Life cycle assessment of natural gas fuelled power plants based on chemical looping combustion technology

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Abstract

Among the different Carbon Capture and Storage (CCS) technologies being developed in the last decades, Chemical Looping Combustion (CLC) stands out since it allows inherent CO₂ capture. In the CLC process, there is a solid oxygen carrier circulating between two reactors in a cycle that allows providing the oxygen needed for combustion. In one of the reactors, named as fuel reactor, the fuel is introduced and combusted while the oxygen carrier reduction takes place. In the second reactor, named air reactor, the oxygen carrier is reoxidized in air. Different materials based on copper, nickel and iron oxides have been proposed as oxygen carriers for the CLC process. This work presents an environmental evaluation of the CLC process for natural gas based on

Life Cycle Assessment (LCA). Five different oxygen carrier materials already tested in pilot plants were considered and the results compared to the conventional natural gas combustion in a gas turbine in a combined cycle without and with CO₂ capture using postcombustion capture with amines. In view of the results, lower impact of the CLC process compared to the base case is expected without and with CO₂ capture. The influence of several variables on the results was considered, such as temperature in the air reactor, lifetime of the oxygen carrier and possibility of recuperation of the depleted oxygen carrier. The nickel-based oxygen carriers were identified as the most adequate to be used in natural gas combustion. However, due to their toxicity, several analyses were also performed in order to identify improvements in the known oxygen carriers that can qualify them to replace nickel-based materials.

Keywords: CO₂ capture; natural gas; chemical looping combustion; oxygen carrier; life cycle assessment

1. Introduction

The reduction of CO₂ emissions to the atmosphere has become one of the most important challenges of the present century in view of the evidences of global warming and the subsequent climate change. An important step further was recently settled through the Paris Agreement commitment which pursues not to exceed the increase in the global average temperature in more than 2 °C by 2100 and develop strategies which could even limit the temperature increase to 1.5 °C above preindustrial levels [1]. In order to reach the total reduction in CO₂ emissions needed, several technological possibilities have been outlined: improving energy efficiency, turning to less carbon intensive fuels, deployment of renewables and of carbon capture and storage (CCS)

technologies. The later are the only technologies capable of delivering significant emission reductions from the use of fossil fuels in power generation an industrial applications in the short to medium term [2]. Among CCS technologies, chemical looping combustion (CLC) represents a promising technology due to its inherent CO₂ capture and the low cost/energy penalty associated [3]. This explains the intensive research and development that this technology has experienced since it was first considered for CO₂ capture [4] to its current level of technology readiness (TRL) equal to 6 [5] . CLC has been investigated for all types of fossil fuel, i.e. gaseous, liquids and solids. The largest experience was accumulated on gaseous fuel combustion (mainly syngas or natural gas). Therefore, CLC-based combined cycles (GCLC-CC) could be thought as an alternative to the actual gas turbine combined cycle without and with CO₂ capture, GTCC and GTCC-Amine, respectively, used for electricity production [6, 7]. A scheme of these three processes has been included in Figures 1(a) to (c). Amine scrubbing of exhaust gases is considered as the reference process for CO₂ capture in GTCC systems [8], see Figure 1(b). It is a commercially available post-combustion CO₂ capture technology based on solvent-based chemisorption. In the CO₂ capture using amine-based technology, the CO₂-rich exhaust gas is forced through an aqueous amine solution. The amine solvent reacts reversibly with the CO₂, forming water-soluble salts. The energy needed for the regeneration of the amine solvents implies a significant efficiency penalty [9].

The objective of the present paper is to evaluate the environmental impact of the CLC-based natural gas combined cycles (GCLC-CC) compared to that of GTCC and GTCC-Amine in light of life cycle assessment (LCA). LCA is a powerful tool to understand all the environmental hazards associated to the GCLC-CC process and therefore consider its future forecast as a CCS technology.

