Carbon Dioxide Capture using Aqueous Ammonia

I. Introduction

The project involves a complete detailed design of a carbon-capture process using post-combustion absorption with aqueous ammonia to remove 90% of the carbon dioxide in the flue gas a power plant.

The design basis was for a power plant with a net output of 550 MWe and an exhaust gas composition of 13.50 mol% CO_2, 15.17 mol% H_2O, 68.08 mol% N_2, 2.43 mol% O_2, and 0.82 mol% argon at a temperature of 57.2°C and 103 kPa. The capture process must extract 90% of the CO_2 from the inlet flue gas and the CO_2 outlet stream must be supercritical. Additionally, due to environmental regulations, the treated flue must contain less than 150 ppm (wt/wt) of ammonia. The criteria for economic analysis were a 20-year time period and a 5% interest rate with a current cost of electricity of $0.06/kWh. A design that minimizes the cost of electricity is required.

II. Background

II.A. Absorption Using Aqueous Ammonia

II.A.1. Overview of Process

The ammonia absorption process for the removal of carbon dioxide from flue gas has been reported to be very effective. Any ammonia absorption process has the same topology as seen in Figure 1.

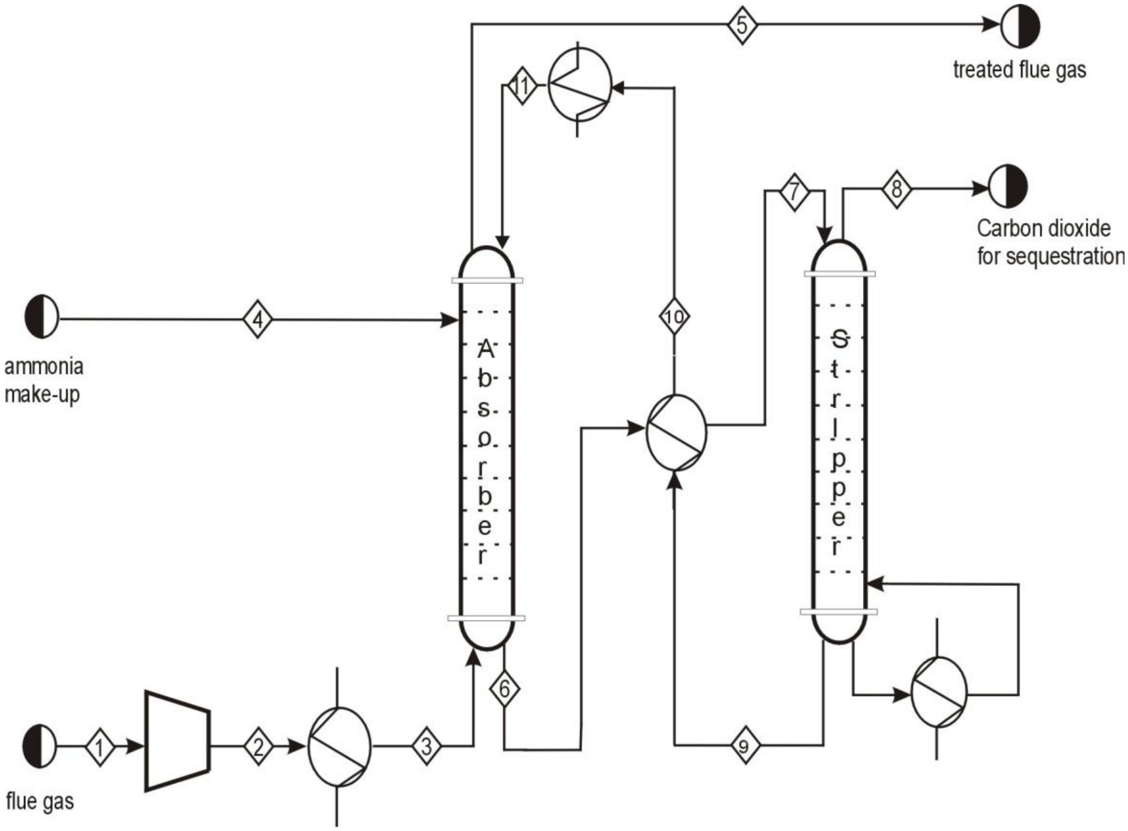

Figure 1: Block Flow Diagram (BFD) of an aquous ammonia absorption process

The system operates using an absorber to capture the carbon dioxide into a liquid stream followed by a stripping unit to regenerate it as gas. First, the flue gas (Stream 1) is compressed enough to overcome the pressure drop in the subsequent heat exchanger. This flue gas cooler uses refrigerated water to cool the flue gas to a suitable temperature for absorption. This temperature is critical for controlling the exothermic reactions in the absorber (Sherrick et al). The cooled flue gas enters an absorption tower that flows countercurrently to (Stream 3) a liquid solution of ammonia in water (Stream 11). At this low temperature, the carbon dioxide has an affinity for liquid ammonia and water at this low temperature, causing carbon dioxide from the flue gas to absorb into the liquid stream (Stream 6) (Sherrick et al, Kozak et al 2009). The outlet streams from the absorber are CO_2-lean flue gas (Stream 5) and CO_2-rich aqueous ammonia

