Modeling and Analysis

GREENHOUSE GASES SCIENCE AND TECHNOLOGY

Life-cycle performance of natural gas power plants with pre-combustion CO_2 capture

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Abstract: CO_2 capture and storage involves technologies that separate, capture, and store CO_2 from large facilities, such as fossil fuel power plants. Although it is a promising measure to meet environmental standards on carbon pollution, proposed technologies in power plants are energy demanding and decrease the energy generated per unit of input fuel when compared to business-as-usual scenarios. In this paper, we evaluate the environmental performance of two similarly structured combined-cycle power plants with pre-combustion capture. The first power plant performs methane steam reforming in an autothermal reformer, while the second plant uses a reactor that includes a hydrogen-separating membrane. The two plants are compared both to one another and to a business-as-usual scenario using six environmental impact potentials (abiotic depletion, global warming, ozone layer depletion, photochemical oxidant formation, acidification, and eutrophication). The goal is to pinpoint environmental weaknesses and strengths of the two capture technologies. We find that the two plants result in similar impacts, decreasing the contribution to global warming of conventional operation but, at the same time, increasing other impacts, such as ozone layer depletion and photochemical oxidant formation. Additionally, the two capture plants result in higher cumulative non-renewable and total energy demands, as well as in lower life-cycle energy balances and efficiencies. The most direct measure to decrease the environmental impacts of the examined techniques would be to increase their efficiency, by decreasing the requirements of the processes in natural and energy resources.

Keywords: autothermal reforming; CO_2 capture; life cycle assessment; membrane reactor; pre-combustion; steam reforming

Introduction

Electricity generation through the combustion of fossil fuels in power stations is associated with high emissions and environmental pollution. Power plants that include systems for separating and capturing the generated emissions (CO_2 capture and storage, CCS) provide a possibility of generating clean energy and decreasing the anthropogenic influence on the climate.[1]

CO_2 capture is classified into post-, pre-, or oxy-combustion technologies.[2–5] In pre-combustion

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capture, i.e., the focus of this work, the carbon included in the fuel is captured before the combustion process and, in this way, the combustion is performed with a relatively clean, hydrogen-rich fuel that reduces harmful emissions. The most common application of pre-combustion method for CO_2 capture is the gasification of coal and the combustion of the generated syngas in a combined-cycle power plant. Although integrated gasification combined cycles (IGCCs) operate with a relatively high efficiency,[6–8] they are complex and associated with numerous operational challenges and high investment costs.[9] Overall, the implementation of pre-combustion CO_2 capture technologies both in coal and natural gas power plants requires either the construction of new facilities or significant modifications of already existing, conventional power plants.

Although CCS is an attractive choice for decreasing emissions from energy generation, processes like the separation itself and the compression of the captured CO_2 are energy intensive and decrease the operating efficiency of a power station.[1,10,11] Additionally, the incorporation of new components increases the associated investment and operating costs.[1,12–15] Because newly introduced processes for CCS make the overall evaluation of power plants more complex, thermodynamic and economic analyses of such plants must be accompanied by a parallel evaluation of their environmental performance.[16]

In this paper, we evaluate and compare the environmental performance of natural gas-fired power plants with two different pre-combustion capture technologies, along with a business-as-usual scenario. The analysis presented herein is based on similarly structured plants simulated in detail using the software EBSILONProfessional, software suitable for the simulation of thermodynamic processes.[17] The operation of the power plants with CO_2 capture is based on methane steam reforming.[18] The environmental evaluation is conducted using the life cycle assessment (LCA) methodology.

LCA is used as a tool for comprehensively calculating the environmental performance of the examined plants and determining whether the involved technologies are worthy of implementation or whether they should be further improved before implementation. LCA is a well-established and standardized methodology for the comprehensive evaluation of products with a holistic perspective;[19,20] it has already been used for the environmental evaluation of other CCS systems.[21–23] Here, in addition to the life-cycle environmental performance of the evaluated power plants, we also calculate their cumulative energy demands[24] and life-cycle energy balances and efficiencies, in order to assess the viability of the capture technologies and compare them with a conventional case. The present paper supplements previously presented analyses of CCS systems, since the life-cycle environmental and energy assessment of the proposed natural-gas-fired power plants with two different pre-combustion capture technologies had not yet been addressed.[1]

