Modelling and Multi-stage Design of Membrane Processes Applied to Carbon Capture in Coal-fired Power Plants

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Abstract

According to recent predictions, energy generation from coal will continue to play a key role in the next decades. The UK 2008 Climate Change Act requires a reduction of 80 % of greenhouse gases emissions by 2050, and carbon capture and storage will have a key role in order to meet this target.

This work focuses on post-combustion capture from coal-fired power plants based on membrane separation. A detailed multi-stage design is presented: the developed flowsheet includes cross-flow and countercurrent-sweep stages. The cross-flow stages are based on a 2D model implemented by our research group able to predict the separation through spiral-wound permeators. Different process configurations are analysed, with the aim of reducing both energy consumption and membrane area.

An economic analysis is also included: both capture and avoidance costs are evaluated. The estimated values are compared with data available in the literature for processes based on amine capture technology.

1.   Introduction

According to major international organisations [1-3], climate change due to increased greenhouse gas emissions is a key issue and it will affect the energy policies of the next decades. Despite the increasing investments in renewable energies, combustion of fossil fuels will still play a major role for energy generation in the near future. An example is given by China [4], where the coal demand is expected to reach 4.3 billion tonnes by 2025.

In this scenario CCS – Carbon Capture and Storage – is a solution that has the potential of helping the reduction of the global emissions. Significant research efforts are being carried out on both engineering and geological aspects of CCS.

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Membrane gas separations are already applied in smaller scale for processes like natural gas sweetening and production of oxygen-enriched air and they have the potential of playing a competitive role in second generation carbon capture technologies. Thanks to strengths such as absence of regeneration apparatus, modularity and small footprint, they may represent an alternative to other carbon capture solutions for coal-fired power plants.

Zhao et al. [5] propose a study of the possible dual-stage configurations including a detailed economic and energetic analysis based on countercurrent stages, starting from the flue gas of a coal-fired power plant. An optimal membrane CO_2/N_2 selectivity (in the range $50 \div 100$) is identified and a capture cost of 31 €/t$_{CO2}$ is estimated.

A similar approach can be found in the work of Hussain and Hägg [6]: a dual-stage countercurrent investigation based on facilitated transport membranes is presented, including an economic evaluation of the capture costs. Considering a value for the Gas Processing Cost (GPC) of 1.5 $/MSCF typical for amines, a cost of 0.85 $/MSCF is estimated for membranes.

Merkel et al. [7] propose a hybrid solution including a refrigeration stage integrated into the compression of the CO_2 up to 150 bars. The material suggested for the membrane modules is Polaris[TM] and simulations are run integrating countercurrent and cross-flow stages. A new configuration involving the recycle of part of the CO_2 to the boiler is investigated: the identified capture costs are 23 $/t$_{CO2}$. The possibility of recycling the CO_2 to the boiler is also investigated by RTI [8, 9] focusing on the Generon polycarbonate high flux membrane: a capture cost of 30 $/t$_{CO2}$ is reported.

This work is focused on membrane separation applied to post-combustion carbon capture from a coal-fired power plant, based on process simulations and their possible optimisation. A complete flowsheet and a cost analysis are presented: an overall estimation of the feasibility of this technology is given, in alternative to the traditional solvent-based capture. The simulations are run using UniSim Design®, the Honeywell process simulator.

2.   **Material and process conditions**

The feasibility of membrane process separation for carbon capture is directly related to the materials and their properties. Different polymers reported in the literature [10-13] achieve both high CO_2 permeability and selectivities over the other components. For example, Polaris[TM] [7] is a promising polymeric material and tests with real flue gas have been carried out with promising results [14, 15]. PIMs – Polymers of Intrinsic Microporosity [16] – belong to another category of materials showing promising performances, particularly regarding high CO_2 permeabilities. In the case of PIM1 post-treated with methanol, a value of 12600 Barrer [17] has been achieved, but issues related to the stability of the polymer and the aging effect are still under investigation.

