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Energy Procedia 1 (2009) 487–494
Energy Procedia
www.elsevier.com/locate/procedia

GHGT-9

Opportunities for CO_2 capture through oxygen conducting membranes at medium-scale oxyfuel coal boilers

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Abstract

Ecofys and ECN conducted a study commissioned by the IEA Greenhouse Gas R&D Programme to identify the most suitable combinations of medium-scale CO_2 sources (1 – 100 MW_{th}) and capture technologies, with respect to potential and costs (see parallel paper by Hendriks et al.). An industrial coal-fired Circulating Fluidized Bed (CFB) boiler with oxyfuel combustion and Oxygen Conducting Membranes (OCM) appeared to be an economically attractive combination to capture CO_2. This paper describes the principle and economic evaluation of this combination in comparison with a reference coal boiler without CO_2 capture, as well as a coal boiler with CO_2 capture based on amine scrubbing.

Keywords: CO_2 Capture; Medium-scale; Oxyfuel; Oxygen conducting membranes

1. Introduction

A medium-scale coal-fired CFB boiler used for industrial steam generation appears to be particularly suitable for equipment with OCM that allow oxyfuel combustion. Oxygen is the only component transported through these dense membranes, while the closed configuration of these boilers results in negligible air-in leakage. This results in a limited build-up of nitrogen in the recycle, and consequently in a relatively pure CO_2 product stream. However, it must be noted that OCM are in a relatively early stage of development and not presently commercially available.

To facilitate sufficient oxygen transport through the membranes, OCM require elevated temperatures (800 °C) and feed pressures. Therefore the OCM unit is placed in a configuration that resembles the gas generator part of a gas turbine. Two approaches for air preheating were examined. One system was based on natural gas combustion in a combustion chamber (Case 1), and a second system based on radiant heat exchange panels inside the boiler (Case 2). Furthermore, a coal-fired CFB boiler with MEA scrubbing was evaluated (Case 3) as a reference case with CO_2 capture.

This paper describes the detailed assessment of a new-build oxyfuel coal boiler with (OCM). This alternative has been selected for further evaluation described in a recent study commissioned by the IEA Greenhouse Gas R&D Programme [1]. It comprises a 50 MW_{th} industrial Circulating Fluidized Bed (CFB) boiler for generation of low-pressure steam (10 bar, 200 ºC).

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doi:10.1016/j.egypro.2009.01.065

2. Plant Description

In Case 1 "natural gas and coal", natural gas is combusted to preheat air that is fed to the OCM. Case 2 "all coal" involves preheating of air fed to the OCM, by mounting a heat exchanger inside the boiler.

Air must be heated to 800 °C to facilitate oxygen permeation through the membrane, and moreover sufficient difference in oxygen partial pressure on the feed and permeate side of the membrane is required [2]. It must be noted that OCM are still subject to research, therefore little is known about the effect of constituents such as SO_x, chlorines and mercury on its performance. Moreover, the chemical and mechanical stability of the membranes as well as the membrane sealing form important issues in pursuit of commercial implementation.

The simplified process layout is depicted in Figure 1, the air inlet stream is compressed to 18 bar and further heated by the OCM retentate outlet stream. For Case 1, the targeted temperature of 800 °C is reached by addition of natural gas in the combustion chamber. This results in a lower carbon capture ratio since flue gases obtained from natural gas combustion are vented into the atmosphere. Case 2 comprises further preheating of air by a gas-to-gas heat exchanger mounted inside the coal boiler (not depicted).

The oxygen-lean air downstream of the OCM is expanded and used to pre-heat Boiler Feed Water (BFW). The oxygen-lean air is subsequently released to the atmosphere at 105 ºC. Coal is fed to the oxyfuel CFB boiler together with limestone to capture SO_x in-situ. The boiler generates steam (10 bar, 200 ºC) from Boiler Feed Water (BFW). The flue gas from the boiler is led to a cyclone and a dry Electro-Static Precipator (ESP) where ash is removed. The LP steam conditions allow a flue gas temperature of 150 ºC, which is above the acid dew point temperature due to in-situ desulphurisation. The CO_2-rich primary recycle maintains the boiler temperature at 900 ºC, by recycling approximately 75% of the particulate-free flue gas to the inlet of the boiler. Approximately half of the remaining flue gas is fed to the knockout drum, while the other half is preheated by the permeate outlet stream of the OCM. The retentate stream is enriched with oxygen that permeated through the OCM, and is subsequently cooled and used as secondary recycle to the boiler.

