

Implementation of Chemical Looping Combustion technology with Waste-Derived Fuels: Process analysis and comparison with other prominent CO₂ capture technologies

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Abstract

Solid wastes management constitutes an unavoidable issue in modern overconsuming societies, but apart from that, it is also an energy source. Combustion of waste, like other carbonaceous fuels, emits carbon dioxide, which needs to be mitigated in order to achieve the Paris Agreement targets, concerning the limitation of carbon dioxide emissions in the atmosphere. The CCS implementation at waste to energy (WtE) plants is an attractive strategy to achieve this. Chemical Looping Combustion (CLC) is considered as a very promising combustion for power plants to produce efficiently thermal energy, given that it includes an inherent CO₂ capture, avoiding in this way the cost or energy penalties that accompany other existing CO₂ capture technologies. In this study, the integrated model of a CLC unit fuelled with waste derived fuel for power production with the simultaneous CO₂ capture in an effective way is presented. Several aspects that affect the overall plant efficiency such as the heat recovery configuration, the steam pressure level and the fuel type are assessed. Moreover, the CLC is benchmarked with other two competitive CO₂ capture technologies, amine scrubbing and calcium looping.

Keywords: Chemical looping combustion; waste to energy plants; CO₂ capture; process modelling; Calcium looping; amine scrubbing

Introduction

Chemical Looping Combustion (CLC) is an emerging technology for waste incineration, with an inherent CO₂ capture. It is characterized by low energy penalty and high CO₂ capture efficiency. The basic idea behind the Chemical Looping Combustion technology lies in the recirculation of an oxygen-carrier material between two interconnected fluidized bed reactors, reacting with atmospheric air in the one reactor (Air Reactor) and with the feedstock in the other (Fuel Reactor). The oxygen-carrier material, usually in the form of a metal-oxide [1], has the ability to transfer oxygen from the atmospheric air to the fuel, therefore avoiding the formation of nitrogen oxides (NO_x) and leading to the production of almost pure CO₂ and H₂O in the Fuel Reactor. After condensation of the Fuel Reactor flue gas, vapour is removed and hence CO₂ can be captured and transported or further utilised. The Air Reactor flue gas consists mostly of N₂ and O₂ that did not react with the oxygen-carrier. What is more, apart from the fuel, a gas stream enters the Fuel Reactor needed for its fluidization, which is usually either steam or recirculated CO₂. Chemical Looping Combustion is a rather new and up-and-coming technology, so there is not so much experience yet with using waste as fuel. However, since waste incineration has been conducted already in fluidized beds in Waste-to-Energy plants, the transition to CLC is not expected to pose such a big challenge. The process flowsheet diagram of a simplified CLC system is depicted in Figure 1.

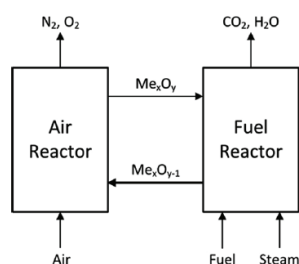


Figure 1: Schematic diagram of the Chemical Looping Combustion process (source: [2])

Regarding the post-combustion techniques for CO₂ capture, chemical absorption in an aqueous MEA solution is widely considered as the most mature available technology. In this process, CO₂ reacts with the solvent in an absorber column, forming chemical dissolved compounds. Subsequently, the solvent is regenerated in a stripper column due to the reversal of the chemical reactions at higher temperatures [3]. The necessary heat duty for the MEA regeneration is delivered by steam extraction of the initial power plant's Rankine cycle.

Calcium looping (CaL) is a promising post-combustion technology. This process relies on two reversible chemical reactions: carbonation and calcination. The first one occurs in a carbonator reactor, where CO₂ is captured by reacting with solid lime (CaO) and limestone (CaCO₃) is formed. Afterwards, the sorbent is regenerated in the calciner and CO₂ is released and purified in a PCU [4]. As the calcination is an endothermic reaction, fuel (RDF) combustion is necessary to maintain a constant temperature whereas pure O₂ is used as oxidizing agent in order to achieve high CO₂ concentrations in the calciner flue gas.