Figures

Figure 1. Scheme of (a) natural gas combustion combined cycle (GTCC) (b) natural gas combustion combined cycle with post-combustion CO_2 capture using amine absorption (GTCC-Amine) (c) natural gas combustion combined cycle with inherent CO_2 capture using chemical looping technology (GCLC-CC)

2. Background

2.1 Chemical looping combustion process

Figure 2 presents a simple scheme for the CLC process. There are two reactors, namely fuel and air reactors and a solid oxygen carrier circulating between them. The oxygen carrier is normally a metal oxide (M_xO_y) and the commonest configuration for this system is two interconnected fluidized bed reactors [10]. The operation of a CLC system have significant similarities with the operation of a circulating fluidized bed (CFB) boiler which is a well-known and commercially available technology for combustion. In CLC the combustion process is split in two (see reactions R1 and R2): in the fuel reactor, the fuel is converted to CO_2 and H_2O and the reduction of the oxygen carrier takes place; in the air reactor, the reduced oxygen carrier is oxidized again using air so that a new redox cycle can start and this way continuous combustion of the fuel can be maintained.

Figure. 2. Scheme of the CLC process.

The heat of combustion for CLC and conventional combustion is the same although the heat duty in each reactor will vary depending on the fuel and oxygen carrier used. Currently, one of the main cornerstones in CLC is oxygen carrier development. Two types of oxygen carriers have been considered for CLC, i.e. natural (minerals/ores) and synthetic materials. The selection of an oxygen carrier for CLC depends on the application intended, i.e. gas, liquid or solid fuel combustion [3]. The combustion of each type of fuel presents special features that could advise the use of one type of oxygen carrier or another. This is the case of CLC of solids fuels. In this process, the ashes from the solid fuel can accumulate during combustion and therefore should be frequently removed from the CLC system. In the drainage process, some losses of oxygen carrier are expected together with the ashes. Thus, low-cost oxygen carriers are preferred to synthetic [11]. In the case of gaseous fuels, which are the focus of the present paper, the absence of ashes in the fuel allows using both natural and synthetic materials. Nevertheless, only a few metal oxides are adequate to be reduced and oxidized under operational conditions in a CLC system and reach high selectivity to H_2O and CO_2 . Most of the oxygen carriers proposed for CLC of gaseous fuels are based on CuO , Fe_2O_3 , NiO or Mn_3O_4 [12]. Moreover, many of the oxygen carriers tested in continuous CLC units are synthetic materials, mostly prepared by co-precipitation, impregnation, freeze-granulation or spray-drying [3, 13]. Potential oxygen carrier tested in the last years at laboratory or CLC pilots with a wide range of gaseous fuels (natural gas, syngas, sour gas, acid gas ore refinery gas) have been gathered in recently published comprehensive reviews [3, 13]. Results for above 2000 oxygen carriers can be found in literature, although not more than 15 have been evaluated in continuous combustion in CLC units of different size (0.3 kW_{th} to 1 MW_{th}) [12]. In the early years of CLC development, most of the attention was focused on NiO -based materials, due

their high reactivity with methane, the main component in natural gas or refinery gas. [14, 15]. However, nowadays the focus is shifted to find alternative oxygen carriers with lower cost and more environmentally-friendly. Thus, promising materials mostly based on Fe_2O_3 and CuO have been identified [16, 17]. Even more, the use of Fe_2O_3 -based oxygen carriers has demonstrated that is possible to work in CLC with sulphur-containing gases [18] thus avoiding previous fuel cleaning processes.

In the scale-up of CLC of gaseous fuels there are several aspects to be considered. One of them and of utmost importance is the lifetime of the oxygen carrier since the losses of material due to particle attrition in the CLC system create the necessity of a make-up flow that constitutes the main additional cost [19, 20]. Moreover, the disposal of elutriated oxygen carriers is an environmental issue still to be addressed. If possible, the active metal oxide in the oxygen carrier should be recovered, especially when it is a highly expensive material [21].

The experience accumulated to date both in oxygen carrier development and pilot plant operation together with the low cost/energy penalties that the CLC technology presents compared to other CCS technologies should be boosting factors for CLC scale-up in the next future, especially when a carbon constrained world is envisaged.

