernandez et al., 2013), kinetics (Kumar et al., 2003c) and physical properties (Han et al., 2012; Kumar et al., 2001) have been carried out.

The few studies that have evaluated CO2 removal using potassium taurate have focused on purification from flue gases of coal-fired power plants (Ho et al., 2019; Raksajati et al., 2016; Sanchez Fernandez et al., 2013). However, also CO2 removal from flue gases of natural gas-fired power units needs to be evaluated, especially since there is an increasing number of new gas fired power plants being built, coupled with the conversion of many coal to natural gas boiler in the residential and industrial sectors (IEA, 2018).

* 1. Aim of the work

This work uses ASPEN Plus® simulation to compare the applicability of potassium taurate solvent for chemical absorption of carbon dioxide from flue gases exiting from coal and natural gas fired power plants.

* 1. Case studies

The flue gas stream of a 500 MW coal-fired power plant (Moioli et al., 2018b) and the one of a Natural Gas Combined Cycle (NGCC) unit for production of 630 MW (Fout et al., 2015) have been considered in this work. The characteristics of the two streams are detailed in Table 1.

Table 1: Characteristics of the streams to be purified.

|  |  |  |
| --- | --- | --- |
| Parameter | NGCC | coal-fired |
| Flowrate [kmol/s] | 36.26 | 19.6 |
| Temperature [K] | 313 | 313 |
| Pressure [atm] | 1 | 1 |
| Component | mole fraction | mole fraction |
| Carbon dioxide | 0.0391 | 0.13 |
| Water | 0.0841 | 0.07 |
| Nitrogen | 0.7441 | 0.75 |
| Oxygen | 0.1238 | 0.05 |
| Argon | 0.0089 | 0 |

As flue gases from combustion in the presence of air, most of the existing gas is composed of nitrogen (about 74% for both the streams), with remaining oxygen and water and CO2 formed during combustion. The mole fraction of carbon dioxide (13% for the coal-fired exiting stream and 3.91% for the natural gas flue gas) is strongly dependent on the type of fuel feed employed for producing power as reported also by the Global CCS Institute (Global CCS Institute, 2013). The amount of produced carbon dioxide per generated energy is very different for the NGCC case (0.10 kgCO2/MJ) and for the coal-fired case (0.22 kgCO2/MJ). The amount of carbon dioxide to be removed influences the performance of the CO2 removal plant.

* 1. Methodology
     1. Simulation tool

The work has been carried out by employing a tool for simulation previously developed by Moioli et al. (Moioli et al., 2017) based on the commercial software ASPEN Plus®, which has been user customized for the representation of the system with potassium taurate, not present by default in the simulator (Moioli et al., 2018a). With this user-developed tool, the Vapor-Liquid-Solid Equilibrium (VLSE) is considered in the thermodynamic model and rate-based simulations, taking into account also the kinetics of the system, are performed.

* + 1. Process model

The process plant for treating the NGCC flue gas has been designed considering the very large gas flowrate. For this reason, the modelling includes three parallel absorption columns, each one treating one third of the total gas flowrate.

For both the coal-fired and NGCC flue gases, removal of 85% is set. The absorber is modelled using counter current flow based on an aqueous solution of 4M potassium hydroxide and 4M taurine. The rich solvent, in the form of slurry with some precipitated taurine, is sent to the dissolution heat exchanger for dissolution of solids and to the lean-rich heat exchanger for heat recovery, before being sent to the distillation column for regeneration and recycle of the lean regenerated solvent to the absorption section, after cooling and make-up.

Because the CO2 to be removed is at low concentration for NGCC, despite the huge flue gas flowrate, the liquid flowrate is lower and can be treated in one single regeneration column.

The same scheme has been considered in this work also for purifying flue gas from the coal-fired power plant. However, because of the different characteristics of the stream to be purified, different dimensions of the columns have been selected. It was found that to minimize the reboiler duty, a lean loading of 0.31 was required for the NGCC flue gas stream and a lean loading of 0.39 for the coal-fired one.

Table 2 reports the characteristics of the plant for the two case studies.

Table 2: Characteristics of the CO2 removal plant for purification of the two different types of flue gas stream.

|  |  |  |
| --- | --- | --- |
| Parameter | NGCC | coal-fired |
| Absorber |  |  |
| Number of units | 3 | 3 |
| Height [m] | 30 | 15 |
| Diameter [m] | 13.3 | 12.2 |
| Dissolution heat exchanger |  |  |
| Temperature [K] | 349.15 | 349.15 |
| Lean-rich heat exchanger |  |  |
| Tapproach [K] | 10 | 10 |
| Regenerator |  |  |
| Number of units | 1 | 1 |
| Height [m] | 17.6 | 17.6 |
| Diameter [m] | 8.5 | 13 |
| Lean solvent cooler |  |  |
| Temperature [K] | 313 | 313 |

* 1. Results and Discussion

The mass balance around the absorber are reported in Table 3 for NGCC power plant and in Table 4 for the coal-fired power plant.