The permeance, defined as the ratio between permeability and membrane thickness, is the parameter generally used in process simulations. A CO_2 permeance of 12000 GPU has been assumed: it can represent the performance of a membrane with a permeability of 12000 Barrer and a thickness of 1μm. The selectivity of CO_2 towards Nitrogen and Oxygen, respectively, has been assumed 50 [7] and 15. These have been considered as promising characteristics for a future material combining high CO_2 permeability and selectivities over the other components.

Another important issue is the water content and its influence on the process: the temperature at the inlet of the modules is a key parameter in determining the water concentration. It is well-known that water achieves high permeabilities in dense polymeric membranes and that it can improve the separation acting as an internal sweep. However, the stability of the polymers, both to high temperature and to high water content, is one of the major problems [18]. For the purpose of this work, a CO_2/H_2O selectivity equal to 1 is assumed [19].

The reference power plant for the simulation is the DOE case 9 – subcritical coal-fired power plant [20]. However, the Argon content resulting from the power plant simulation has been considered as nitrogen. The flue gas properties and conditions are reported in Table 1.

Table 1: flue gas properties

Molar Flow	$2.26 \cdot 10^4$ mol/s
T	57°C
P	1 atm
Molar fractions	
CO_2	0.13
N_2	0.67
O_2	0.03
H_2O	0.17

An important aspect in the separation is the pressure ratio [18, 21], the ratio between the feed and the permeate pressure. According to recent studies [7], vacuum at the permeate side has been identified as optimal option. With the aim of reducing the energy consumption, the simulations are run using moderate compression (blower) at the feed side, fixing the pressure ratio to 5 as suggested in the literature [7, 14]. Consequently, the feed stream is compressed up to 1.1 bar while for the permeate side a pressure of 0.22 bar is set.

An efficiency of 80% is considered for pumps, compressors and expanders [6, 7]. After performing simulations fixing the feed temperature of 50°C [5-7], a feed temperature of 25°C is assumed, since the water content is one of the key parameters in the simulations. However, the dependence of the permeability with temperature is neglected.

3.   Multi-stage design

3.1   *Modelling and integration into UniSim Design*[®]

Different models have been analysed and implemented, with the aim of having predictions closer to the performance of industrial modules. In particular, the models adopted to simulate the membrane stages are nondisperse plug-flow (countercurrent flow pattern with sweep) and the 2D cross-flow implemented by our research group. The main assumptions are no pressure drops, constant permeances along the module, ideal gas and isothermal behaviour.

A key step in the modelling process is the implementation in UniSim Design[®] (version R400), the Honeywell software simulator: the models are completely integrated into the simulation software as an exstension unit operation. This allows the user to run simulations into the Unisim environment (Figure 1).
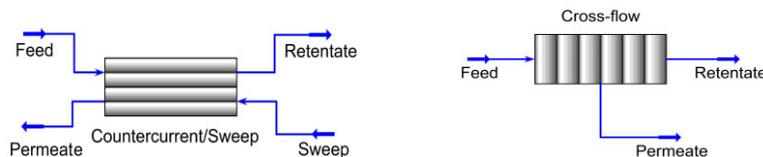

Figure 1: Membrane_UoE (1.0 and 2.0)

3.2 Investigation strategy

Preliminary single-stage simulations have been carried out: an analysis of the results obtained shows that a multi-stage scheme is required to achieve the fixed targets in terms of overall performances.

A complete flowsheet including the power plant, the multi-stage scheme and the final compression train is presented in order to evaluate the overall costs, directly related to the energy consumption and membrane area. The investigation is focused on 2 options: a pure retrofit - starting directly from the flue gas - and a possible modification of the complete flowsheet.

The coal-fired power plant is the DOE case 9 – subcritical coal-fired power plant – simulated by our research group. All the simulations are investigated by fixing the CO_2 recovery at 90%.

3.3 Retrofit option

After performing single and dual-stage simulations, the benefit of combining countercurrent-sweep and cross-flow stages is confirmed with a reduction of both energy consumption and membrane area.