Several studies and projects can be found in the literature that are dedicated in the implementation of CO₂ capture technologies in Waste-to-Energy plants. In [5], Fortum Oslo Varmer AS used Shell's proprietary amine-based solvent DC-103 (previously untested in WtE flue gases) as a CO₂ capture technology in a WtE plant in Oslo and achieved high carbon-capture efficiency (around 90-95%) while also diminishing its amine emissions (fewer than 0.4ppm) and operating successfully for over 5000 hours. In Saga city, Japan, Toshiba has constructed a CCU facility, using an alkaline aqueous amine to capture CO₂ from a WtE plant situated nearby, accomplishing capturing up to 10 tons of CO₂ every day and further utilising it to cultivate crops and algae [6], [7]. Martin Haaf et al. in [8] presented the results of a 1MW_{th} CaL pilot plant, using SRF as fuel in the calciner, which captured carbon dioxide from the flue gases, provided by the combustion of pulverized coal or natural gas. In [9], the implementation of a CaL unit with a WtE plant was studied, with SRF, natural gas, and coal being tried out as the additional fuel. Their study showed that the use of SRF achieved the lowest net electrical efficiency out of them all. In [10], a process simulation was carried out for a 60MW_{th} CLC plant, fired by SRF, which achieved a very high carbon capture efficiency of 97% and had an oxygen demand of 17%, in order for the flue gas to be fully converted in the post-oxidation chamber. What is more, Yaqub et al., in [11] conducted experiments with plastic and paper waste in a batch fluidized-bed reactor, which showed that the paper waste had a higher fractional conversion of CO to CO₂, due to its augmented volatile content.

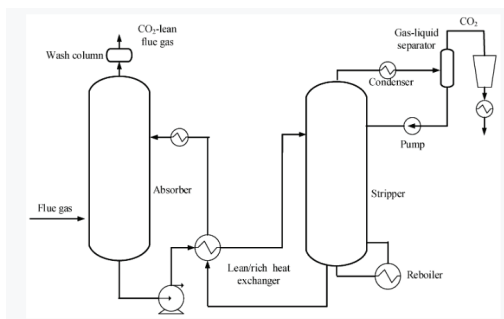


Figure 2: Amine Scrubbing process

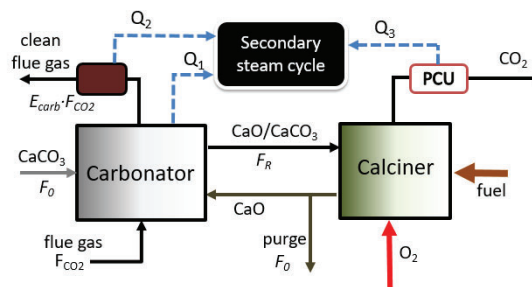


Figure 3: Calcium Looping process

This study analyses the implementation of the CLC technology with waste-derived fuels. The main purpose of this paper is to indicate which of the three abovementioned carbon capture technologies is more energy effective, taking into account in each case the net electricity production derived from the RDF combustion.

2. Model description

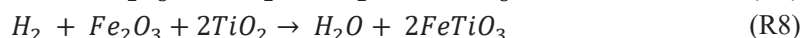
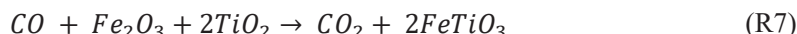
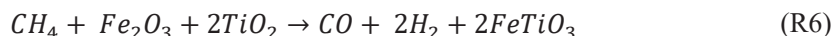
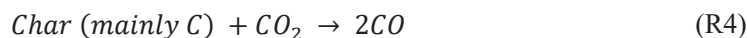
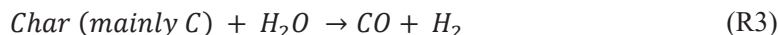
2.1 Chemical Looping Combustion

The process simulations for all technologies are performed with ASPENPlusTM. The properties of the RDF fuel used in all cases are summarized in Table 1.