Material and methods

Goal and scope

In this work we evaluate the life-cycle performance of two combined-cycle systems for power generation with pre-combustion CO_2 capture and that of a similarly structured reference plant without emission control (business-as-usual scenario). The potential impacts of the power plants with CO_2 capture are compared both to one another and to those of the reference power plant. The simulations of the power plants have been realized with the software EBSILONProfessional used in the simulation of thermodynamic processes.[25] The reasons for choosing this specific software were its proven accuracy, operational support, and user-friendly environment.[17]

The reference plant (Fig. 1(a)) is a conventional combined-cycle power plant operating with natural gas and generating approximately 410 MW of electricity. The Rankine Cycle of the plant includes three pressure levels and one reheat stage. Detailed structural and operational characteristics of the reference power plant can be found in Petrakopoulou *et al.*[26]

Simplified flow diagrams of the pre-combustion power plants are shown in Figs 1(b) and 1(c). In both pre-combustion plants a hydrogen-rich fuel is generated and is subsequently burned in the combustion chambers of the plants. The CO_2 generated during the reforming process is separated and compressed.

The first power plant examined (MSR-H2, Fig. 1(b)) operates with a membrane reformer, in which methane steam reforming takes place (with a mass ratio methane/vapor 1:4). The modeling and thermodynamic limits of hydrogen-separating membranes were studied by Marigliano *et al.* that also presented a similar membrane reactor design to the one presented in this paper.[27,28] The simulation and

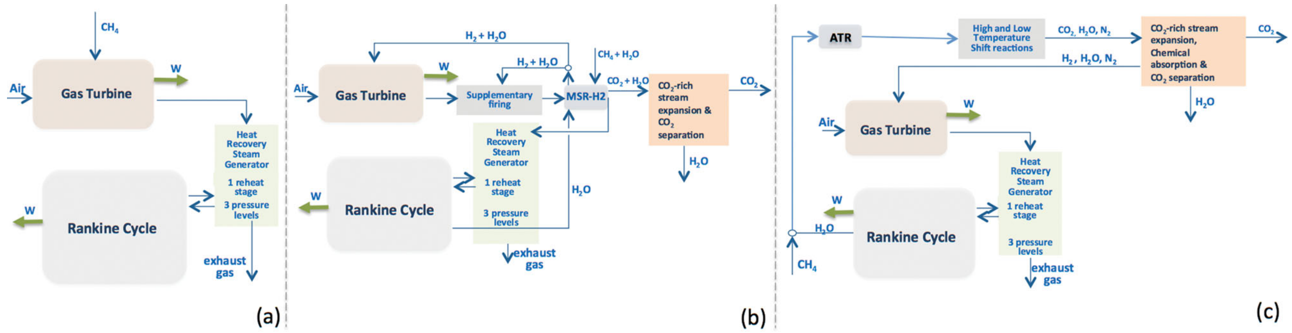

Figure 1. Simplified diagrams of the reference plant (a), MSR plant (b), and ATR plant (c).

application requirements of membrane reactors for H_2 separation in IGCC plants have also been investigated, by, for example, Koukou *et al.* and Amelio *et al.*[29,30] However, these studies assume stream concentrations and operating conditions that differ from the ones assumed in the present paper. For the purpose of the present work, the membrane is programmed to function in a steady-state mode under the specific operating requirements of the power plant presented. The simulation of the membrane reactor was based on data derived from Jordal *et al.*[31] (adjusted to the operating conditions) who studied conditions relevant to the pressure and temperature requirements of gas turbine power plants with CO_2 capture.

In the Pd-based membrane, 99.8% of the incoming methane is reformed and 99% of the produced CO is shifted.[32] The thermal energy required for the reforming process is provided from the combustion gases the temperature of which is increased to about 960°C in the supplementary firing of the plant.[31] Before the mixture of methane and water vapor is led to the membrane, it is preheated from the combustion gases in a gas/gas heat exchanger.

The generated hydrogen is swept by intermediate-pressure steam and led to the combustion chamber of the gas turbine system of the plant. The sequence of H_2 separation steps has been based on the work of Jordal *et al.*,[31] until a separation of 99.7% of the generated hydrogen is achieved. The CO_2 generated in the reforming process undergoes a four-level inter-cooled compression and it is separated via water condensation.

In the second power plant (ATR, Fig. 1(c)) a mixture of natural gas and water vapor is led to an autothermal reformer, where the methane is reformed at a temperature and pressure of 850°C and 15 bar, respectively. The energy required for the reforming of the methane in this component is provided by both partial and complete combustion (with air extracted from the gas turbine of the plant).