With respect to Case 2 it must be noted that the implementation of the air preheater inside the boiler is highly challenging. Gas-to-gas heat exchangers typically require large surface areas due to the low heat transfer coefficient, and moreover state-of-the-art heat exchangers are suitable for operation at relatively low temperatures. Research on heat exchangers capable of heating pressurised air up to temperatures of approximately 1000 ºC is ongoing, pursuing application in externally fired combustion cycles [3]. These heat exchangers are to be positioned in a pulverised coal boiler, after which the air is further heated by natural gas addition and subsequently expanded in a turbine. Two prototypes are under development: a finned-tube convective arrangement and a radiant panel where tubes a covered with refractory liner. Experiments demonstrated that the first arrangement suffers from particulate deposition on the finned-tubes while the refractory liner in the second arrangement is particularly vulnerable with respect to alkali slag corrosion, resulting in refractory replacement at regular intervals [4]. Moreover, the application within a CFB boiler operated at 900 ºC results in a relatively large heat exchanger surface area compared with a pulverised coal boiler operated at more elevated temperatures.

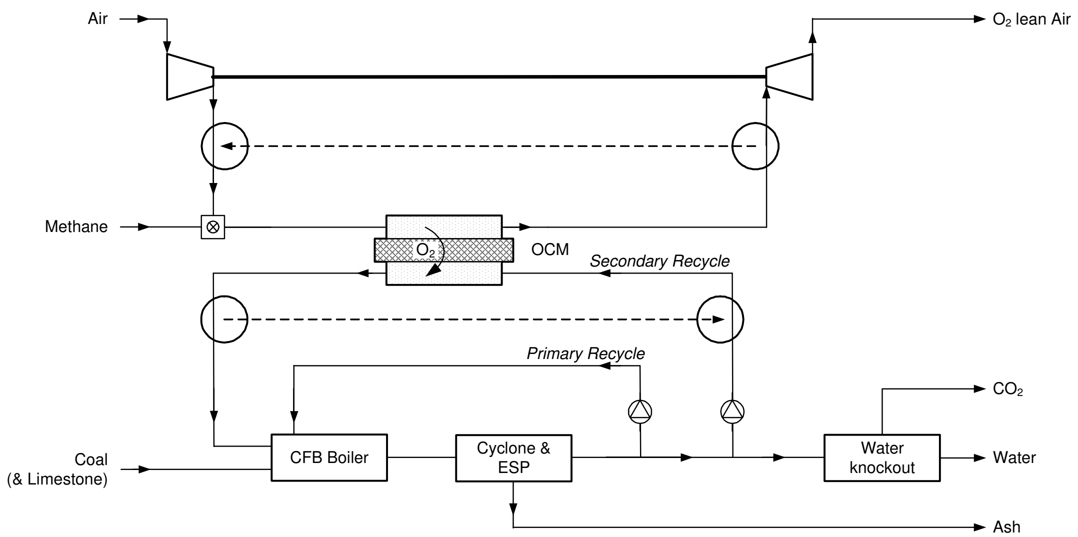

Figure 1 Simplified process layout coal-fired oxyfuel boiler with OCM (Case 1)

Coal boilers require significant start-up and shut down periods to prevent excessive thermal stresses in the refractory liner, which could result in failure. A typical start-up and shut down period of 8 hours is appropriate for a boiler of the size specified in these cases. If the heating rate of the OCM amounts two Kelvin per minute, the start-up and shut down period matches with the boiler. This seems practically feasible for materials used for the OCM. The flow of the secondary recycle can be gradually increased if both temperature and pressure at the feed side of the membrane allow oxygen permeation.

The combustion of methane in Case 1 allows start-up of the OCM prior to start-up of the boiler. This implies that oxygen is also produced and used as oxidation agent during start-up of the boiler. Moreover, the primary recycle is operated to recycle CO_2 to the boiler during start-up, which results in reduced size of the AGR/SCR section since both throughput and NO_x formation are significantly reduced. Moreover, CO_2 can be captured from the flue gases of the coal boiler during start-up.