Table 1. Fuel properties

C (% d.b.)	H (% d.b.)	O (% d.b.)	N (% d.b.)	S (% d.b.)	Cl (%) d.b.)	Ash (% d.b.)	Moisture (w/w %)	LHV (kJ/kg w/w)
40.03	4.1	24.9	0.47	0.19	0.51	29.8	27.8	9598.9

In CLC with solid waste, after the feedstock enters the Fuel Reactor, devolatilization and gasification of char take place, the products of which react with the oxygen-carrier. The reduced particles of the oxygen carrier (here ilmenite) are then transported to the Air Reactor to be oxidised, carrying some unconverted char particles with them. That char, if it reaches the Air Reactor, is oxidised by the air and transformed into CO₂, which will later on be emitted in the atmosphere. Both reactors are modelled as an RSTOIC in the ASPEN flowsheet. The reactions taking place in the AR are the (R1) and (R2), while the reactions (R3) until (R8) are the ones occurring in the FR:



The rates of the reactions taking place in the FR were defined through 'Design Specs', so that the final composition of the FR exhaust gases would be in accordance with the composition of the

corresponding gases from an application of a CLC plant with biomass (see Table 2). Given that there aren't many literature sources regarding CLC operation with SRF, and that SRF has a large biogenic fraction, the admission was made that these two fuels should have a similar performance. Regarding the impurities such as SO₂, NO and HCl that form in CLC applications with SRF, these were produced in a separate RSTOIC in the ASPEN flowsheet, preceding the FR, the reaction rates of which were based on data from different literatures sources. It should be stated that in reality, all these reactions take place in one reactor (the FR). In the AR, both the char and the oxygen-carrier were considered to convert fully to their products in the reactions (R1) and (R2) respectively.

In order to reduce the amount of these CO₂ emissions, a Carbon Stripper is used, separating the char particles from the oxygen-carrier and returning them back to the Fuel Reactor to be gasified. Unconverted gases in the FR flue gas can be fully oxidised to CO₂ and H₂O in a Post-Oxidation Chamber (POC), using a pure stream of O₂. The thermal energy of the flue gases exiting the two reactors is used for the production of electricity in an adjoining Rankine Cycle and the preheating of air and steam needed in the AR and the FR respectively. For the superheating of steam, the heat of the AR flue gas is used, given that it is a stream clean from impurities, hence overcoming the problem of possible corrosion faced in most Waste-to-Energy plants, which limits the superheated steam temperature to 400°C. Therefore, the steam in this model was able to be superheated to a temperature of around 520°C and expanded in 3 stages in steam turbines [1]. The FR flue gas, being rich in CO₂, after being cooled down to just before its dew point in the Rankine Cycle, is purified and gradually compressed and condensed to reach the appropriate conditions for CO₂ capture and delivery [12]. On the other hand, the AR flue gas, after being cooled down to around 80°C, consisting mainly of N₂, O₂ and CO₂, is released in the atmosphere, therefore constituting the only CO₂ emissions of the CLC system.

Regarding the CO₂ emissions of the plant, the carbon capture efficiency was introduced, calculated as the amount of carbonaceous gases (measured in kmol/s) in the off-gas that is captured to the amount of the total carbonaceous gases both captured and emitted to the atmosphere.

$$\eta_{CC} = \frac{(F_{CO_2} + F_{CO} + F_{CH_4})_{FR \text{ flue gas}}}{(F_{CO_2} + F_{CO} + F_{CH_4})_{FR \text{ flue gas}} + (F_{CO_2})_{AR \text{ flue gas}}} \quad (1)$$

Table 2: Main characteristics of the CLC case

Temperature in FR / AR (°C)	950 / 1000
Feedstock mass flow (kg/sec)	5.21
Oxygen-carrier mass flow (kg/sec)	250
FR flue gas mass flow (kg/sec)	8.29
AR flue gas mass flow (kg/sec)	11.22
Supercritical steam conditions (°C / bar)	520 / 80
Total feedwater mass flow (kg/sec)	12.17
O ₂ needed in POC (kg/sec)	1.22
AR outlet flue gas composition (%vol)	97% N ₂ , 2% CO ₂ , 1% O ₂
FR outlet flue gas composition (%vol)	28% CO ₂ , 4% CO, 56% H ₂ O, 8% H ₂ , 3% CH ₄ , 1% impurities (SO ₂ , NO, HCl, N ₂)
FR outlet flue gas composition range requirements (%vol d.b.) [13]	65-70% CO ₂ , 10-12% CO, 11-18% H ₂ , 5-8% CH ₄

