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Life cycle emissions of greenhouse gas for ammonia scrubbing technology

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Abstract−It is thought that the CO_2 emissions from coal-fired power plants contribute greatly to the total anthropogenic CO_2 emissions. Ammonia solvent can be used to absorb the CO_2, called ammonia scrubbing. However, as has been pointed out, the production of ammonia would emit CO_2; therefore, the effectiveness of ammonia scrubbing is doubted. The paper focuses on the problem. Two systems are defined in the paper. System I is CO_2 absorption by ammonia scrubbing, and system II is industrial production of ammonium bicarbonate. The total CO_2 emissions of the two systems are calculated by means of life cycle assessment. The paper shows that the total CO_2 emissions of ammonia scrubbing are less than that of the industrial production of fertilizer ammonium bicarbonate. It can be concluded that ammonia scrubbing is an effective way to reduce the anthropogenic CO_2 emissions.

Key words: Ammonia Scrubbing, CO_2 Emissions, Life Cycle Assessment, Coal-fired Power Plant, Ammonia Bicarbonate

INTRODUCTION

The CO_2 emissions of China are the second largest in the world, to which the combustion of coal contributes about 85%. Some strategies have been produced to control the CO_2 emissions to the atmosphere. These strategies include energy conservation, improvement of energy efficiency, and utilization of renewable energies [1]. However, as the impact of these strategies is limited, it is very important to remove CO_2 from flue gas of coal combustion. Generally, there are six kinds of technologies: chemical solvent absorption, physical adsorption, cryogenic separation, membrane separation, biological fixation as well as the O_2/CO_2 process. Amongst these techniques, the chemical solvent absorption method is considered as a reliable and relatively low cost method to reduce CO_2 from fossil fuel power plants [2]. Ammonia scrubbing is one of chemical solvent absorption methods, which is widely used in the fertilizer industry, is proven experimentally to be more effective than MEA scrubbing that is so far the most acceptable method [3]. However, the production of ammonia will produce CO_2 also. So the effectiveness of ammonia scrubbing has been doubted. The paper focuses on the problem. The following part will give an introduction of ammonia scrubbing technology. Then two systems are defined: one is the ammonia scrubbing system; the other is the industry production of the fertilizer ammonium bicarbonate. Based on the systems, life cycle CO_2 emissions are evaluated.

AMMONIA SCRUBBING TECHNOLOGY

Ammonia scrubbing technology involves spraying aqueous ammonia into the flue gas of a power plant to capture CO_2. The NH_3 will react with CO_2 to produce ammonium bicarbonate [4,5]. The gas-liquid chemical reactions can be expressed by the following reaction equation:

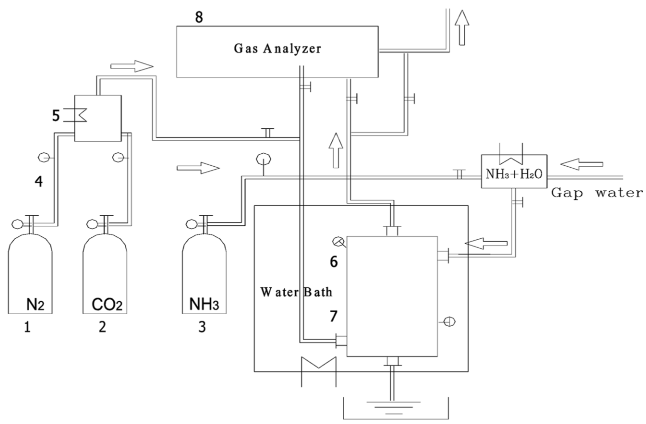

Fig. 1. Eexperimental setup of ammonia scrubbing.