As a result of the investigation, a multi-stage design starting directly from the flue gas of the plant is proposed in Figure 2.
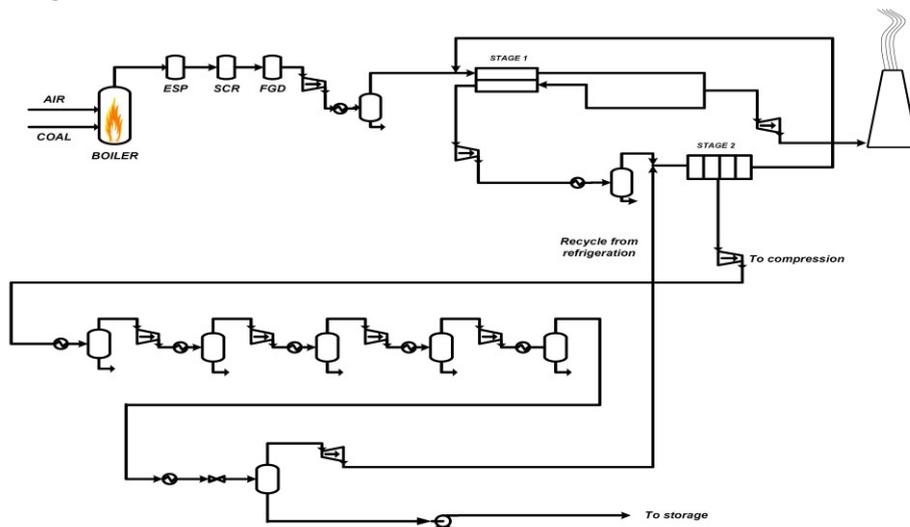

Figure 2: retrofit option - flowsheet

The flue gas is sent to a high-recovery countercurrent stage 1 using its retentate as sweep to improve the driving force for the separation (and, consequently, the overall recovery). Stage 2 is a cross-flow/high-purity stage: the retentate, due to the high CO_2 content (50 %), is recycled to stage 1, while the permeate stream is sent to the compression train. A multi-stage compression system with intercoolers to 45°C has been included into the simulation, according to the US Department of Energy guidelines [20].

After being compressed up to 30 bar, the CO_2 rich stream is cooled to – 20°C [7] and then expanded via a Joule-Thompson valve, in order to decrease the temperature and increase the liquid fraction. The gas stream (which presents CO_2 content of 70 %) is sent as recycle to stage 2 after being expanded, while the liquid stream is pumped up to 150 bars.

Compared to the direct compression of the stream from stage 3 to 150 bar, a reduction of both energy consumption and membrane area is achieved. Thanks to the refrigeration stage, the final CO_2 purity is also improved. An efficiency of 50% is assumed for the refrigeration in order to estimate the overall energy consumption.

Table 2: multi-stage simulations - results

Configuration	Compression [MW]	Refrigeration [MW]	En. Consumption [kJ/mol$_{CO2}$]	Area [10^5 m^2]	Purity
Retrofit	115	20	50	3.00	0.98
Boiler	104	20	46	2.30	0.98