In Case 2 coal is combusted with air during start-up; hence the primary recycle is not used because oxygen is already sufficiently diluted by nitrogen. This results in an increased NO_x formation, since air is used as oxidation agent. Application of the secondary recycle is omitted during start-up, since both temperature and pressure on the feed side of the OCM do not result in oxygen permeation through the membrane until the preheated air approaches a temperature of 800 ºC.

In Case 1, the upper part of the flow sheet (above the OCM) resembles the gas generator part of a gas turbine. The application of methane to increase the air inlet temperature of the OCM allows enhanced adaptation of the oxygen separation section of the plant upon thermal load fluctuations. The flexibility upon thermal load fluctuations is slightly hampered by the plant configuration in Case 2, due to the integration of air preheating within the CFB boiler.

3. Modelling

Case 1 and 2 have been thermodynamically assessed with Aspen Plus (version 13.1, AspenOne). The dedicated membrane model developed by ECN [5] was used during the simulations in Aspen Plus. This Fortran based model allows selection of logarithmic transport behaviour across membranes, amongst other transport mechanisms. The most important assumptions are displayed in Table 1.

Table 1 Assumptions modelling

Section	Parameter	Units	Value
OCM	Temperature	°C	800
	Air inlet pressure	bar	18
	Average oxygen permeation	$g\ m^{-2}\ s^{-1}$	2.50
	Secondary recycle (stream <19> & <20>)	mol/s	120.0
Gas generator	Isentropic efficiency compressor	-	0.88
	Isentropic efficiency turbine	-	0.88
	Mechanical efficiency	-	0.98
Boiler	Adiabatic temperature	°C	900
	Outlet temperature	°C	150
	Carbon content in ash[1]	wt%	2.0
	Heat loss	MW_{th}	2.0
ESP	Separation efficiency ash	wt%	100.0
Limestone injection	Total separation efficiency SO_x	wt%	95.0
	Limestone/Sulphur molar ratio	kg/kg	2.5
CO_2 compressor (five-stage)	Isentropic efficiency per stage	-	0.85
	Mechanical efficiency per stage	-	0.90

[1] CFB coal boiler, amount allows application of ash in construction materials

The natural gas composition is based on the standard composition of Norwegian North Sea natural gas (LHV = 46.88 MJ/kg); the coal composition appertains to Eastern Australian coal (LHV = 25.87 MJ/kg) as described in IEA-GHG report no. 2005/9 [6]. The prices of Norwegian North Sea natural gas and Eastern Australian coal were established at 5 and 2 €/GJ respectively. Addition of limestone to reduce SO_x emissions was not included during modelling to allow rapid convergence of the flowsheet; moreover the amount added is rather small. Moreover, the costs related to ash, limestone and gypsum disposal is omitted since application in construction materials is foreseen.

During modelling a CO_2 purity of 95.0 mol% is pursued, however this purity is governed by the stoichiometric ratio over the boiler, being 1.2 (20% O_2 excess). The secondary recycle is fixed at 120 mol/s to allow rapid convergence of the Fortran-based membrane model in Aspen Plus. The average oxygen permeation through the OCM amounts 2.5 g m^{-2} s^{-1} (10.5 nml cm^{-2} min^{-1}), which corresponds to recent experimental results by Vente et al. [7] upon correction for higher oxygen partial pressures at the feed side of the membrane in this particular case. An oxygen permeation of 10 nml cm^{-2} min^{-1} is generally accepted as lower boundary limit required for cost effective OCM operation. Heat transfer through the membrane is neglected.

The development of boilers that are specifically adapted for oxyfuel combustion in combination with OCM has a significant potential. Increasing the air in-leakage from 0 to 10%, results in an increase of the NO_x concentration in the CO_2 product stream from 6 to 65 ppm, due to thermal NO_x formation. However, the SO_x concentration decreases from 27 to 22 ppm upon increasing the air in-leakage, due to nitrogen dilution. The air in-leakage is neglected since a CFB-boiler is used.

CO_2 obtained from oxyfuel combustion can be further purified by cryogenic distillation. A small fraction of compressed CO_2 is expanded to facilitate sufficient cooling duty. The feed conditions of the mixture approach the triple point of CO_2. When cooled CO_2 condenses while incondensable compounds such as nitrogen and oxygen remain in the vapour phase. Expansion of 1.8 wt% of the captured CO_2 is reported to result in a CO_2 purity increase from 95.90 to 99.97%, moreover the increase in power requirement for CO_2 liquefaction amounts 5% [8]. The economic evaluation for each case was prepared both with (Table 2) and without [1] compression to 110 bar and purification by cryogenic CO_2 distillation.