At the exit of the autothermal reformer, the generated gases undergo high- and low-temperature shift reactions in order to convert the produced CO into CO_2 and H_2. However, because the exiting gas includes a considerable amount of nitrogen that complicates the separation of the included CO_2, chemical absorption is realized (chemical absorption unit, CAU).[33,34] In the CAU[9], 85% of the CO_2 included in the stream is separated and the thermal energy required for the regeneration of the chemical solvent (monoethanolamine, MEA) is provided from a low-pressure steam extraction. After the CAU, the captured CO_2 is led to the compression unit, while the hydrogen-rich gas (fuel) is sent to the combustion chamber of the gas turbine system of the plant.

More details on the operation of the evaluated plants can be found in Petrakopoulou and Tsatsaronis.[25]

The functional unit (FU) of the LCA of the three considered power systems is 1 kWh of net electricity product (at plant). A cradle-to-gate approach is followed, and capital goods are excluded from the study.

In the simulations, the power plants are fed with the same fuel quantity and generate different amounts of electricity, depending on their operational efficiency. For the LCA, the product of the systems is kept constant. This means that the fuel inputs of the plants are adjusted to compensate for different thermodynamic efficiencies. In other words, when the efficiency of power plant A is lower than that of power plant B, power plant A will need more fuel to generate the same amount of electricity.

Six environmental impact potentials are evaluated: abiotic depletion (ADP), global warming (GWP), ozone layer depletion (ODP), photochemical oxidant formation (POFP), acidification (AP), and eutrophication (EP). These impact potentials are quantified using the CML method,[35] with the exception of GWP, which is evaluated according to the characterization factors (100-year period) reported by the Intergovernmental Panel on Climate Change (IPCC).[36] In addition to these environmental impact categories, the cumulative non-renewable energy demand (CED_{nr}) and the total cumulative energy demand (CED_t) of each system are also calculated.[24] The CED_{nr} involves fossil and nuclear energy, whereas the CED_t includes renewable and non-renewable energy. The selected categories and evaluation methods can be characterized as well-established, since they correspond with common choices in LCA studies of energy systems.[23,37] The software SimaPro 8 has been used to facilitate the calculation of the life-cycle impacts.[38]

To further examine the life-cycle performance of the alternative plants, we also calculate their life-cycle energy balances and efficiencies. The life-cycle energy balance of each system is calculated as the difference between the potential energy output (i.e., $1\ \text{kWh}\cdot\text{FU}^{-1} = 3600\ \text{MJ}\cdot\text{FU}^{-1}$) and the CED_{nr} indicator of each power system.[23,39,40] In this respect, a positive balance is desired, as it indicates a system whose energy production exceeds its non-renewable (fossil and nuclear) energy requirements. The life-cycle energy efficiency of each system is estimated by dividing the potential energy output with the CED_t indicator. Unlike the life-cycle energy balances that usually only take the fossil and nuclear energy demand into account (in order to not penalize the use of renewable resources), the efficiencies consider the total (renewable and non-renewable) energy required by the system.[37]

Data acquisition

Table 1 presents a selection of key inventory data for each of the systems. The main inventory data were obtained from the process simulation of the systems using EBSILONProfessional. Analytical tables can be found in the literature.[25,26] Data for natural gas production and background processes were taken from the ecoinvent database.[41,42]

The higher CO_2 emissions of the ATR plant are justified by the less effective separation process used in the plant, i.e., the chemical absorption unit. The necessary chemical absorption in the ATR plant (due to the coexistence of N_2 and CO_2 in the flue gases) separates only 85% of the generated CO_2. In contrast, the CO_2 separation of the MSR plant is more effective, since the flue gases consist of only CO_2 and water vapor.

Table 1. Main inventory data for each power system (values per FU: 1 kWh of net electricity, at plant).

	Reference plant	MSR plant	ATR plant
INPUTS			
From the environment			
Air (kg)	5.36	5.63	6.26
Water (kg)	0.54	0.49	0.45
From the technosphere			
Natural gas (g)	122.18	150.63	148.32
Water (kg)	—	0.85	0.33
Catalysts (mg)	—	0.28	0.92
MEA solution (g)	—	—	2.72
OUTPUTS			
Products			
Electricity (kWh)	1.00	1.00	1.00
Carbon dioxide (kg)	—	0.41	0.31
Waste to treatment			
Catalysts (mg)	—	0.28	0.92
Wastewater (kg)	0.65	0.51	0.51
Emissions to the air			
CO_2 (g)	337.93	2.80	101.88
NO_x (g)	0.45	0.38	0.49
H_2O (kg)	0.31	0.98	0.51
MEA (g)	—	—	1.09

Results and discussion

The life-cycle inventories of the three power systems were implemented into SimaPro 8.[38] The results of the environmental characterization of the power systems are presented in Table 2. These life-cycle impact profiles are entirely allocated to the net product of the plants, i.e., the electricity generated.