2.2 Reference Case

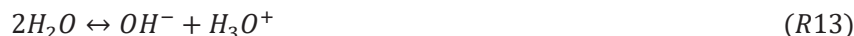
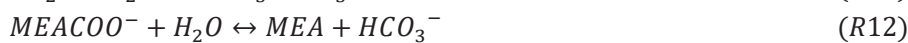
As for the reference case, a typical Waste-to-Energy plant is considered. This plant consists of a typical WtE supercritical boiler that produces steam at 400 °C in order to avoid corrosion problems [9], two reheaters and three steam turbines at escalated pressures. The feedwater preheating is performed using part of the thermal content of the flue gas. The main characteristics for the reference plant are summarized in Table {3}:

Table 3: Main characteristics of the reference case

Boiler efficiency (%)	93.6	HP/IP/LP Turbine isentropic efficiency (%)	92/94/88
Turbine inlet HP/IP/LP pressure (bar)	40/17.5/6.3	Superheated steam temperature/pressure (°C/bar)	400/40
Condenser pressure (bar)	0.077	Feedwater temperature (°C)	143.6
Flue gas outlet temperature (°C)	330		

2.3 Chemical Absorption with MEA

In MEA scrubbing, flue gas is in countercurrent flow with an aqueous MEA solution in an equilibrium absorption column that operates in atmospheric pressure. CO₂ separation from the flue gas is conducted via the following exothermic reactions [3]:



Subsequently, the abovementioned reactions are reversed and the solvent is regenerated in a stripper column. The necessary heat for solvent regeneration is given from low pressure steam (3.09 bar), which is extracted from the second turbine of the reference plant. The whole process is simulated and ELECNRTL is being used as property method. Variables like reboiler temperature (has to be lower than 120 °C in order to avoid thermal degradation), amine loading, etc. are presented in Table 4:

Table 4: MEA scrubbing key parameters

<i>MEA scrubbing</i>	
Lean solvent w/w	30 %
Lean solvent temperature	40 °C
Lean solvent loading	0.2
Rich solvent loading	0.49
L/G ratio	3.11 kg/kg
Specific heat duty	3.74 MJ _{th} /kg _{CO2}
Absorption capacity	349.3 g _{CO2} /kg _{MEA}
Auxiliary power demand	0.37 MJ _e / kg _{CO2}
Reboiler temperature	118 °C
Steam temperature/pressure	133.5 °C/ 3.09 bar

2.4 Calcium Looping

In this study, chemical equilibrium between the inlet CO₂ (F_{CO_2}) and the average fraction of the available CaO is assumed in order to form CaCO₃ ($X_{max} \cdot F_R$). This assumption is valid and widely found at literature [14], as the high operating temperature and fluidization phenomena of both carbonator and calciner lead to equilibrium conditions. Therefore, Gibbs free energy minimization is applied for the simulation of carbonator and calciner. As for the X_{max} parameter, it practically represents the fraction of the sorbent that is available to react and is a function of the make up limestone stream (F_0) and the solid recirculation ratio (F_R). It can be estimated by the following semi-empirical correlation [15]:

$$X_{ave} = \frac{f_m(1 - f_w)F_0}{F_0 + F_R(1 - f_m)} + f_w \quad (2)$$

where f_m, f_w are constants based on each sorbent characteristics. The carbon capture efficiency in the carbonator is calculated via the following equation:

$$E_{carb} = \frac{F_{CO_2,carb,in} - F_{CO_2,carb,out}}{F_{CO_2,carb,in}} \quad (3)$$

where $F_{CO_2,carb,in}$ is the inlet CO₂ flow in carbonator and $F_{CO_2,carb,out}$ the outlet CO₂ flow of the carbonator. Regarding the CO₂ capture efficiency of the whole Calcium Looping process, the following equation is applied:

$$E_{tot} = \frac{F_{CO_2,calc,out}}{F_{CO_2,carb,in} + F_0 + F_{CO_2,RDF}} \quad (4)$$

where $F_{CO_2,carb,in}$ is the inlet CO₂ flow in the carbonator and $F_{CO_2,RDF}$ is the CO₂ flow due to the RDF combustion in the calciner.

