Membrane-integrated oxy-fuel combustion of coal: Process design and simulation

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ARTICLE INFO

ABSTRACT

A membrane-integrated oxy-fuel combustion process is designed and simulated in UniSim Design®. The results of the simulation indicate that a net efficiency of 31.8% is obtained for a coal-fired power plant of 182 MW_{th} (assuming only carbon in the coal), including the compression of CO_2 to 100 bar. The specific electrical energy demand for CO_2 capture in this process, including oxygen production and CO_2 compression is $\sim$0.58 MJ/kg. The required membrane area for air separation is $\sim$80,000 m^2 based on Ta-doped $SrCo_{0.8}Fe_{0.2}O_{3-\delta}$ (SCF) membranes. By using the same protocol, the net efficiency is lower for an oxy-fuel combustion process using oxygen from cryogenic distillation of air (29.6%).

1. Introduction

Coal-fired power plants contribute to around 30% of the total anthropogenic CO_2 emission to the atmosphere, which is considered to be one of the main reasons for the global climate change. In the past 20 years several techniques are developed to reduce CO_2 emission and several overviews of these techniques are published [1–4]. Generally, these techniques can be classified into 3 categories: post-combustion, pre-combustion and oxy-fuel combustion.

Post-combustion CO_2 capture involves the removal of CO_2 from the flue gas of a combustion process. The advantage of post-combustion CO_2 capture is that it can be implemented to an existing combustion process (e.g. in a power plant) without making major adjustments in the plant. A disadvantage is that the CO_2 concentration in the flue gas is very low (usually less than 15 mol%) because the main component is N_2 and the flue gas is at near atmospheric pressure. Several techniques are developed for the separation of CO_2 from flue gas such as absorption/desorption, membrane separation and cryogenic distillation [5–9]. The most mature technique in the post-combustion CO_2 capture area is the absorption/desorption method with a monoethanolamine (MEA) solution. The overall energy consumption for CO_2 capture in this process is approximately 1.22–1.62 MJ/kg, which results in a net efficiency drop of 13–15%-points for the whole power plant [10–12].

Pre-combustion CO_2 capture is a process that produces a carbon-free fuel before combustion. This process comprises two main steps: fuel reforming and H_2/CO_2 separation. In the first step fuel is reformed by steam or oxygen or both, and a mixture of H_2 and CO (syngas) is obtained [13,14]. The produced syngas undergoes a water gas shift reaction where CO is converted to CO_2 and more H_2 is produced [15]. After these reactions H_2 is separated from CO_2 and the produced H_2 is combusted with air in a gas turbine to generate electricity. In most cases a heat recovery steam generator (HRSG) and a steam turbine system is coupled to increase the overall efficiency. This process is called integrated gasification combined cycle (IGCC) with CO_2 capture if coal is used as fuel, and natural gas combined cycle (NGCC) with CO_2 capture if natural gas is used as fuel.

Another promising technique for CO_2 capture is oxy-fuel combustion where pure oxygen instead of air [16] is used for the combustion of coal. The result of this process is a concentrated CO_2 flue-gas stream, enabling efficient CO_2 capture, because no amine absorption/desorption process is required [17]. Pure oxygen, required for the oxy-fuel combustion process, can be produced via cryogenic distillation of air, which is a mature technique for oxygen separation from air. However, the power consumption for oxygen production, using this technique, is relatively high. For a standalone cryogenic air separation unit the specific electrical energy for oxygen production is about 0.72–0.86 MJ/kg [18] (oxygen under 0.1 MPa and at 15°C), which leads to a net efficiency drop in the power plant of $\sim$10%-points. This efficiency drop is much smaller than that in the amine based post-combustion CO_2 capture process. In addition, the energy, necessary for oxygen

production by cryogenic distillation, can be further decreased by optimizing this process, but 0.58 MJ/kg seems to be a realistic limit [19]. Pressure swing adsorption (PSA) is another way for the production of pure oxygen; however, this technique is only suitable for small/medium scale oxygen production, not for large scale like in a power plant. In addition, oxygen produced by using PSA usually has a purity of less than 95% [19,20].

Membrane separation is another technique for air separation. The separation can be performed with polymeric membranes at ambient temperature or with ceramic oxygen transport membranes at elevated temperature [21,22]. Polymeric membranes have the advantages of low price, flexibility and easy handling, but O_2/N_2 selectivity is usually limited so only oxygen-enriched air, rather than pure oxygen, can be obtained. In contrast, dense ceramic oxygen transport membranes have infinite oxygen selectivity, because only oxygen can go through these membranes. The membrane can be used in a standalone system to produce pure oxygen, or integrated into chemical processes to supply oxygen for reactions [10,23–25]. One of these processes is the Advanced Zero Emissions Power Plant (AZEP). In this concept, natural gas is used as fuel and an oxygen transport membrane is implemented for oxygen supply. Here, a combined (gas turbine and steam turbine) cycle arrangement is applied for power generation. Griffin et al. [26], performed process calculations and the results indicate that the AZEP technique will result only in a loss of about 4%-points in efficiency including the pressurization of CO_2 to 100 bar, as compared to approximately 10%-points loss when using cryogenic distillation technique to produce oxygen.