Figure 2 compares the life-cycle impacts of the three systems. It can be seen that while the life-cycle performance of the MSR and ATR plants is found to be significantly more favorable than that of the reference plant in terms of the GWP (79% and 54%

Table 2. Characterization results of the power systems (values per FU: 1 kWh of net electricity, at plant).

	Reference plant	MSR plant	ATR plant
GWP (kg CO_2 eq)	0.40	0.08	0.19
ADP (kg Sb eq)	$3.49 \cdot 10^{-3}$	$4.30 \cdot 10^{-3}$	$4.28 \cdot 10^{-3}$
ODP (kg CFC-11 eq)	$5.02 \cdot 10^{-8}$	$6.19 \cdot 10^{-8}$	$6.13 \cdot 10^{-8}$
POFP (kg C_2H_4 eq)	$1.91 \cdot 10^{-5}$	$2.36 \cdot 10^{-5}$	$2.39 \cdot 10^{-5}$
AP (kg SO_2 eq)	$4.50 \cdot 10^{-4}$	$4.67 \cdot 10^{-4}$	$5.31 \cdot 10^{-4}$
EP (kg PO_4^{3-} eq)	$8.42 \cdot 10^{-5}$	$7.74 \cdot 10^{-5}$	$9.69 \cdot 10^{-5}$
CED_{nr} (MJ)	7.15	8.81	8.77
CED_t (MJ)	7.16	8.82	8.79

lower, respectively), the capture plants are generally associated with more detrimental impacts in the rest of the categories evaluated. Despite the improved GWP results, the comparable AP of the MSR and reference plants, and the relatively lower EP of the MSR plant due to its reduced direct NO_x emissions, the two power plants with CO_2 capture generally show worse performance when compared to the business-as-usual scenario. Similar observations (e.g. regarding GWP, ADP, ODP, and CED results) have been reported in scientific literature when comparing other type of CCS systems with one another or with conventional power plants.[23]

The ATR plant results in higher AP and EP values than both the reference and MSR plants. This is mainly due to the increased use of natural gas (when compared to the reference plant) and the increased direct NO_x emissions (compared to both the reference and MSR plants).

The ADP, indicating the depletion of abiotic resources, is found to be similar for the two pre-combustion plants, with the ATR plant performing slightly better. This is due to the relatively higher efficiency of the ATR plant, as well as to the lower amount of CO_2 separated in the process. These two factors result in a lower natural gas demand for power generation and lower electricity requirements for CO_2 compression.

Furthermore, the ATR plant results in a somewhat higher POFP value when compared to the MSR plant. This stems from the additional use of the MEA solvent in the ATR plant.

Overall, the MSR and ATR plants show similar results for the environmental impact categories ADP, ODP, and POFP, while the first shows significantly lower GWP, EP, and AP values. Lastly, as also seen in Fig. 2, the two capture plants result in similar values of CED_{nr} and CED_t.

Normalization of the life-cycle impacts provides insight into the relative magnitude of each indicator.[19] This is particularly useful for examining, for example, the benefit of GWP mitigation at the expense of other increased impacts. When normalizing according to the factors for the world in 1995,[35] it is found that ADP is the category with the highest relative impact. Therefore, even though the normalized results for GWP are

