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A parametric study of CO_2/N_2 gas separation membrane processes for post-combustion capture

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ABSTRACT

Capture of CO_2 from flue gases produced by the combustion of fossil fuels and biomass in air is referred to as post-combustion capture. Chemisorbent processes are considered to be the most feasible method and are already at an advanced stage of development, but gas separation membranes are attracting more and more attention as a possible alternative. This paper describes a detailed parametric study of mass and energy balances for a simulated single membrane process. Typical operating conditions (CO_2 concentration in the flue gas, pressure and temperature, etc.) together with the influence of the membrane quality (permeability, selectivity) and membrane area on membrane performance (CO_2 separation degree and CO_2 purity) are simulated over a wide range of parameters.

1. Introduction

Carbon dioxide (CO_2) capture and storage (CCS) is a process consisting of the separation of CO_2 from industrial and energy-related sources, transport to the storage location and long-term isolation from the atmosphere [1,2]. There are several different types of CO_2 capture systems: post-combustion, pre-combustion, oxyfuel combustion and other industrial separation techniques [3].

The so-called post-combustion process captures CO_2 from flue gases produced by the combustion of fossil fuels and biomass [2]. Power plants emit more than one-third of all CO_2 emissions worldwide. Because the CO_2 concentrations in the flue gas are low – typically, 3–5 mol% in gas plants and 13–15 mol% in coal plants – the cost of capture would be significant [1,4]. Separation processes such as absorption, membranes, adsorption and cryogenic fractionation are listed as potential candidates for CO_2 capture in the post-combustion process. Furthermore, the ammonia scrubbing process based on chemical absorption with a monoethanolamine (MEA) solvent plays a dominant role [5], at least in the near future. Many companies are currently involved in building full-scale demonstration power plants with MEA absorption process, e.g. RWE 500 MW_e coal-fired power plant (PCC) in Tilbury, UK; Statoil/Shell 860 MW_e natural gas power plant (NGCC) in Tjeldbergodden, Norway; Statoil/Dong 280 MW_e natural gas power plant (NGCC) in Mongstad, Norway [6,7]. MEA was developed more than 60 years ago as a general, nonselective solvent to remove acid gases, such as CO_2 and H_2S, from natural gas streams [8]. However, the chemical processes associated with the degradation of the MEA are still not completely understood, which leads to increased material costs, waste disposal costs, and energy demands for the CO_2 capture process [8]. Of course, in order to extend the solvent life the high demands made by the SO_x (<10 ppm) and NO_2 (<20 ppm) [9] content in the flue gas leads to extraordinarily high expenditure for the pre-capture process. Furthermore, almost 50% of the low-pressure steam from the intermediate/low-pressure (IP/LP) steam turbine should be employed for regeneration of the solvent used to release CO_2, which requires a considerable amount of thermal energy generated by the power plant [5]. A life cycle assessment (LCA) of MEA-based carbon capture shows that the reduction of CO_2 emissions using this method is achieved at the expense of increasing other emissions and corresponding environment impacts and calls for a further investigation of membrane separation processes [10]. All above this is the motivation for investigating the most competitive alternative technique – gas separation membranes.

In view of the application field for gas separation membranes, especially for CO_2 flood EOR (enhanced oil recovery) and purification in pipeline natural gas projects, it is well known that since the early 1980s polymer gas separation membranes have achieved developmental and commercial success in comparison to conventional amine and cryogenic fractionation [11,12]. Several features distinguish the gas separation membranes used in post-combustion at power plants from the aforementioned application

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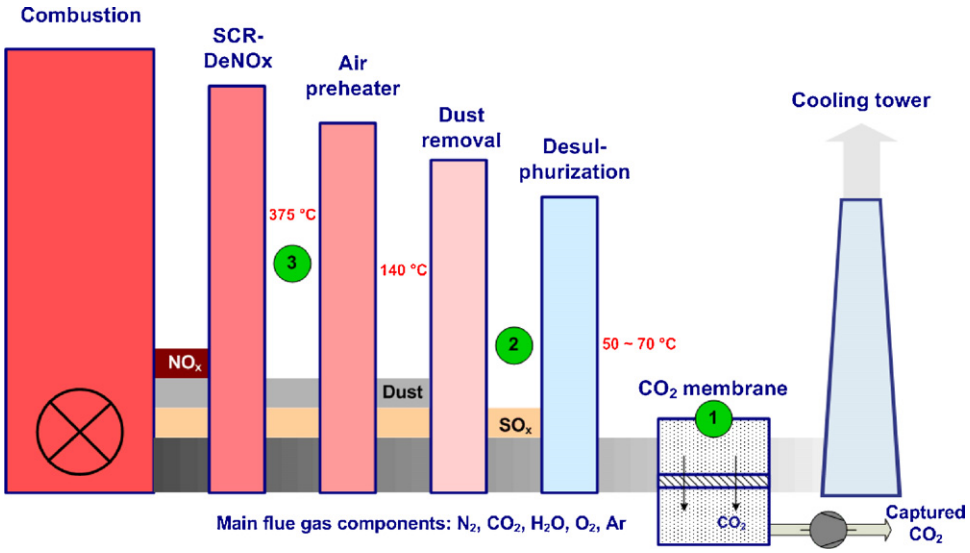