1. N_2 Cylinder	5. Electronic heater
2. CO_2 Cylinder	6. Temperature controller
3. NH_3 Cylinder	7. Reaction column
4. Mass flow controller	8. Gas analyzer

$$NH_3+CO_2+H_2O \Leftrightarrow NH_4HCO_3 \quad (1)$$

Experimental study on ammonia scrubbing was done at the Department of Thermal Engineering, Tsinghua University. The experimental setup is shown in Fig. 1. The reactor, which was made of stainless steel with inside diameter of 250 mm and height of 450 mm, was installed in a temperature controlled water bath. The main part of the experimental system is composed of a sieve-plate tower, which is gas-stream reverse flow. Its schematic diagram is shown in Fig. 2. Five layers of sieve plates, which were made of stainless steel with thickness of 3 mm and bore diameter of 3 mm, are set in the reactor in order to make CO_2 and the NH_3 solution react thoroughly.

Studies on CO_2 removal by ammonia scrubbing have been per-

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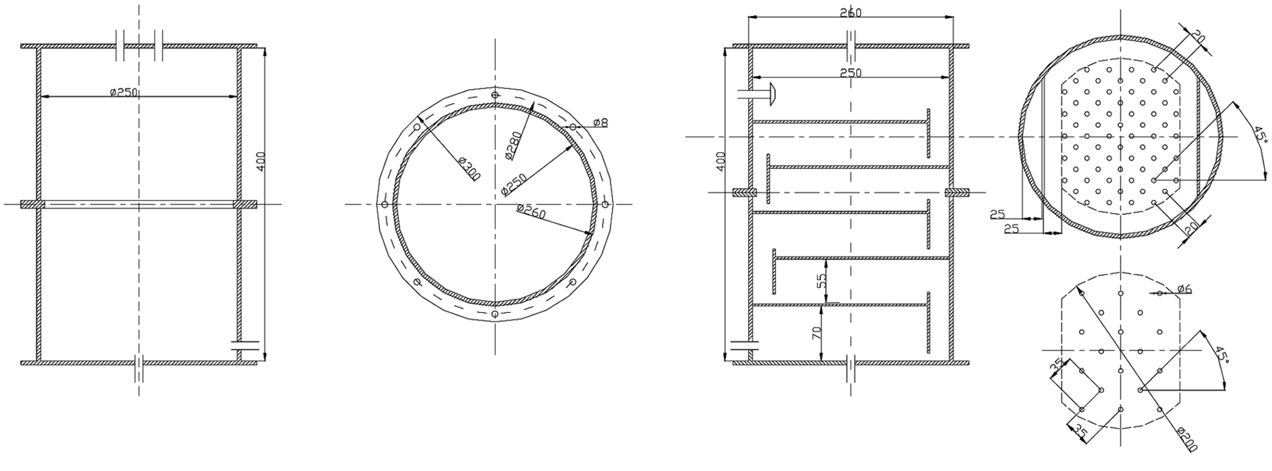

Fig. 2. The structure and parameters of the absorption tower.

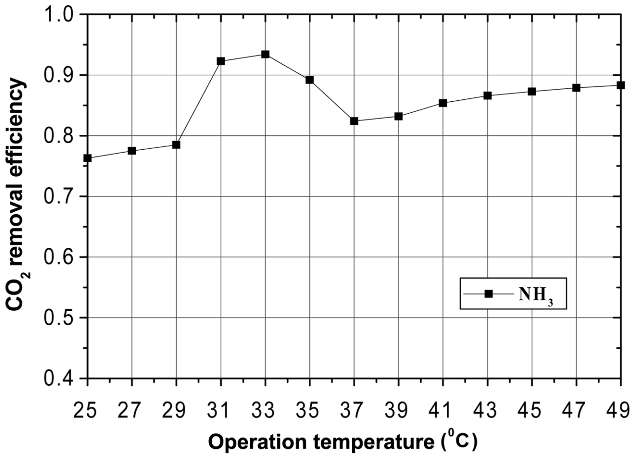

Fig. 3. CO_2 removal efficiency as a function of operation temperature. The operating conditions were at CO_2 inlet concentration of 12% (v/v), and ammonia concentration of 0.066 mol/*l*.

