



The environmental impact of post-combustion CO₂ capture with MEA, with aqueous ammonia, and with an aqueous ammonia-ethanol mixture for a coal-fired power plant

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ARTICLE INFO

Article history:

Received 18 February 2010

Received in revised form

7 December 2010

Accepted 23 December 2010

Available online 3 February 2011

Keywords:

LCA

Ammonia

Post-combustion

CO₂

Capture

Aspen plus

ABSTRACT

In this paper the authors compare monoethanolamine (MEA) to aqueous ammonia (AA) and a solvent mixture of aqueous ammonia and ethanol (EAA) with respect to their post-combustion CO₂ capture performance and their environmental impact. Simulation of all processes was carried out with Aspen Plus[®] and compared to experimental results for CO₂ scrubbing with ammonia. Of special interest was the formation of stable salts, which could be observed in the experimental CO₂ capture with both ammonia solvents. If CO₂ can be captured in the form of ammonium salts, energy requirements are greatly reduced, since no energy is required for solvent regeneration and CO₂ compression. The environmental impact of CO₂ capture was investigated for a 500 MW pulverised coal power plant employing Life Cycle Assessment (LCA) using the software SimaPro[®]. For a comprehensive evaluation of this impact, influencing factors such as solvent production and solvent emissions were included. With kinetics taken into account, no salt formation could be observed in CO₂ removal with aqueous ammonia. The necessary reduction of ammonia emissions leads to further energy requirements, and solvent production as well as the remaining ammonia losses to the environment have a more significant environmental impact than CO₂ removal with MEA.

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1. Introduction

The still rising demand for energy and the increase in CO₂ emissions typically involved has become one of the most important environmental topics. Coal is a resource still readily available. Its use in power generation comes with a cost, however, since its combustion releases a high amount of CO₂. In 2006, 41% of the global electricity was produced with coal as fuel. Even if this percentage is to half down to 21% according to the 450 ppm CO₂ mitigation scenario of the IEA, coal will remain one of the principal sources for electricity generation [1].

In order to reduce the environmental impact of power generation from fossil fuels, the emission of greenhouse gases such as CO₂ needs to be reduced. The most mature technology of CO₂ absorption in gas purification processes uses amines such as MEA as absorbent [2]. This technology can be applied to post-combustion

CO₂ capture, which is used to remove the CO₂ from the flue gas stream. Post-combustion capture has the advantage that it can also be applied to retrofit existing power plants. Due to high economic and efficiency penalties this technology is still not applied to CO₂ absorption in commercial power plants.

A novel method of reducing CO₂ emissions from power plants is the use of an aqueous ammonia solution [3,4] to capture the CO₂ as a valuable solid product [5,6]. Thus we focused our research also on the capture of CO₂ as an ammonium salt, as this allows for a much more rational use of the CO₂ than by compression and necessary storage, if the carbon dioxide is captured in gaseous form. Therefore a solution of ethanol, ammonia and water (EAA) was investigated. Using ethanol helps reduce the solubility of the ammonium salts that can then be obtained by filtration or sedimentation. The produced salts are a basic ingredient of fertilisers [7] and could hence be exploited commercially. NH₃ has frequently been used as reactant for the De-NO_x process in power plants, and it is also interesting for the removal of SO₂ and HCl from flue gases [8].

In this paper the authors compare the primary amine MEA to aqueous ammonia and a solvent mixture of aqueous ammonia and ethanol, with respect to their CO₂ capture performance.

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Table 1
Power plant parameters.

Coal feed	50.4 kg/s		
HHV coal	27060 kJ/kg		
Net efficiency (HHV)	41.4%		
HP turbine	$p_{in} = 152$ bar	$T_{in} = 533$ °C	$P_{el} = 130.7$ MW
MP turbine	$p_{in} = 42.5$ bar		$P_{el} = 190.1$ MW
LP turbine	$p_{in} = 4.67$ bar		$P_{el} = 243.7$ MW
Condenser	$p_{cond} = 0.0068$ bar		$Q_{cooling} = 265$ MW _{th}

Life Cycle Assessment is a method for the determination of the environmental impact of all substances used and emissions caused by a product or activity during its entire lifetime or Life Cycle.

After the definition of goal and scope of the study as a first step, the next step in a Life Cycle Analysis is the inventory of all the materials included in assembly, operation, and maintenance, and of the generated wastes. It is also important to include avoided materials such as the avoided emission of greenhouse gases. Last but not least, the location of the investigated process can be very important. In the consecutive impact assessment step, the impacts caused by the use of the involved materials or their emission are evaluated.

2. Process simulations

All simulations were carried out with Aspen Plus® [9], which has become a widely used standard application for computerised flowsheet simulations in the chemical industry.

2.1. Pulverised coal power plant

For all capture processes we assumed the flue gas stream to exit from a pulverised coal power plant, which has a maximum net output of 565 MW_{el}, if no CO₂ capture is applied. The maximum net output of the power plant with solvent based post-combustion capture is in the range of 400 MW_{el}. The relevant parameters of the Rankine cycle power plant [10] can be found in Table 1, and the composition of the flue gas stream is presented in Table 2.

For the thermal solvent regeneration in a stripper column, steam is extracted after the medium pressure turbines.