4. Results

Based on the thermal input of coal and natural gas, the thermal efficiency of the oxyfuel boiler amounts 87.6% on LHV basis for Case 1. This relatively high efficiency is attributable to application of gas for air preheating, as well as the higher extent of heat recovery from the oxygen-lean air. The electricity required for the CO_2 compressor is not taken into account with respect to this efficiency, but solely to determine the operating costs and the overall amount of avoided CO_2 emissions. The stoichiometric oxygen ratio in the boiler amounts 1.20, which results in a CO_2 purity of 92.3 mol% for Case 1 without cryogenic CO_2 distillation. Total moisture removal would result in a CO_2 purity of 96.6 mol%.

The CO_2, H_2O, O_2 and N_2 concentrations as function of the stoichiometric ratio are displayed in Figure 2; these are similar for both cases. This chart demonstrates that relatively large stoichiometric ratios still allow high CO_2 purities, which is mainly ascribed to the absence of nitrogen permeation during oxygen production in the OCM (nitrogen present at the boiler outlet solely originates from coal-bound nitrogen). Moreover, relatively low boiler temperatures require a larger amount of inert gas at the boiler inlet; this is obtained by a larger primary recycle and subsequently results in an elevated CO_2 concentration at the boiler outlet. The latter poses an advantage for application of CFB boilers in comparison with pulverised coal boilers.

Additionally, oxygen production in an Air Separation Unit (ASU) typically contains 3-5% nitrogen, which predominantly builds up in the primary recycle over the boiler. This gives rise to research in coal combustion at low stoichiometric ratios: total combustion is reported at stoichiometric ratios of 1.01 to 1.03 [9].

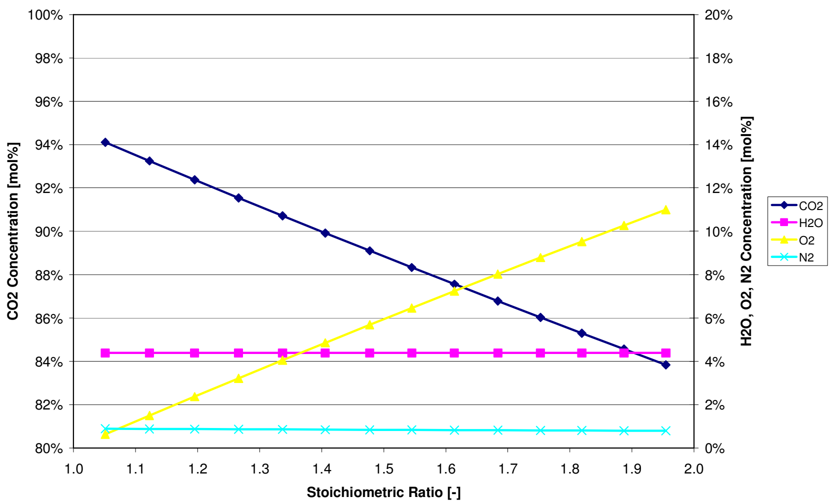

Figure 2 Stoichiometric oxygen ratio versus CO_2, H_2O, O_2 and N_2 concentrations downstream of knockout drum

The results of the economic evaluation are displayed in Table 2. It demonstrates that operation of an oxyfuel coal boiler with OCM is economically viable at market prices above €22.22 per tonne CO_2 avoided for case 1. The costs for to coal consumption slightly decrease in comparison with the reference case (no CO_2 capture). The costs for natural gas and electricity that are introduced upon carbon capture are more significant.

The results for the second case demonstrate that an oxyfuel coal boiler with OCM is economically viable at market prices above €21.46 per tonne CO_2 avoided. A third case is introduced to compare both cases with conventional MEA scrubbing, downstream of an air-fired coal CFB boiler. The MEA case proves to be economically viable at €70.31 per tonne CO_2 avoided.