Fig. 1. Schematic diagram of CO_2 membrane position in a post-combustion flue gas line. The Arabic numerals 1, 2 and 3 show the proposed test positions for different membranes in the EnBW power plant.

fields: one is the different gas separation goal, which is CO_2/N_2, instead of CO_2/CH_4, H_2/CH_4; the second is the low CO_2 concentration in the flue gas – about 14 mol% (the gas recovered from the gas field by EOR obviously has a higher CO_2 concentration); and the third is the pressure of the flue gas, which is in the range of ambient pressure rather than being pressurized.

A feasibility study for the polymer gas separation membranes used in post-combustion by Van der Sluijs [13] claimed that gas separation membranes with a selectivity of at least 200 are required to make them a serious competitor in comparison to other separation techniques. This conclusion is still widely accepted and has been cited by many other researchers [14,15]. Gas separation membranes have recently been rejected again in the early stages of a process selection project [2,5].

CO_2/N_2 gas separation membranes have developed remarkably in the past 15 years [16–18], especially polymer membranes; a PAMAM dendrimer membrane with a selectivity of 230 and 400 has been developed by RITE [19,20]. In addition, systems analysis and design help us to understand and arrange the membrane process effectively. Although several groups, e.g. Favre (French), Hägg (Norwegian) and Hendriks (Dutch), have done a great deal of work in this field [21–25], comprehensive simulation work on operating conditions (feed gas CO_2 concentration, pressure and temperature, etc.) together with the influence of the membrane quality (permeability, selectivity) and membrane area on the membrane performance (CO_2 separation degree and CO_2 purity) has seldom been involved in a coal-fired post-combustion capture process. It is now absolutely necessary to investigate the separation process of the membranes for post-combustion so that a more competitive CO_2 capture method can be developed.

As part of the METPORE project [26], the first membrane test worldwide concerning mechanical stability in a real flue gas environment used for post-combustion CO_2 capture was performed in a power plant of the EnBW company, Germany, in April 2008. The experiment was carried out both for a cermet-type membrane and a polymer membrane. A publication is being prepared about the experimental equipment and results.

The present work is concerned with a detailed parametric study of mass and energy balance simulation for a single-stage CO_2/N_2 membrane process.

2. A single-stage CO_2/N_2 membrane process

The flue gas line for a conventional pulverized coal combustion power plant is shown in Fig. 1. On the basis of the working conditions of coal-fired power plants several different type of membranes (e.g. sol–gel derived stainless-steel-based cermet membrane developed by IEF-1 Forschungszentrum Jülich, Germany; PEBAX polymer membrane (working temperature lower than 70 °C) developed by GKSS, Germany) were proposed for testing in three different positions in the EnBW power plant. Position 1, analogous to amine stripping processes [5] after the deNOx (SCR), dust removal (E-filter) and desulphurization (FGD) processes, was chosen for the first test. This position has been widely accepted to be most suitable for the polymer membrane. The decision to apply the cermet-type membrane at the other positions was based on the fact that this membrane can work in a harsh environment. According to the purity requirements of CO_2 transport (>95 mol% CO_2 purity) [27,28] and re-injection, a purification process should be included (CO_2 purity more than 99 vol%) after capture [29]. The flue gas information given in Table 1 was obtained from the industrial partners.

A single-stage membrane process is shown in Fig. 2. All relevant parameters are listed here. Operating conditions (pressure, temperature, CO_2 concentration and flow rate of the feed gas) act on a certain membrane (selectivity, permeability and area), then the performance of the membrane (CO_2 purity and degree of CO_2 separation) can be predicted. The degree of CO_2 separation is also described as the CO_2 recovery ratio in the literature [13,21–23].

Table 1
Flue gas data after FGD in power plants corresponding to data from the literature [30]

Data	Unit	Power plant 1	Power plant 2
Coal	–	Hard coal	Hard coal
Efficiency	%	45	44
Power output	MW_e	1000	1000
Flue gas flow rate	$m^3\ s^{-1}$	833	850
CO_2	mol%	13.6	14.8
N_2	mol%	70.7	70
Temperature	°C	50	80