3.4   Recycle to the boiler option

Since the aim is to improve the overall performance of the capture system, a different configuration is now analysed. Starting from what is presented in the literature [7], this design uses air as sweep to a high-recovery stage in order to recycle part of the CO_2 to the boiler. In fact, by using this option, the flue gas results in a higher CO_2 concentration and, consequently, the driving force for the separation is improved. The schematic flowsheet of this option is reported in Figure 3.
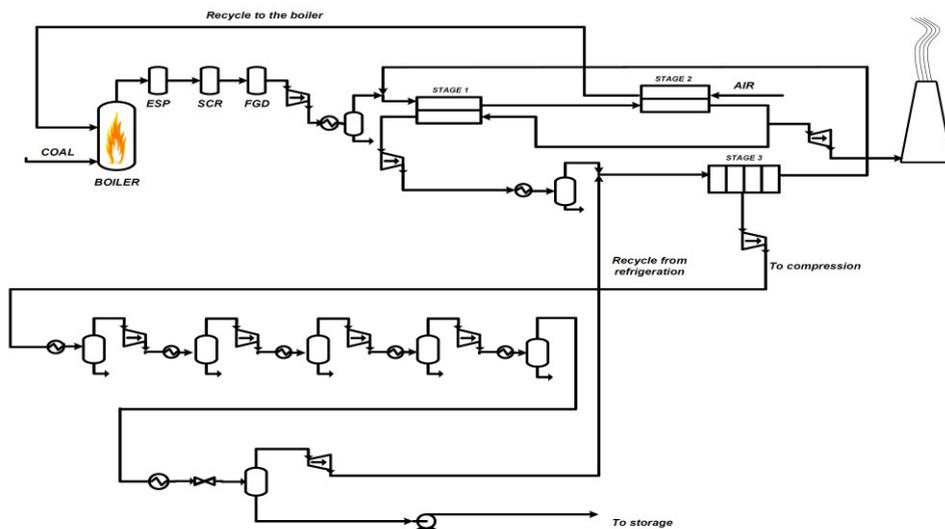

Figure 3: boiler option - flowsheet

The flue gas is sent to a 1st countercurrent stage: the permeate is sent to a cross-flow module in order to achieve higher CO_2 purities. The retentate from stage 1 is then sent to a high-recovery countercurrent module with air as sweep at atmospheric pressure [7]. Consequently, the permeate stream from the 2nd stage results in a stream containing 3% of CO_2 and 19% of oxygen, which is directly sent to the boiler. However, by keeping the same air flow rate as the base plant, 7% of the oxygen content back-permeates in the retentate stream from stage 3 and it is directly sent to the stack.

Differently from the retrofit option, 1st stage has a lower CO_2 recovery due to the additional countercurrent-sweep stage. Consequently, the permeate stream from stage 1 has a lower flow rate and higher CO_2 concentration: this justifies the lower energy consumption. The compression and refrigeration sections are the same as the retrofit option.

As already reported by Merkel et al. [7], this configuration presents the benefits of both reducing the energy consumption and the membrane area (it is referred as *boiler* case in Table 2).

It is evident from the results presented that this configuration, compared to the previous *retrofit* one, is able to improve the overall performance of the capture system. In addition to this, the oxygen content in the final liquid stream is reduced compared to the retrofit option due to its lower content in the flue gas.

4. Economic analysis

After the multi-stage design particular attention given to the economic analysis. The investigations presented in the literature show different calculations of the carbon costs and it is difficult to carry out a comparison among them due to the different markets (mainly US and Europe) and reference plants chosen for the estimation.

The analysis developed in this paper starts from an estimation of the capture costs based on the methodology proposed by Zhao et al. [5]. In order to have a more general estimation, the avoidance costs are calculated. This parameter is a useful tool in order to compare the capture application for different technologies and it is also more general since it based on the calculation of the *LCOE* (Levelised Cost of Electricity), an indicator of the capture costs throughout the plant life.

4.1 Capture costs

An estimation of the capture costs is presented as initial step of the economic evaluation: this indicator represents the annual cost referred to the captured CO_2 in the plant and its value for amines is in the range $40 \div 50$ €/t_{CO_2} [13]. Different cost analyses are reported in the literature [6, 7] showing promising results regarding the possible application of membrane technology to carbon capture.

The reference calculation and methodology for our initial analysis is based on the one presented by Zhao et al. [5], who estimate a capture cost of 31 €/t_{CO_2}. The cost of energy is updated to the actual European reference parameters [22].

An overall result of the capture costs including the main process parameters is reported in Table 3.

Table 3: performances and capture costs

	Power required [MW]	En. Consumption [kJ/mol$_{CO_2}$]	Area [10^5 m^2]	Purity	Capture costs [€/tCO_2]
Retrofit	135	50	3.0	0.98	**27**
Boiler	124	46	2.3	0.98	**23**

The second configuration has the benefit of reducing the capture costs by 10% in comparison to the retrofit case, with an improvement in both the energy consumption and the membrane area. In Figure 4 the contributions of capital, operation & maintenance (O&M) and energy costs are presented.