The hot streams released from the carbonator and calciner can be further exploited in a secondary Rankine steam cycle for steam production, whereas the heat released from the exothermic reactions of the carbonator is being used for water evaporation. Apart from sorbent regeneration, RDF combustion takes place inside the calciner. Therefore, desulfurization and dechlorination of the RDF's combustion flue gas have been taken into account via the formation of CaSO₄ and CaCl₂. Concerning the secondary steam cycle, the efficiency is calculated with the following equation:

$$eff = \frac{Net\ power\ output}{mcp\Delta T_{flue\ gases} + Q_{carb}} \quad (5)$$

Table 5: Calcium Looping key parameters

F_R/F_{CO_2}	7.5
X_{max}	0.25
T_{carb}	650 °C
T_{calc}	900 °C
E_{carb}	89.77 %
Raw supplementary fuel consumption	39.76 % of total fuel
O ₂ /CO _{2 cap}	0.436 kg/kg
ASU specific power consumption	220 kWh/tn O ₂
Air to fuel ratio	1.2
Secondary steam cycle's efficiency	32.89 %

3. Methodology

The fuel input was set as such, so that the total thermal input of the feedstock would be 50MW on an LHV basis. The net electric efficiency of the whole plant was calculated according to the following definition:

$$\eta_{net} = \frac{P_{gross\ electric} - P_{consumptions}}{Q_{thermal\ fuel}} \quad (6)$$

,where the consumptions mostly refer to the work of pumps or compressors needed and the gross electric is the power produced by the steam turbines.

In addition to the aforementioned efficiencies, the CO₂ emissions intensity was also calculated as the amount of carbon dioxide released in the atmosphere (measured in kg) divided by the amount of net total electricity produced by the plant:

$$CO_2\ intensity = \frac{m_{CO_2,emitted}}{P_{net\ total}} \left(\frac{kg_{CO_2}}{KWh_{el,net}} \right) \quad (7)$$

Furthermore, the specific fuel consumption was determined as the total amount of the input fuel to the net total electricity output of the whole plant:

$$\frac{m_{fuel}}{P_{net\ total}} \left(\frac{kg_{fuel}}{KWh_{el,net}} \right) \quad (8)$$

Lastly, for the cases of MEA scrubbing and CaL, where a reference plant already exists, SPECCA (Specific Energy Consumption for CO₂ Avoided) coefficient is introduced and expresses the additional fuel that is required to be consumed in order to avoid the emission of 1kg of CO₂:

$$SPECCA = 3600 * \frac{\frac{1}{n} - \frac{1}{n_{ref}}}{E_{ref} - E} \left(\frac{MJ}{kg_{CO_2emitted}} \right) \quad (9)$$

4. Results and Discussion

Simulations of the reference Waste-to-Energy plant have shown that a net electrical efficiency can be achieved of around 27% on a LHV basis. Figures 4, 5, 6 depict the energy flow through a Sankey diagram in each scenario for amine scrubbing, Calcium Looping and Chemical Looping Combustion respectively.