2.2. Equilibrium CO₂ capture model with ammonia and experiments

The energy penalty for post-combustion CO₂ capture with amines is currently in the range of 4–5 GJ/t CO_{2,captured} [11]. This penalty is mainly due to the input of thermal energy for the regeneration of the solvent in the traditional absorber-desorber configuration (see Fig. 4). Energy requirements are reduced by about 75% in the salt-forming process with ammonia, since heat is only needed for the regeneration of the water in a secondary gas treatment for the reduction of ammonia emissions. Furthermore, the separated CO₂ does not have to be compressed.

CO₂ scrubbing with aqueous ammonia is economically very attractive [12], especially if salts can be separated, since the conventional production of the ammonium salts requires an energy input of around 32 GJ/t salt [13], while the ammonia supply for this

Table 2
Flue gas parameters.

	N ₂	H ₂ O	CO ₂	O ₂	CO	NO	SO ₂	Total flow	Temperature	Pressure
% mole	74.0	7.0	13.8	3.5	0.2	0.4	0.07	550 kg/s	300 °C	1 atm

Table 3
Reactions for the CO₂–NH₃–H₂O system.

$2\text{H}_2\text{O} \leftrightarrow \text{H}_3\text{O}^+ + \text{OH}^-$ (1)
$\text{CO}_2 + 2\text{H}_2\text{O} \leftrightarrow \text{H}_3\text{O}^+ + \text{HCO}_3^-$ (2)
$\text{HCO}_3^- + \text{H}_2\text{O} \leftrightarrow \text{H}_3\text{O}^+ + \text{CO}_3^{2-}$ (3)
$\text{NH}_3 + \text{H}_2\text{O} \leftrightarrow \text{NH}_4^+ + \text{OH}^-$ (4)
$\text{NH}_3 + \text{HCO}_3^- \leftrightarrow \text{NH}_2\text{COO}^- + \text{H}_2\text{O}$ (5)
$\text{NH}_4\text{HCO}_3 \xrightarrow{\text{SALT}} \text{NH}_4^+ + \text{HCO}_3^-$ (6)
$(\text{NH}_4)_2\text{CO}_3 \xrightarrow{\text{SALT}} 2\text{NH}_3 + \text{CO}_2 + \text{H}_2\text{O}$ (7)
$\text{NH}_2\text{COONH}_4 \xrightarrow{\text{SALT}} \text{NH}_4^+ + \text{NH}_2\text{COO}^-$ (8)
$\text{CO}_2 + \text{OH}^- \rightarrow \text{HCO}_3^-$ (9)
$\text{HCO}_3^- \rightarrow \text{CO}_2 + \text{OH}^-$ (10)

process is produced with an energy input of around 19.4 GJ/t NH₃ [14]. Table 3 shows the reactions used for the simulation of CO₂ absorption with aqueous ammonia. The important carbamate formation reaction, Reaction (5), was described to proceed very fast by a so-called shuttle mechanism [14] via a zwitterionic intermediate. Reactions (9) and (10) are slower than the other reactions considered here. Therefore, the kinetic parameters [15] of these reactions are important for a more accurate simulation of the capture process.

Due to the presence of ionic species in these volatile multi-component weak electrolyte systems [16,17], liquid and vapour properties were computed by the electrolyte NRTL (Non-Random Two Liquid) method, an activity-coefficient based model. Henry's law was used to calculate the solubility of CO₂ in water.

The model configuration for the simulation of CO₂ scrubbing with aqueous ammonia (solvent concentration 5% wt., solvent temperature 20 °C) included a packed absorber column and a filter for the extraction of salt products (see Fig. 1).

Subsequently, we investigated the CO₂ removal with ammonia by a series of experiments carried out in a small-scale setup. The CO₂ capture performance of the system was easily determined by weight measurements of all system components before the tests and after certain time intervals. The pH-value and the species in solution were analysed with a ¹³C NMR (Nuclear Magnetic Resonance spectroscopy) apparatus.

In order to separate the ammonium salts by sedimentation or by filtration it is necessary to lower their solubility. Therefore, temperatures down to –5 °C were investigated, which were achieved by immersion of the container in a temperature controlled coolant. A reduction in salt solubility can also be achieved by either raising the concentration of ammonia in the solvent stream [18,19] or by using an alternative solvent. In this work, the latter option was chosen, as otherwise ammonia losses with the exhaust gas become very large. Aiming at a reduction of these losses, the authors then investigated an Ethanol-Aqueous Ammonia (EAA) solution [6].

Ethanol was chosen due to its good miscibility with water, a lower freezing point of the resulting solution, and its good availability. A new series of tests was carried out for various ammonia concentrations and absorber temperatures without changing the described experimental setup. Using the EAA solution, only at lower temperatures could crystalline salts be obtained to a significant amount (see Fig. 2). The measurements are effected by an experimental error. The precision of the balance used to weigh all components is 0.01 g, corresponding to an error of ca. 1%. The accuracy of the gas chromatograph, used to measure the losses of CO₂ from the system, is ca. 5% around the value of calibration (a flow containing 10% vol. CO₂ and 90% vol. N₂ produced by Italian gas company Rivoira). This leads to an error of up to 4% in mass of CO₂ lost, depending on the total amount of CO₂ that has left the system.

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