The main difference between the two options is the energy cost contribution, which is higher for the *retrofit* case. The difference in capital costs is less evident despite the 30% of area reduction because the highest contribution to the cost is due to the skids [5]: their cost is due to the fact that they are responsible for maintaining compression (or vacuum) in the relative module side.
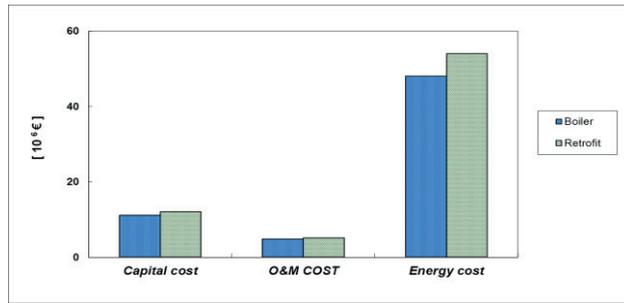
Figure 4: distribution of the capture costs

4.2   Avoidance Costs

As a final step of the analysis presented, an estimation of the avoidance costs (**AC**) is included. The formulation is reported in (1), where **SCE** is the Specific Carbon Emission (ratio between the CO_2 emitted and the net power generated) and *LCOE* the Levelised Cost Of Electricity.

$$AC = \frac{LCOE_{CCS} - LCOE_{ref}}{SCE_{ref} - SCE_{CCS}} \quad (1)$$

The *LCOE* calculation follows the methodology presented by ZEP (Zero Emission Platform [23, 24]) and by the UK DECC (Department of Energy and Climate Change [24, 25]). The data from   IEA (International Energy Agency [3]) are also considered, despite the fact that these do not include the storage costs. The membrane capture contribution is included into the base-case calculation, based on the DOE case 9 subcritical coal-fired power plant [20]. The calculated avoidance costs are reported in Figure 5, where the following estimations with amines are also shown:

- DOE case 10 (base case with integrated amine capture) estimated by using the parameters from the UK DECC [24, 25];
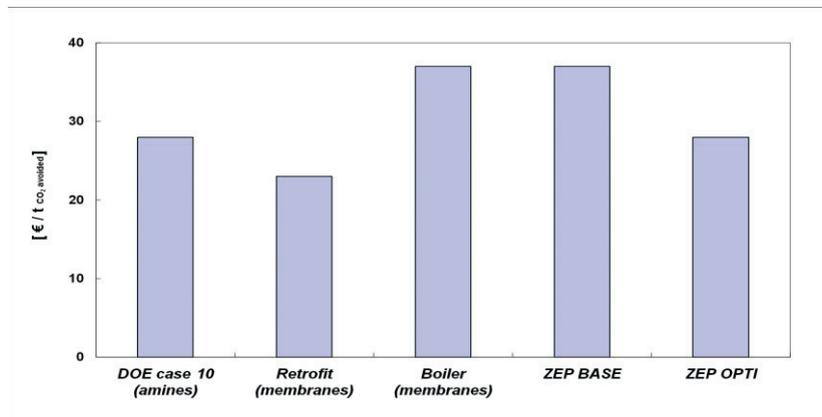- ZEP BASE and OPTI (optimal) [23, 26].

Figure 5: avoidance cost estimation

An analysis of Figure 5 shows that both membrane cases have the potential of reducing the avoidance costs compared to the reported amine cases and exceed even the ZEP OPTI predictions, which are based on the best possible cost options.

5.　　Conclusion

We have presented an analysis based on an integrated process flowsheet simulation in Unisim, which includes specialised modules for the membrane units. The analysis shows that multistage membrane separations can be a competitive solution for post-combustion capture from coal-fired power plants, in alternative to amines. The overall capture performance meets the separation requirements and reduces the capture cost by more than 30%.

Given the stringent requirements on the separation performance on the membrane modules, we are developing more detailed models and simulations to improve the predictions under industrial conditions.

Acknowledgements

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