(Stream 6). The CO_2-lean flue gas with at least 90% of the CO_2 removed is then vented to the atmosphere after possible additional treatment to remove any ammonia that has escaped in the absorber.

The heated CO_2-rich stream (Stream 7) is fed at the top stage of a stripper (Sherrick et al) for the separation of carbon dioxide from the aqueous ammonia. A reboiler is used in the stripper to provide the energy needed for the salts formed to dissociate and for the carbon dioxide to escape to the gas phase. The carbon dioxide exits the stripper (Stream 8) and is subsequently compressed to a pressure suitable for sequestration. The liquid product of the stripper (Stream 9) is the CO_2-lean aqueous ammonia stream that is used as the liquid feed to the absorber.

The liquid outlet from the heated stripper (Stream 9) is cooled before entering the absorber in an integrated heat exchanger that also heats the cool absorber outlet (Stream 6), which is to be sent to the stripper. An additional cooling exchanger is required to lower the aqueous ammonia temperature to that required for the absorber. Finally, the amount of ammonia and water that has escaped to the treated gas and CO_2 product streams is added to the liquid recycle (Stream 4) to the absorber as a make-up ammonia stream to allow the process to run continuously.

II.A.2. Reactions in Absorber and Stripper

Chemical reactions of carbon dioxide in aqueous solution are shown in Equations 1-4 (Yeh 2004).

$$CO_2(g) + H_2O(l) \leftrightarrow CO_2 \cdot H_2O(l) \tag{1}$$

$$CO_2 \cdot H_2O(l) + OH^- \leftrightarrow HCO_3^- + H_2O \tag{2}$$

$$HCO_3^- + OH^- \leftrightarrow CO_3^{2-} + H_2O \tag{3}$$

$$CO_3^{2-} + H^+ \leftrightarrow HCO_3^- \tag{4}$$

Interactions of ammonia with aqueous solution are shown in Equations 5 and 6 (Yeh 2004).

$$NH_4^+ + OH^- \leftrightarrow NH_3 \cdot H_2O \tag{5}$$

$$NH_3 \cdot H_2O \leftrightarrow NH_3(g) + H_2O \tag{6}$$

The absorber and stripper work in tandem by reactively absorbing and desorbing carbon dioxide to and from salts in the aqueous ammonia solution. CO_2 is combined into the salts in the absorber, which are then thermally decomposed to release the CO_2 in the regenerator. Reactions involving the formation of these salts are shown in Equations 7-11 (Yeh 2004).