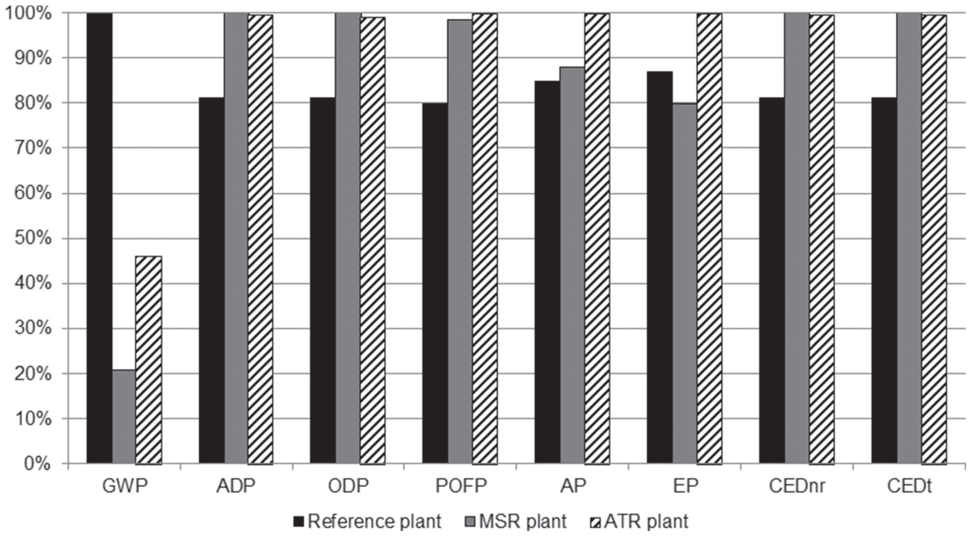

Figure 2. Comparison of the life-cycle environmental profiles of the power systems.

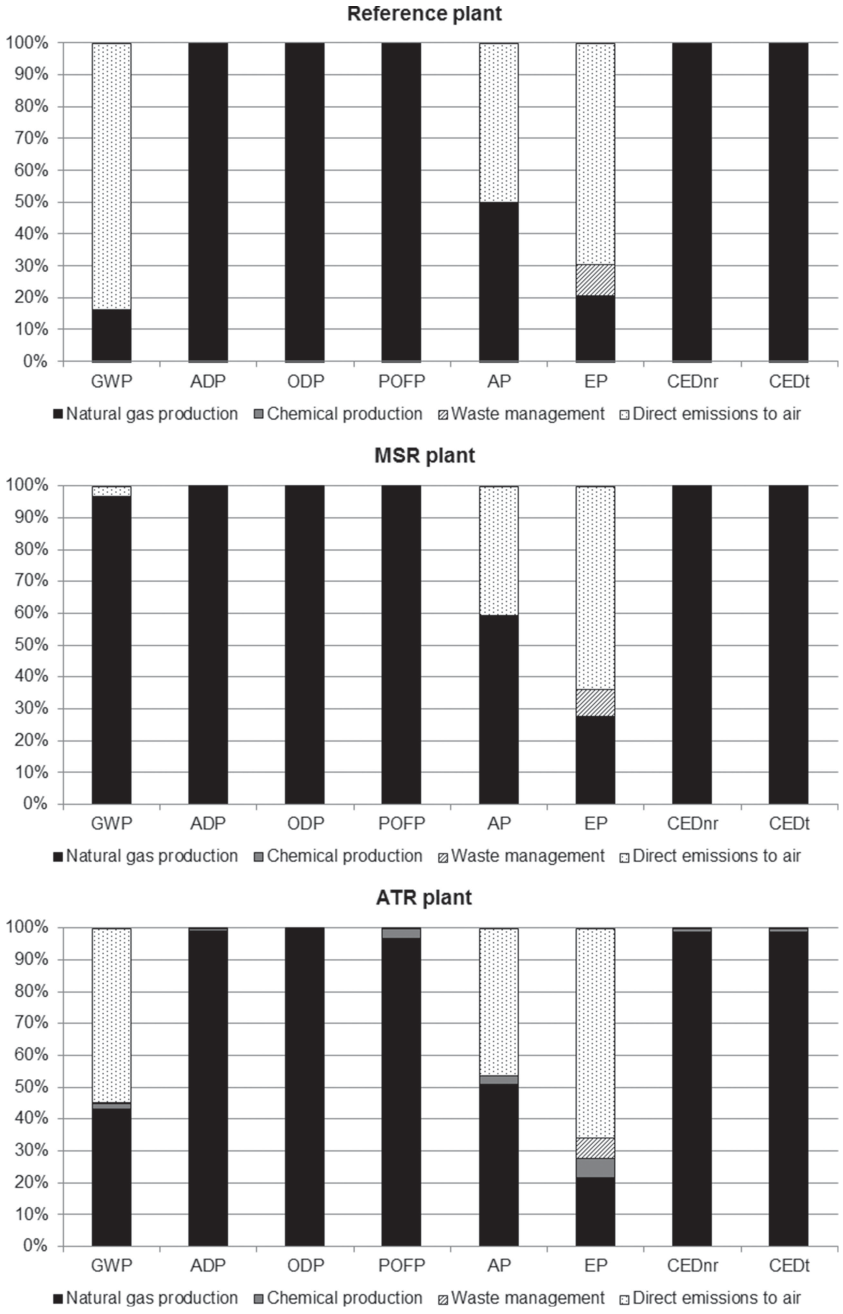

Figure 3. Process contribution to the impact categories for each system.

also found to be relevant (in fact, GWP is ranked second for the set of environmental impact categories), the concerns about the suitability of the evaluated systems – as previously shown by the characterization results – remain. It should be noted that no weighting procedure has been applied to the impacts, because such a weighting would be inherently subjective.