Table 2 Economic evaluation oxyfuel coal boiler with OCM with CO2 purification and compression

		Case 1: Coal/Nat. gas	**Case 2:: All coal**	**Case 3: MEA**	**Reference w/o capture**	**Unit**
Coal (Eastern Australian)		1.827	2.231	3.062		kg/s
LHV Coal (Eastern Australian)		25870	25870	25870	25870	kJ/kg
Natural Gas (Norwegian)		0.209				kg/s
LHV Natural Gas (Norwegian)		46880				kJ/kg
Load		50000	50000	50000	50000	kW_{th}
Annual operation time at 100% capacity		7500	7500	7500	7500	h
Overall thermal efficiency		87.6%	86.6%	63.1%	87.0%	-
Fuel input		57078	57704	79226	57471	kW_{th}
Carbon capture ratio (from coal)		98.2%	98.2%	85.0%	-	-
CO_2 for storage (from coal)		4.304	5.104	6.043		kg/s
CO_2 emission combustion coal & NG		0.679	0.094	1.066	5.156	kg/s
CO_2 emission electricity consumption		0.483	0.551	0.761	0.104	kg/s
CO_2 avoided (net capture rate)		4.099	4.615	3.433		kg/s
Auxiliary power consumption		0.571	0.577	1.188	0.489	MW_e
CO_2 compressor duty		1.689	2.003	2.371		MW_e
Specific power CO_2 compressor		0.109	0.109	0.109		kWh_e/kg
Investment OCM		€ 2,217,109	€ 2,709,605			
Investment coal boiler		€ 15,362,111	€ 18,753,730	€ 25,748,483	€ 18,678,161	
Investment heat exchangers		€ 1,277,214	€ 1,797,389			
Investment compressor & turbine		€ 3,000,000	€ 3,663,783			
Investment CO_2 Compressor		€ 3,397,168	€ 3,841,379	€ 4,338,931		
Investment MEA Plant[1]				€ 15,000,000		
Total capital costs		€ 25,253.602	€ 30,764,438	€ 45,087,413	€ 18,678,161	
Operational costs						
Annuity	11%	€ 2,777,896	€ 3,384,088	€ 4,959,615	€ 2,054,598	€/a
O&M	4%	€ 1,010,144	€ 1,230,578	€ 1,803,497	€ 747,126	€/a
Electricity	0.05 (€/kWh)	€ 880,917	€ 1,030,493	€ 1,563,395	€ 183,190	€/a
Coal	2.00 (€/GJ)	€ 2,552,474	€ 3,116,004	€ 4,278,209	€ 3,102,508	€/a
Natural Gas	5.00 (€/GJ)	€ 1,324,403	€ 0	€ 0	€ 0	€/a
Costs per tonne CO_2 avoided		€ 22.22	€ 21.46	€ 70.31		€/tonne
Costs deducted		-€ 2,458,412	-€ 2,673,742	-€ 6,517,294		€/a
Total annual operational costs		€ 6,087,422	€ 6,087,422	€ 6,087,422	€ 6,087,422	€/a

[1] Derived from Singh et al. [10] upon downscaling; assumed thermal loss 3.6 MJ/kg CO_2 captured

The costs per tonne CO_2 avoided for Case 1 and 2 are more or less equal. The economic evaluation of case 3 (MEA) proves that coal-fired OCM boilers (Case 1 and 2) offer a large potential over commercially available technology. The economic evaluation of the cases presented in Table 2 is also calculated without CO_2 compression and purification; this allows application of the obtained results in the assessment of central CO_2 collection nodes, where further compression occurs. The avoidance costs amount € 10.26 per tonne CO_2 for Case1, € 9.01 per tonne CO_2 for Case 2 and € 45.58 per tonne CO_2 for Case 3.

The footprint of the OCM boiler in Case 1 is significantly larger than the footprint of the reference case, being 236 m^2 versus 48 m^2 respectively [1]. The required surface area is estimated at 150% of the calculated surface area, to allow inspection and maintenance of the unit operations.

5. Sensitivity analysis

The input parameters used for the calculation of the economic performance of each of these processes are subject to uncertainties. Sensitivity analyses are used to investigate the influence of the variations of input parameters on the CO_2 avoidance costs of the different capture processes studied. The input parameters (e.g. component costs, discount rate, etc.) are characterized by variability and/or uncertainty. The variability of a parameter refers to the range of that specific parameter and is determined by external conditions. The uncertainty of a parameter refers to the limited knowledge of the system under investigation (e.g. oxygen flux). For the purpose of the analysis, the economic evaluation of all three cases has been implemented in a stand-alone Excel workbook. The Excel add-in @risk module performs the analysis using Monte Carlo simulation. Distributions are assigned to the most relevant input parameters using the @risk module.