$$HCO_3^- + NH_4^+ \leftrightarrow NH_4HCO_3(l) \tag{7}$$

$$NH_4HCO_3(l) \leftrightarrow NH_4HCO_3(s) \tag{8}$$

$$CO_3^- + 2NH_4^+ \leftrightarrow [(NH]_4)_2CO_3(l) \tag{9}$$

$$[(NH]_4)_2CO_3(l) \leftrightarrow [(NH]_4)_2CO_3(s) \tag{10}$$

$$[(NH]_4)_2CO_3 + CO_2 + H_2O \leftrightarrow 2NH_4HCO_3 \tag{11}$$

Potential reactions describing the release of CO_2 in the regenerator and the corresponding heats of formation are shown in Equations 12-14 (Yeh 2004).

$$2NH_4HCO_3(aq) \leftrightarrow [(NH]_4)_2CO_3(aq) + CO_2(g) + H_2O \quad \Delta H_{rx} = \frac{6.4kcal}{mol} \tag{12}$$

$$NH_4HCO_3(aq) \leftrightarrow NH_3(aq) + H_2O + CO_2(g) \quad \Delta H_{rx} = \frac{15.3kcal}{mol} \tag{13}$$

$$[(NH]_4)_2CO_3(aq) \leftrightarrow 2NH_3(aq) + H_2O + CO_2(g) \quad \Delta H_{rx} = \frac{24.1kcal}{mol} \tag{14}$$

It can be seen that Equation 13 has the lowest enthalpy of dissociation to release CO_2. Therefore, it may be more economical to decompose ammonium bicarbonate in the stripper and circulate the resulting ammonium carbonate back to the absorber for CO_2 capture as shown in Equation 12. Running the system in such a way would cause the CO_2-loading capacity to decrease sharply after the initial start-up cycle before stabilizing at a constant value for all subsequent cycles (Yeh 2004). The benefit of fully reacting all of the ammonium bicarbonate and ammonium carbonate to restore loading capacity completely is a smaller circulating aqueous stream, but the drawback is an increased energy requirement to obtain the complete CO_2 release. These economic concerns must be balanced to determine the appropriate regenerator design and operating conditions that will yield the desired degree of CO_2 recovery.

III.A.3. Advantages of Absorption Using Aqueous Ammonia

When compared to the more commonly used MEA, aqueous ammonia has many advantages. Aqueous ammonia operates at lower temperatures than MEA. The lower the temperature the less chance of ammonia evaporating and being discharged into the atmosphere and the lower the volatility. This process also results in higher removal efficiency and a greater CO_2 loading capacity. (Yeh 1999) Ammonia reacts much faster than MEA, allowing for a wider operating range. The energy consumption for regeneration is also less than that required by MEA, reducing operating costs. Finally, aqueous ammonia eliminates corrosion of equipment and solvent degradation, since NH_3 reacts with SO_x and NO_x to form by-products. These by-products may be sold as fertilizer.

III. Results

An optimized design for post-combustion carbon capture using aqueous ammonia has been completed, and implementation of this design, Unit 100, is expected to result in a total cost of electricity of $0.1045/kWh. For this design, a basis of $0.06/kWh was used as the current price of electricity produced from a 550 MW power plant. Using some of the generated electricity for pumps and compressor drives, electricity for refrigeration cycles for refrigerated water and low-temperature refrigerant, and low-pressure steam from the existing power plant increases the cost of electricity by $0.017/kWh, and capital costs, using a 20-year time period and 5% after-tax interest rate, raw material costs, and additional utility costs add an additional $0.027/kWh. The final power plant capacity is 428 MW. A process flow diagram of Unit 100 is shown in Figure 2.

Implementation of Unit 100 is expected to remove 98.5% by weight of CO_2 from the flue gas of a power plant with a CO_2 product purity of 99.9 mol%. It is also expected to release about 200 kg/h of ammonia vapor into the atmosphere at a concentration of about 125 ppm by weight in the treated flue gas. The overall design is considered conservative.