Table 3: Characteristics of the main streams in the CO2 removal section of the NGCC power plant.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Parameter | flue gas | purified gas | lean solvent | rich solvent |
| Total flowrate [kg/s] | 1029.69 | 990.85 | 3253.19 | 3292.03 |
| Temperature [K] | 313 | 330.28 | 313 | 329.99 |
| Pressure [atm] | 1 | 1 | 1 | 1 |
| Fluid phase | mole fraction | mole fraction | mole fraction | mole fraction |
| Carbon dioxide | 0.0391 | 0.0059 | 0.0324 | 0.0452 |
| Water | 0.0841 | 0.1073 | 0.7589 | 0.7543 |
| Taurine | 0 | 0.0000 | 0.1044 | 0.0955 |
| KOH | 0 | 0.0000 | 0.1044 | 0.1049 |
| Nitrogen | 0.7442 | 0.7616 | 0.0000 | 0.0000 |
| Oxygen | 0.1238 | 0.1252 | 0.0000 | 0.0000 |
| Argon | 0.0089 | 0.0000 | 0.0000 | 0.0000 |
| Solid phase | weight fraction | weight fraction | weight fraction | weight fraction |
| Taurine | NA | NA | 0.0000 | 0.0338 |

Table 4: Characteristics of the main streams in the CO2 removal section of the coal-fired power plant.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Parameter | flue gas | purified gas | lean solvent | rich solvent |
| Total flowrate [kg/s] | 580.29 | 483.84 | 9943.96 | 10040.41 |
| Temperature [K] | 313 | 318.02 | 313 | 324.63 |
| Pressure [atm] | 1 | 1 | 1 | 1 |
| Fluid phase | mole fraction | mole fraction | mole fraction | mole fraction |
| Carbon dioxide | 0.13 | 0.0220 | 0.0404 | 0.0482 |
| Water | 0.07 | 0.0756 | 0.7526 | 0.7589 |
| Taurine | 0 | 0.0000 | 0.1035 | 0.0886 |
| KOH | 0 | 0.0000 | 0.1035 | 0.1043 |
| Nitrogen | 0.75 | 0.8460 | 0.0000 | 0.0000 |
| Oxygen | 0.05 | 0.0564 | 0.0000 | 0.0000 |
| Argon | 0 | 0.0000 | 0.0000 | 0.0000 |
| Solid phase | weight fraction | weight fraction | weight fraction | weight fraction |
| Taurine | NA | NA | 0.0000 | 0.0566 |

Although the flue gas flowrate in the coal-fired power plant is smaller than for the NGCC (19.6 kmol/s instead of 36.26 kmol/s), the much higher carbon dioxide concentration of 13%, compared to 3.91% for NGCC, results in 2.1658 kmol/s of carbon dioxide being captured, compared with 1.2051 kmol/s for NGCC. This is because of the fixed CO2 removal, considered equal to 85% for both the cases.

The lower amount of carbon dioxide in the stream from the NGCC makes the separation more difficult. To achieve the target of 85% CO2 removal, very tall absorption columns (Table 2) and high energy consumption per unit of carbon dioxide removed (Figure 1) are needed. As for the overall reboiler duty, however, since the coal-fired power plant produces higher amounts of carbon dioxide, the needed total circulating flowrate and, consequently, the total energy requirement (303 MWth compared to 215 MWth for the NGCC) are higher (Table 4).



Figure 1: Comparison between the CO2 removal section of the NGCC and the coal-fired power plants considered in this study on the basis of the total reboiler duty and specific reboiler duty per kg of absorbed CO2.

Precipitation of solid taurine was found to occur for both the flue gas streams, but with the rich solvent in the NGCC plant being characterized by a lower amount of solid taurine (w = 0.0338) than the one of the rich solvent in the coal-fired plant (w = 0.0566) (Table 3 and Table 4). This is related to the composition of the rich solvent in the liquid phase, which influences the solubility of the taurine. The higher the amount of absorbed carbon dioxide (higher the CO2 molar fraction), the higher the amount of solid taurine that is formed. Because of the precipitation of pure taurine, the resulting mole fraction of potassium hydroxide and of taurine in the liquid phase for the two flue gases is different.



Figure 2: Comparison between the CO2 removal section of the NGCC and the coal-fired power plants considered in this study on the basis of column dimensions (for the absorber, the dimension of the single absorber is reported).

The required volume of packing in the regeneration column was found to be also influenced by the different concentrations and molar flow of carbon dioxide in the flue gas streams. Figure 2 shows that for the NGCC flue gas, the regenerator packed volume is less than half (43%) of the one needed for the coal-fired power plant, while the packed volume for the absorber is three times larger.

* 1. Conclusions

Coal is the primary focus of most of the CO2 capture activities and it is expected to be widely employed in power generation for many decades to come. Nevertheless, natural gas, the most clean fossil fuel, is increasingly becoming used, with many conversions of coal boilers to gas ones. Though CO2 emissions from gas are lower per kilowatt of power generated compared to those from coal–fired power plants, carbon dioxide is still produced and must be removed in order to achieve a low carbon economy.

Precipitating solvents such as potassium taurate have started to be considered for use in the CCS sector for reduction of CO2 emissions because of their advantages over traditional amines.

In this work, simulations with a user-developed tool have been carried out to evaluate whether the process may be suitable for application to different types of flue gases from power plants (NGCC and coal-fired units). The results show that the technology may be applied to both the types of power plants, and precipitation of the solids occurs even at low concentrations of carbon dioxide such as those found in NGCC flue gas streams. The needed power is different for removing carbon dioxide from a flue gas stream of NGCC power plant or of a coal-fired power plant.

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