The input parameters with significant uncertainty and/or variability have been identified [1]. For each input parameter a probability distribution is defined that best approaches the variability or uncertainty of the parameter. The type of probability distribution that is chosen (normal, uniform, exponential) depends on the expected uncertainty or variability in the parameter values. To give an example, a normal distribution (also called Gaussian distribution) is the most common way to describe the uncertainty within a parameter. While the mechanisms underlying these phenomena are often unknown, the use of the normal model can be theoretically justified if one assumes many small (independent) effects contribute to each observation in an additive fashion. A uniform distribution, sometimes also known as a rectangular distribution, is a distribution that has constant probability. This type of distribution is used when no extra information is available or when all values within a range have equal probability (like for the discount rate). The first step of the sensitivity analysis is to define the type of distribution for each input parameter. The second step is to define the minimum and maximum values for each parameter. The basis for the specification of these minimum and maximum values is a general understanding of the status of technologies and components. For example, the range used to the probability distribution of costs for the coal boiler is assumed to be smaller than the range of the probability distribution for the innovative (and unproven) concept of the OCM. The probability distribution reflects that there is much uncertainty regarding the cost development of the OCM.

Figure 3 through 5 show the distribution of the CO_2 avoidance costs for Case 1, 2 and 3 respectively. Based on the assumed distributions of the input parameters, 90% percent of the economic evaluation results lie between 16.12 and 29.40 €/tonne CO_2 avoided. The distribution of the costs for Case 2 is slightly smaller than Case 1, the costs range from 17.28 to 27.67 €/tonne CO_2 avoided, while for Case 3 the distribution of the costs ranges from 54.78 to 90.57 €/tonne CO_2 avoided.

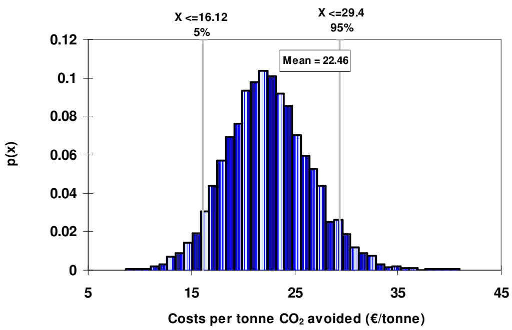

Figure 3 Distribution avoidance costs Case 1

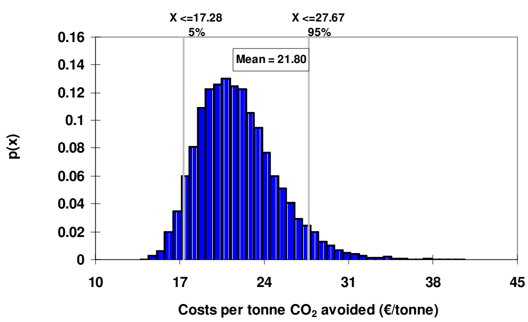

Figure 4 Distribution avoidance costs Case 2

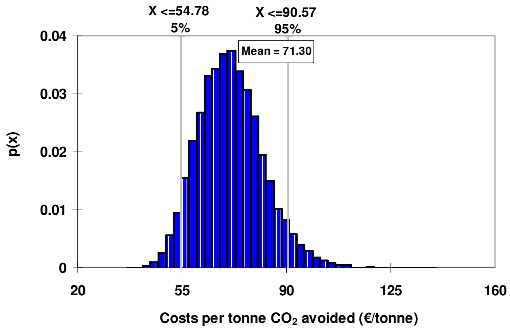

Figure 5 Distribution avoidance costs Case 3

The sensitivity of the costs per tonne CO_2 avoided (€/tonne) for selected parameters is analysed and presented by means of Tornado graphs. The input parameters are ranked in terms of their impact on the output parameter 'costs per tonne CO_2 avoided'. The graph shows which input distributions are 'significant' in determining the value of the output variable (in this case costs per CO_2 avoided). The ranking of the parameters has been done by applying regression analysis. The Monte Carlo simulation drops a variable if the impact on the output variable is close to zero.