When coal is used as fuel in a power plant, however, a combined cycle is difficult to be applied and only a steam turbine is used for power generation; thus power plant efficiency is expected to be lower. In the membrane-integrated oxy-fuel combustion concept, oxygen, as separated from air by a membrane, is swept by CO_2, while this O_2/CO_2 gas mixture is used for the combustion of fossil fuels. After purification, the exhaust gas mainly consists of CO_2. Part of the CO_2 is recycled as sweep gas, and the other part is compressed for delivery and storage.

A lot of research is performed to explore suitable membrane materials for air separation for this application [27–33]. However, little work has been done on the overall evaluation of a membrane-integrated oxy-fuel combustion process [34,35], such as membrane area and energy balance estimations or to show whether the currently developed membranes are sufficient for, or far from, industrial application.

In this study, a membrane-integrated oxy-fuel combustion process is designed for a coal-fired power plant of 182 MW_{th}. This process is simulated in UniSim Design® using pure carbon to represent the coal, while the membrane area, necessary for air separation, the net efficiency of the power plant and the energy demand for CO_2 capture are calculated. For comparison, two other combustion processes are presented: a conventional coal-fired power plant, using air, without CO_2 capture and an oxy-fuel combustion process using oxygen from cryogenic distillation of air. Especially the differences between these processes are discussed.

2. Coal-fired power plant without CO_2 capture

In a conventional coal-fired power plant air is used to combust the coal and the CO_2-containing exhaust gas is emitted to the atmosphere. A simple scheme of this process is shown in Fig. 1. Here air is compressed to 0.17 MPa (A1) to carry the pulverized coal to the burner, where combustion occurs. An excess of 15% oxygen is designed to ensure a complete combustion and this excess level is maintained for the other processes studied in this
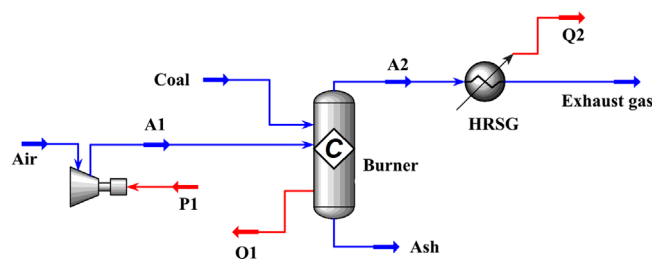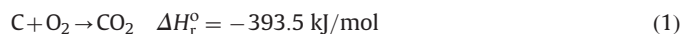

Fig. 1. Process for a conventional coal-fired power plant without CO_2 capture (see Table S1 in Supporting information for details).

work. It is assumed that there is only carbon in the coal and that the reaction between carbon and oxygen is stoichiometric:

$$C + O_2 \rightarrow CO_2 \quad \Delta H_r^o = -393.5 \ kJ/mol \qquad (1)$$

The thermal energy, released from the burner, is used by a steam turbine for electricity generation. The temperature of the flue gas (A2 in Fig. 1) from the burner is further decreased to 50 °C by a Heat-Recovery-Steam-Generator (HRSG), which energy (Q2) is also used for the steam turbine system. After the HRSG the exhaust gas consists of some impurity (acid) gases like SO_2 and NO_x, which have to be removed in a purification step before emission to the atmosphere is allowed. However, as is mentioned above, it is assumed that there is only carbon in the coal and thus here a purification step is omitted. The same strategy holds for all studies in this work.

As mentioned above, the thermal power from the burner and HRSG is used for electricity generation, and the conversion ratio is called the thermal efficiency (η_{th}). In principle, η_{th} has a theoretical maximum Carnot efficiency of ~64% assuming the water/steam cycle operates between room temperature (25 °C) and 565 °C (=creep temperature of stainless steel). However, an actual efficiency of up to 42% for a modern coal-fired power plant is usually expected [3]. In this study η_{th} is calculated based on a reheated Rankine cycle (Fig. 2), which is commonly used in fossil fuel fired power plants for electricity generation. In this cycle, water is compressed to 220 bar, evaporated and heated to 550 °C. The supercritical steam expands in a high pressure steam turbine to generate electricity, and the exhaust gas is reheated to 550 °C for a next expansion. The outlet of the low pressure steam turbine is directly connected to a condenser where the temperature is as low as 20–30 °C, which leads to a steam pressure of ~0.05 bar. The efficiency η_{th} is defined as the net electricity generated divided by the thermal energy ($\dot{Q}_s = Q1 + Q2 + Q3$) from the burner and the HRSG (equals to $Q1 + Q2$ in Fig. 1 in the relevant process):

$$\eta_{th} = \frac{P1 + P2 + P3 - P4}{\dot{Q}_s} \qquad (2)$$

In a Rankine cycle the value of η_{th} is mainly determined by the steam temperature. Generally the higher the steam temperature the higher the efficiency, but the upper limit of the steam temperature is restrained by the creep temperature of the material used for steam turbines. In this work an efficiency of 40% is obtained when the steam temperature is set to 550 °C. To ensure such an efficiency, the exit temperature of the burner is maintained higher than 550 °C, and this protocol applies to all cases in this study. The heat of the HRSG cooler (T ranges from 550 to 50 °C, duty in the range of 31 to 40 MW) is used to cover the initial heating of the high pressure water in Heater-1 (Fig. 2). The rest of the duty of Heater-1 is obtained from the burner.

The electric power output (P_{out}) and net efficiency (η_r) of the process are calculated by the following equations:

$$P_{out} = (Q1 + Q2) \times \eta_{th} - P1 \qquad (3)$$