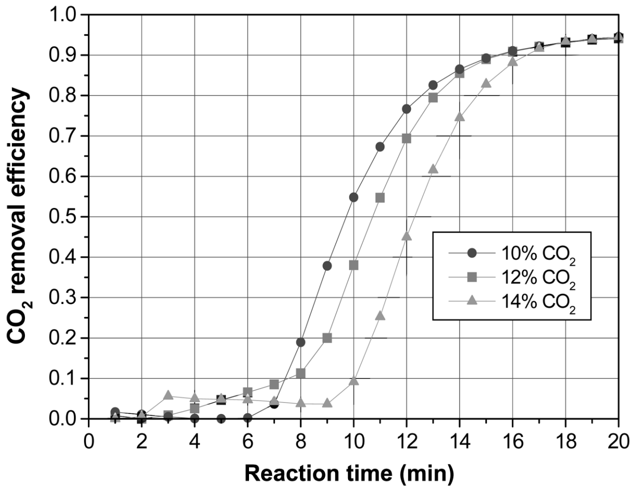

Fig. 4. CO_2 removal efficiency as a function of reaction time for various CO_2 concentrations. The operating conditions were at water bath temperature of 28 °C, and ammonia concentration of 0.140 mol/*l*.

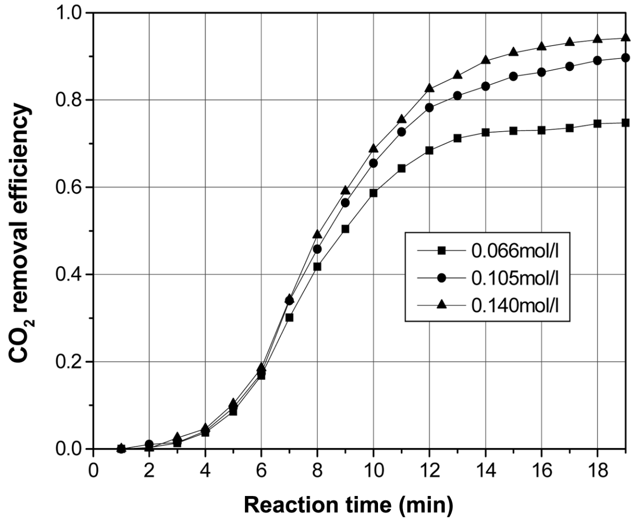

Fig. 5. CO_2 removal efficiency as a function of reaction time for various ammonia solution concentrations. The operating conditions were at CO_2 inlet concentration of 12% (v/v), water bath temperature of 28 °C.

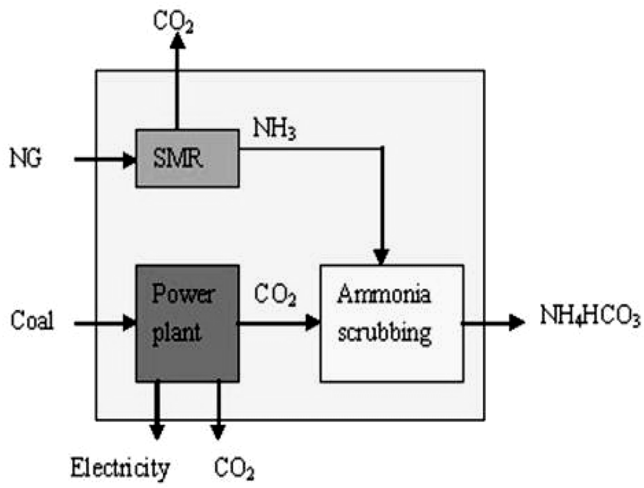

Fig. 6. Schematic of system I.