The regression analysis for Case 1 shows that CO_2 avoidance costs are most sensitive to the price of natural gas. It has been assumed that the gas price varies between 3 and 7 €/GJ. The second largest impact on costs results from fluctuations in electricity price, which range has been set to 0.036 to 0.08 €/kWh. The regression coefficient of coal price and annual operation time is negative for both parameters, which implies that higher coal prices and increasing annual operation hours results in lower costs per tonne CO_2 avoided.

The price of coal and the costs per tonne CO_2 avoided are negatively correlated (correlation coefficient of -0.30). This negative correlation implies that higher coal prices affect the economics of the oxyfuel coal boiler in a positive way, since coal consumption and therefore the costs of coal are higher in the reference case without CO_2 capture. In the Case 1, part of the coal consumption is replaced by natural gas. Higher coal prices also result in lower prices per tonne of CO_2 avoided for Case 2, although the effect is much smaller. With increasing coal prices Case 2 becomes competitive to the reference case at lower prices per tonne CO_2 avoided. The same argument holds for the impact of annual operation time on the CO_2 avoidance costs.

When only coal is used to fuel the system (represented by Case 2) electricity prices have the strongest positive correlation with the cost per tonne of CO_2 avoided. Natural gas prices are not included since no natural gas is consumed in Case 2. While coal prices have a negative correlation with CO_2 avoidance cost in Case 1, in Case 2 the correlation of coal prices with the output parameter is positive (0.006). For Case 1, the effect of higher coal prices is small compared to the reference case, because part of the fuel consumption is fulfilled by natural gas. In Case 2 all coal is used and since capture also consumes energy coal consumption is slightly higher (57,704 kW_{th}) than in the reference case without capture (57,471 kW_{th}).

Several input parameters for Case 2 show a negative correlation to the output parameter costs per tonne of CO_2 avoided: annual operation time, estimated oxygen flux and coal price. Increasing values for these parameters result in lower costs per tonne of CO_2 avoided. For Case 3, both the plant lifetime and annual operation time are negatively correlated to the costs per tonne CO_2.

6. Conclusions

Coal-fired boilers with capture of CO_2 based on Oxygen Conducting Membranes may provide an effective way to reduce emissions of CO_2 to the atmosphere. In this detailed analysis two cases were assessed; Case 1 "natural gas and coal" using additional natural gas to preheat air and Case 2 "all coal" using an additional heat exchanger inside the coal-fired boiler to preheat air. A third case is introduced to compare both cases with conventional MEA scrubbing (Case 3).

The results of the economic evaluation of all cases show that Case 1 and 2 are almost equally attractive from an economic point of view. The difference in costs per tonne CO_2 avoided is less than 1 Euro. However, the capture ratio is significantly larger for case 2 (78% for case 1 versus 88% for case 2) and the uncertainty in costs is significantly smaller. For case 1, the costs are most strongly dependent on the price of natural gas. The variability of electricity prices has largest effect on CO_2 avoidance costs in case 2. The large uncertainty in the required surface area of the OCM makes that CO_2 avoidance costs are quite sensitive to changes of this parameter.

Adversely, the capital investment of the OCM boiler is significantly lower for case 1 compared with case 2, being 25 M€ versus 31 M€ respectively. Moreover, the development of the gas-to-gas heat exchanger to be positioned inside the coal-fired boiler as applied in Case 2 is still in a premature phase. Particulate fouling (CFB-boiler) and slag corrosion (PC-boiler) of these heat exchangers remain substantial challenges to overcome.

Furthermore it should be noted that the Oxygen Conducting Membranes with elevated oxygen permeation (2.5 g m^{-2} s^{-1}) only exist on lab-scale at present, while scale-up and increased membrane stability is required to facilitate technical and economic viability in advanced energy conversion systems. It might require several years before these membranes can be used on the described scale.

However, the application of Oxygen Conducting Membranes for oxygen production in oxyfuel coal boilers results in elevated CO_2 purities compared with conventional oxygen production in Air Separation Units. This is mainly attributable to the high purity of the produced oxygen, which significantly reduces the nitrogen build-up in the primary recycle over the coal boiler. As a consequence a coal boiler with OCM can be operated at higher stoichiometric ratios than a coal boiler with an ASU, both resulting in equal CO_2 purities and under the assumption that air leakage into the boiler can be neglected.

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