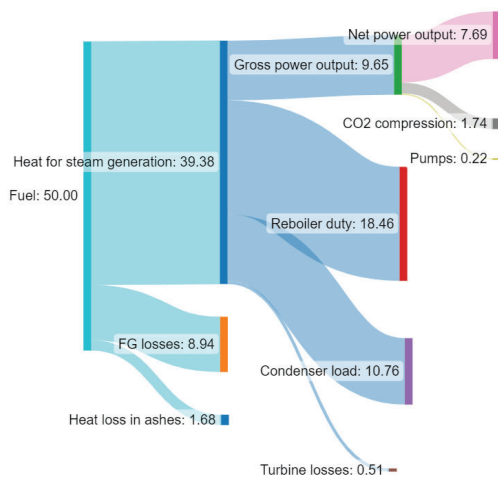


Figure 4: Sankey diagram for MEA scrubbing

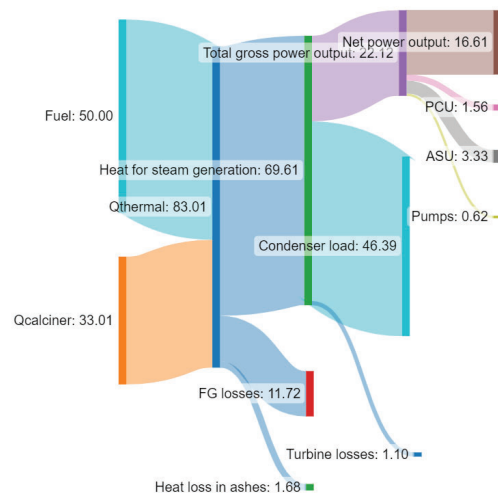


Figure 5: Sankey diagram for CaL

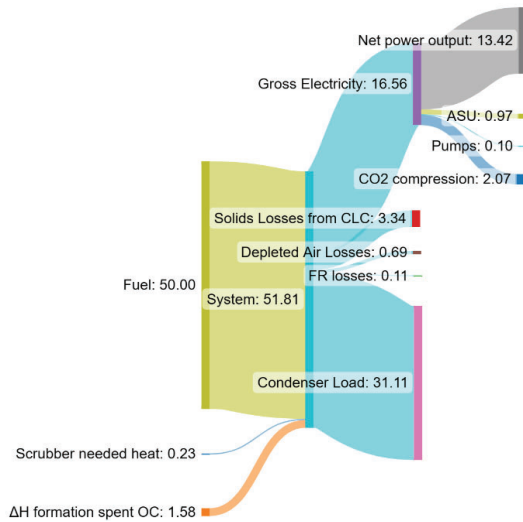


Figure 6: Sankey diagram of CLC plant

In the CLC case, around $51.5\text{MW}_{\text{th}}$ is inserted in the system, of which 50MW_{th} derive from the fuel thermal input, 220kW_{th} is needed for the Scrubber's operation and 1.6MW_{th} come from the difference of the enthalpy flows between the make-up material and the spent oxygen carrier. 31MW_{th} of that input is lost as waste heat in the condensers, 16.5MW_{e} of electricity is produced through the steam turbines and 4MW_{th} are heat losses in the atmosphere (in the form of the hot depleted air exiting the AR, the ash and spent OC exiting the FR and FR heat losses). Out of the 16.5MW_{e} of electricity produced, 2MW_{e} of electricity is consumed for the gradual compression of the CO_2 stream, 100kW_{e} is consumed for the pumps operation and considering the use of an ASU for the oxygen production needed in the POC (requires $220\text{kWh}/\text{tn}_{\text{O}_2}$) [16], which consumes about 970kW_{e} , the remaining 13.4MW_{e} penetrate the grid. The overall net electric efficiency of the plant ended up being 26.84%.

Regarding the MEA scrubbing case, 50MW_{th} of fuel is consumed. From this thermal input $39.38\text{MW}_{\text{th}}$ is used for steam generation, while the rest thermal energy is considered to be waste heat. The gross electrical output of the initial plant steam cycle is approximately 9.65MW_{e} , but taking into account the electric consumption of pumps and the CO_2 compression unit, the net electrical output of turbines is estimated at 7.7MW_{e} . Hence, the net electrical efficiency of the whole process is about 15.4%. The rest of the useful heat to the Rankine cycle is used for solvent regeneration ($18.46\text{MW}_{\text{th}}$), as condenser load ($10.76\text{MW}_{\text{th}}$) and about $0.51\text{MW}_{\text{th}}$ are turbine losses.

As for the Calcium Looping process, the total thermal input is $83.01\text{MW}_{\text{th}}$, of which 50MW_{th} is used as heat source for the steam cycle of the reference plant and $33.01\text{MW}_{\text{th}}$ in the calciner for the Ca sorbents regeneration. The useful heat for steam generation in both steam cycles (Rankine cycle of initial plant and secondary Rankine cycle of CaL process) is estimated at $69.61\text{MW}_{\text{th}}$, while heat losses is $13.4\text{MW}_{\text{th}}$. The gross electrical output of the whole process is $22.12\text{MW}_{\text{e}}$. However, the electrical consumptions in PCU and ASU lead to a net power output of $16.61\text{MW}_{\text{e}}$ and a net electrical efficiency equal to 20%. The rest of the useful heat is consumed in the condensers of the two cycles ($46.39\text{MW}_{\text{th}}$) and turbine losses ($1.10\text{MW}_{\text{th}}$).