To further justify the results and comprehend the environmental behavior of the plants, the main processes responsible for the evaluated impacts are identified. As shown in Fig. 3, the same two leading sources of impact are identified for the three systems: natural gas (with varying contributions depending on the impact category) and direct emissions to the air (with significant contributions to GWP, AP, and EP). Overall, these two aspects clearly arise as the two major environmental impact sources, overshadowing the contribution of the remaining processes to the

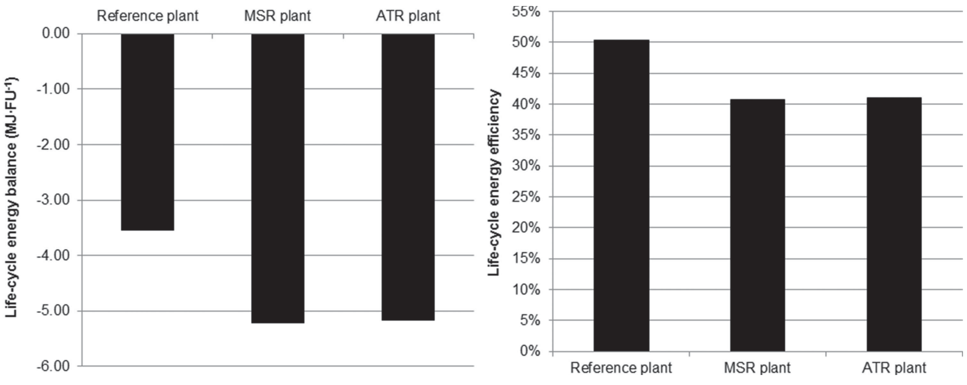

Figure 4. Life-cycle energy performance of the power systems in terms of life-cycle energy balance and efficiency.

potential impacts. Regarding GWP, direct emissions to the air are found to be the main source of impact in the reference plant. The same is true in the case of the ATR plant (due to the relatively low percentage of CO_2 captured in the plant), while in the MSR plant emissions play a secondary role.

The sensitivity of the results to variations in the inventory elements can be determined from the analysis of the processes contributing to the impacts (Fig. 3). In this respect, relevant changes in the natural gas input would dramatically affect the evaluated impacts. Additionally, important changes in the emission levels (direct emissions to the air) would significantly affect the GWP, AP, and EP results.

Figure 4 shows the comparison of the life-cycle energy performance of the different power systems. Unfavorable (i.e., negative) balances are obtained for all three systems since their CED_{nr} indicators exceed the energy output of the electricity produced, with the reference plant showing the best result. Additionally, when compared to the reference plant, the MSR and ATR plants also perform worse in terms of life-cycle energy efficiency due to higher CED_t values. These results are strongly driven by the increased natural gas consumption in the MSR and ATR plants required to generate the same amount of electricity. These findings agree with the results of the exergetic efficiency reported for these combined-cycle power plants in the literature.[25]

Conclusions

This paper presented the LCA of two power plants with pre-combustion CO_2 capture and of a reference combined-cycle power plant (business-as-usual scenario without emission control). Both capture plants were based on methane steam reforming, the first incorporating a reactor including a hydrogen-separating membrane and the second an autothermal reformer. These power plants were compared both to each other and to the business-as-usual scenario.

It was found that the two capture power plants decrease global warming significantly, but at the same time, present a potentially significant negative impact on abiotic depletion, ozone layer depletion, and photochemical oxidant formation. Unlike the autothermal reformer plant, the acidification and eutrophication potentials of the membrane plant (associated with lower direct NO_x emissions) were at comparable levels to those of the business-as-usual case.

Although the cumulative non-renewable and total energy demands of the power plants with CO_2 capture were found to be similar, they were significantly higher than those of the reference plant. Hence, the life-cycle energy balances and efficiencies of the capture plants were found to be less favorable than those of the reference case.

From the results presented, it was seen that in order for the two examined pre-combustion technologies to become more attractive environmentally, the use of natural gas per unit of generated electricity should be significantly reduced and any additional energy requirements should be kept to a minimum.

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