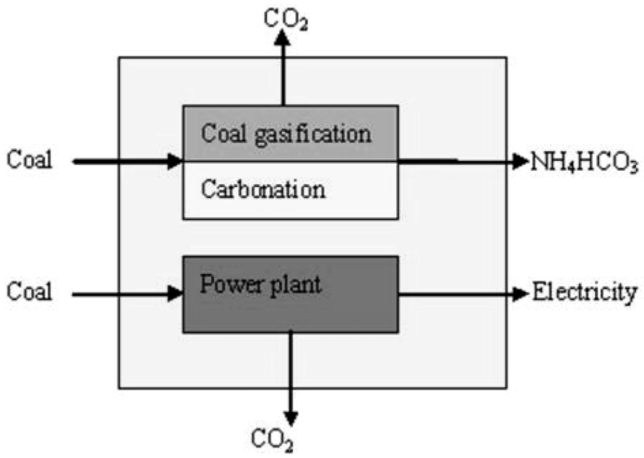

Fig. 7. Schematic of system II.

formed in an open continuous flow reactor. The temperature of the ammonia solution was designed to be in the range from 25 to 55 °C. The inlet concentrations of CO_2 were 10%, 12% and 14% (v/v), and the ammonia solution concentrations were 0.066, 0.105 and 0.140 mol/*l*, respectively. Some results can be seen in Figs. 3, 4, and 5. The overall CO_2 removal efficiencies in this study can be above 95% when the reaction temperature is about 33 °C. It is proved that ammonium bicarbonate is the main product of the CO_2-NH_3 reaction in this study.

Although ammonium bicarbonate is a cheap and widely used fertilizer in China, it is easily decomposed. Lately, some researchers have made great progress in the area of long effect ammonium bicarbonate, so ammonium bicarbonate is promising to be a fine fertilizer. If the need for large amounts of ammonia is not satisfied, the crystalline products can be heated to release CO_2 and NH_3, as shown in Eq. (2).

$$NH_4HCO_3 \Leftrightarrow NH_3 + CO_2 + H_2O \quad (2)$$

CO_2 and NH_3 can be segregated to have sequestration ready CO_2, and the ammonia solution is thus recycled and used as the scrubbing agent.

SYSTEM BOUNDARY SETTING

Two systems are defined in order to compare the life cycle CO_2 emissions. System I is CO_2 absorption by ammonia scrubbing, which includes three parts: ammonia production by steam methane reform (SMR), coal-fired power plant with installed capacity 300 MW, and CO_2 absorption by ammonia scrubbing with the byproduct ammonium bicarbonate. System II is industry production of ammonium bicarbonate, which includes three parts: syngas production by coal gasification, ammonium bicarbonate production by carbonation, and coal-fired power plant with installed capacity 300 MW.

One point needing to be explained is the reason that coal is used to produce ammonium bicarbonate through gasification and carbonation, not natural gas. Coal gasification can produce enough CO_2 that is used to produce ammonium bicarbonate. However, CO_2 must be produced in some other way if SMR is used because SMR cannot afford enough CO_2 for production of ammonium bicarbonate. For simplification, coal gasification is used in system II.

The function unit is the yearly yield of ammonium bicarbonate with ammonia scrubbing to absorb the yearly CO_2 emissions of 300 MW coal-fired power plant, as well as the yearly electricity generated by the power plant.

LIFE CYCLE CO_2 EMISSIONS

1. General Information for Calculation

The coal-fired power plant is assumed to operate at rate capacity for 7,000 hours per year. Therefore the total electricity generated is about 2.1×10^9 kWh per year. In China, the CO_2 emission factor for coal-fired power plant is about 0.8 kg/kWh. Then the annual CO_2 emissions are about 1.68×10^6 tonnes. The CO_2 removal efficiency of ammonia scrubbing is about 95%. It is assumed that the product is fully ammonium bicarbonate. Then the annual ammonium bicarbonate can be as large as 2.87×10^6 tonnes.

Therefore, the unit function can be described as electricity of 2.1×10^9 kWh and ammonium bicarbonate of 2.87×10^6 tonnes.

2. CO_2 Emissions of System I

At present, the absorption ability of ammonia is about 0.85-1.2 $kgCO_2/kgNH_3$ [3]. The excess ammonia can be recycled to absorb CO_2 again. Then the total absorption factor is about 2.5 $kgCO_2/kg\ NH_3$. The annual ammonia consumption will be 0.64×10^6 tonnes.