Table 6: Comparison of main indexes

	Reference WtE plant (no CO_2 capture)	MEA scrubbing	Calcium Looping	Chemical Looping Combustion
Capture efficiency (%)	-	90	93.41	94.64
Net electric efficiency (%)	27.13	15.38	20.00	26.84

Specific fuel consumption ($\text{kg}_{\text{fuel}}/\text{MWh}_{\text{el}}$)	1382	2438	1874	1397
CO ₂ emission intensity ($\text{kg}_{\text{CO}_2\text{emitted}}/\text{MWh}_{\text{el_net}}$)	1449.63	254.76	151.45	79.31
Energy penalty (%)	-	-11.75	-7.13	-0.29
SPECCA ($\text{MJ}/\text{kg}_{\text{CO}_2\text{emitted}}$)	-	8.48	2.63	Not defined

Based on the data shown on Table 6, CLC technology can achieve a higher net electric efficiency than the other two methods, slightly lower than that of the reference plant. All three methods perform at a high capture efficiency, with no significant difference between them. Regarding the specific fuel consumption and the CO₂ emission intensity, MEA scrubbing has the lowest net efficiency of the three technologies, with CaL coming in second and CLC achieving the best performance. The energy penalty was calculated as the difference between the efficiency of each CO₂ capture technology and the efficiency of the reference plant. The SPECCA index cannot be defined for the Chemical Looping Combustion technology, given that it is not an equipment that can be implemented on an existing Waste-to-Energy plant. However, out of the two other methods, CaL achieved the lowest SPECCA index.

5. Conclusions

This study presented the comparison of three different CO₂ capture technologies implemented on Waste-to-Energy plants. In the case of MEA scrubbing, the net electric efficiency was calculated to be around 15%, mainly due to the significant amount of steam that is extracted from the power plant in order to regenerate the aqueous MEA solvent. Calcium Looping achieved a better efficiency, around 20%, with a rather low SPECCA of only 2.63 MJ/kg_{CO₂emitted} and seems to be less energy consuming than MEA scrubbing. Chemical Looping Combustion achieved the highest net electric efficiency out of the three technologies, while also being the least intense in terms of CO₂ emissions to the atmosphere. In CLC's case, a significant amount of heat was lost due to the thermal losses of the solids exiting the Fuel Reactor. Overall, all three technologies managed to reduce severely the CO₂ emission intensity of the reference plant, raising however the specific fuel consumption and decreasing, depending on the technology, the net efficiency. In conclusion, Chemical Looping Combustion is the least energy consuming and most effective technology and therefore is highly suggested as a CO₂ capture technology in case of a new power plant construction.

Acknowledgements

This project has been subsidized through ACT (EC Project no. 691712) by the German Federal Ministry of Economic Affairs and Energy (grant no. 03EE5096), the Research Council of Norway (grant no. 329886), the Greek General Secretariat for Research and Technology (grant no. T12EPA5-00023), and the Scientific and Technological Research Council of Turkey (grant no. 221N265).

Nomenclature

<i>AR</i>	Air Reactor
<i>ASU</i>	Air Separation Unit
<i>CaL</i>	Calcium Looping
<i>CLC</i>	Chemical Looping Combustion
<i>E_{carb}</i>	carbonation efficiency, -
<i>E_{tot}</i>	total carbon capture efficiency, -
<i>eff</i>	efficiency of secondary steam cycle, -
<i>F₀</i>	looping ratio, kmol/kmol

F_R	make-up ratio, kmol/kmol
FR	Fuel Reactor
m	mass flow rate, kg/s
n	molar flow rate, kmol/s
Q	heat stream, kW
P	power, kW
POC	Post-Oxidation Chamber
OC	Oxygen Carrier
T	temperature, °C
X	average conversion of solids in the carbonator/calciner, -

Greek symbols

η efficiency

Subscripts and superscripts

<i>ave</i>	maximum average
<i>calc</i>	calcination
<i>carb</i>	carbonation
<i>th</i>	thermal
<i>tot</i>	total

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