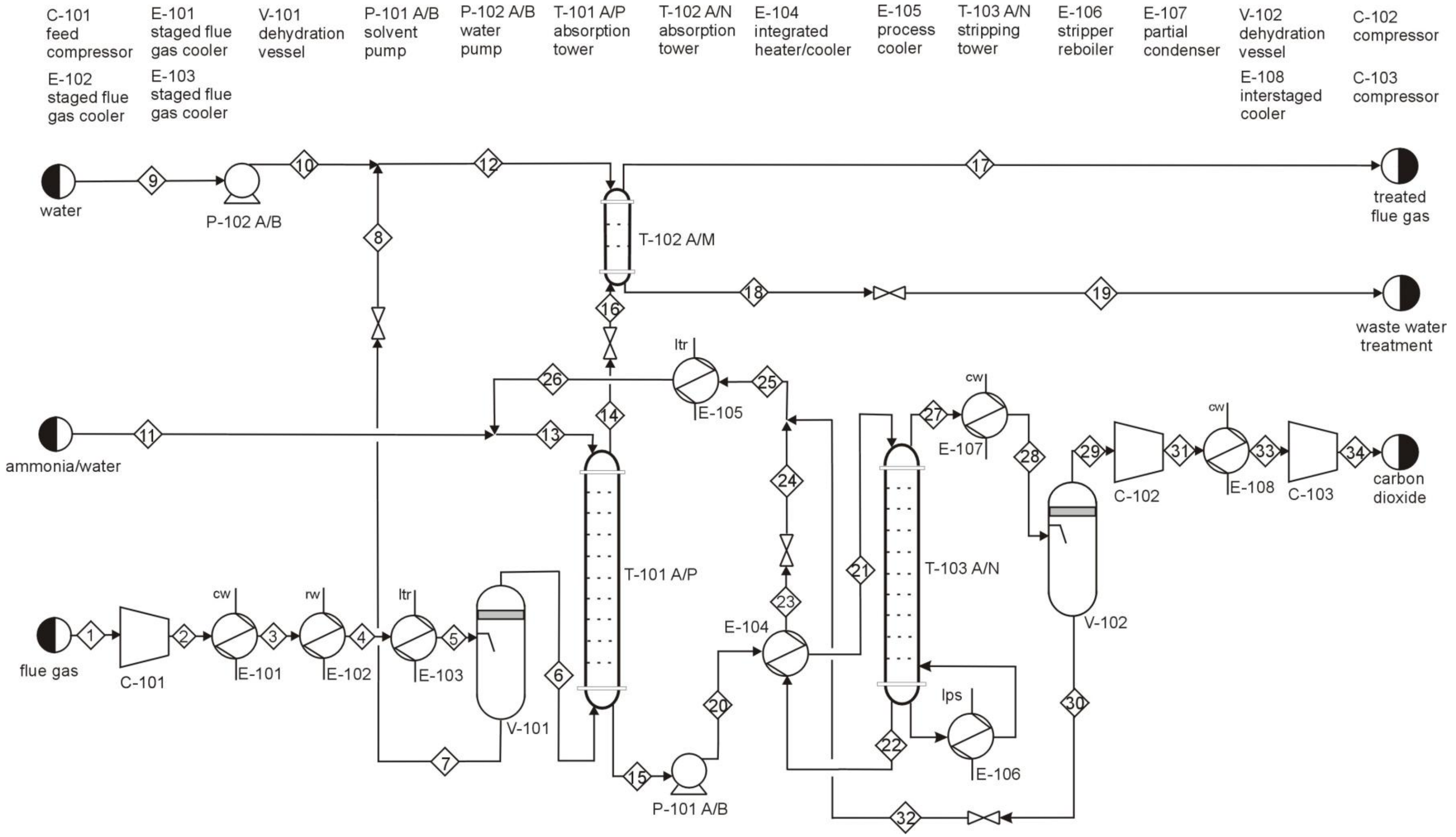

Figure 2: Process Flow Diagram (PFD) for Unit 100

IV. References

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