For industry production of ammonia by SMR, the CO_2 emission factor is about 1.22 t CO_2 per tonne ammonia [6] (It is 0.97 in theory). Therefore, the annual CO_2 emissions by SMR are about 0.78×10^6 tonnes.

For the ammonia scrubbing process, about 130 kWh electricity is used per tonne ammonium bicarbonate [7]. To produce the electricity, additional CO_2 will be emitted, about 104 $kgCO_2$/tonne. For the annual production of ammonium bicarbonate, the CO_2 emissions will be 0.3×10^6 tonnes.

Not considering the CO_2 emissions in the other process such as transportation, the total CO_2 emissions of system I can be evaluated as 1.16 Mt.

3. CO_2 Emissions of System II

For industrial production of ammonium bicarbonate by coal gasification, the material consumption is 0.25 tonnes of NH_3, and 0.65 tonnes of CO_2 for one tonne of NH_4HCO_3 [7]. For coal gasification, the CO_2 emission factor is about 3.59 tonnes per tonne NH_3 [6]. It can be seen that there is excess CO_2 for NH_4HCO_3 production in the coal gasification process, which is about 0.248 tonnes per tonne NH_4HCO_3. Same as in system I, the additional CO_2 emission will be 104 kg CO_2/tonne NH_4HCO_3 for electricity consumption. Therefore the annual CO_2 emissions in the process are about 1.01 Mt. Putting the CO_2 emissions of power plant together, the total CO_2 emissions of system II can be evaluated as 2.69 Mt.

All the calculations above are summarized in Table 1.

4. Discussions

Due to the lack of data information, the consumption of water, steam, and catalysis in both systems is not involved in the analysis. Their production will emit CO_2 also from the point of view of life cycle. The CO_2 emissions during the manufacturing of equipment are overlooked also. According to experience of life cycle assessment [8], the CO_2 emissions from these overlooked factors are generally less than 5% of total emissions of the system. Therefore, the simplification will not affect the results.

Table 1. Summary of the calculation

No.	Item	Unit	Equation	Result
(1)	Electricity Production	kWh	$7{,}000\times300\times10^3$	2.1×10^9
(2)	CO_2 emissions of PP	tonne	$(1)\times0.8\times10^{-3}$	1.68×10^6
(3)	NH_4HCO_3 production	tonne	$(2)\times0.95\times79/44$	2.87×10^6
(4)	Ammonia production	tonne	$(2)\times0.95/2.5$	0.64×10^6
(5)	CO_2 emissions of SMR	tonne	$(4)\times1.22$	0.78×10^6
(6)	CO_2 emissions for electricity consumption in the process of NH_4HCO_3 production	tonne	$0.8\times10^{-3}\times130\times(3)$	0.3×10^6
(7)	CO_2 emission factor of CG	tonne/tonne	$3.59\times0.25-0.65$	0.248
(8)	CO_2 emissions of CG	tonne	$(7)\times(3)$	0.71×10^6
(9)	CO_2 emissions of system I	tonne	$(2)\times0.05+(5)+(6)$	1.16×10^6
(10)	CO_2 emissions of system II	tonne	$(2)+(6)+(8)$	2.69×10^6

1. PP is simplification for Power Plant.
2. CG is simplification for Coal Generation.

CONCLUSIONS AND SUGGESTIONS

From the analysis of the paper, it can be concluded that the life cycle CO_2 emissions of ammonia scrubbing are about 1.16 Mt a year for a 300 MW coal-fired power plant, whereas 2.69 Mt for industry production process of ammonium bicarbonate. It can be concluded that ammonia scrubbing is an effective way to absorb CO_2 from the flue gas of coal-fired power plants.

The difference in life cycle CO_2 emissions between the two systems is due to the ammonia originating from different sources. Less CO_2 emission of system I is due to the ammonia originating from natural gas, instead of coal in system II. Therefore, ammonia originating from low-carbon sources is significant from the point of view of CO_2 removal.

Furthermore, decomposing ammonium bicarbonate to regenerate ammonia is a better option rather than using it as a kind of fertilizer.

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