2.1 Life cycle assessment

Life Cycle Assessment (LCA) allows the quantitative analysis of the material and energy efficiency of a process, the identification of environmental hazards as well as the establishment of a reference to compare. Many LCA studies can be found in literature for the GTCC processes that provide significant information regarding the environmental impacts associated. In the GTCC processes, methane leaks in natural gas extraction and transport affect life cycle emissions of greenhouse gases (GHG). Moreover, the NO_x emissions during natural gas combustion augment the terrestrial

acidification potential of the process [22, 23]. The addition to the GTCC system of a CO₂ capture process based on amine scrubbing (mostly using monoethanolamine, MEA) reduces GHG emissions [24], approximately by 50-60% [22], but increases other environmental impacts [8, 22, 25-27]. This increase is related to the loss in efficiency of the GTCC process and the corresponding additional demand for fuel, the loss of solvent and the generation of other emissions/wastes, such as ammonia emissions during MEA production [8], ethylene oxide emissions from MEA supply [8] or the emissions of MEA and formaldehyde during the capture process [26]. Singh et al [26] estimated the values corresponding to the impact of these environmental effects and established an increase of 43% in acidification, 35% in eutrophication, and 120–170% in various toxicity impacts (freshwater and marine aquatic ecotoxicity and terrestrial ecotoxicity impacts). It should be also pointed that, according to Singh et al [26], the transport and storage of the captured CO₂ contributes very little to GHG emissions.

Therefore, advantages and disadvantages of incorporating CO₂ capture to GTCC should be carefully considered. In order to reduce the environmental impacts linked to the incorporation of MEA-based absorption processes for postcombustion CO₂ capture, new solvent formulations would be needed so that the energy requirements for their regeneration would be reduced as well as the problems associated to solvent degradation [26]. Moreover, the use of the captured CO₂ in enhanced oil recovery (EOR) processes would also reduce GHG emissions [28]. Another option to reduce the environmental impacts of amine scrubbing would be to use other capture technologies. In this respect, the study of Berstad et al. [29] evaluated the calcium looping process (CaL) as an alternative to MEA for postcombustion CO₂ capture from GTCC. The CaL process is based in the reversible carbonation reaction of CaO and the subsequent calcination of the CaCO₃ formed. The exhaust gases from the GTCC process enter a first reactor

(carbonator) where CO_2 reacts with CaO forming CaCO_3 . After that, the CO_2 -lean stream at the carbonator outlet is released to the atmosphere. The produced CaCO_3 is conducted to another reactor (regenerator) where the calcination reaction takes place in order to regenerate the CaO and produce a pure CO_2 stream. Compared to the GTCC-Amine process, the GTCC-CaL process has the advantage that there are no emissions of potentially toxic chemical residues and by-products to the environment. However, a large heat supply at high temperature is required by the oxycombustion process demanded to calcine CaCO_3 . In addition, the purity of the CO_2 captured in the CaL process is lower than the corresponding CO_2 purity with the MEA-based absorption since there is excess oxygen and inert nitrogen/argon in the CO_2 concentrated stream. Considering these findings of Berstad et al. [29], the advantages of the CaL process over chemical solvents as postcombustion CO_2 capture technology applied to GTCC are not clear.

In this context, the combustion of natural gas using the CLC process (GCLC-CC) could incorporate significant advantages to natural gas combustion in a combined cycle. First, the formation of NO_x in natural gas combustion would be avoided since the fuel and the air supplied for combustion are never mixed in CLC and the temperatures reached are significantly lower compared to those existing in GTCC. Second and regarding some of the negative environmental impacts associated with CO_2 capture, the avoidance of fuel/air mixture makes that the CO_2 generated during combustion is not diluted in air nitrogen and a concentrated CO_2 stream is produced instead. Thus, CO_2 capture is intrinsic to the process. This fact makes the difference with respect to other CCS technologies, where the main cost and energy penalty are associated with the gas separation stages needed in order to isolate CO_2 from other companion gases [5].

Finally, the risk of emissions of hazardous chemical compounds associated with the solvents used in amine scrubbing is suppressed.

All of these facts mentioned above give an advantageous position for the GCLC-CC process compared to other CO₂ capture technologies to be implemented in GTCC systems. However, only few LCA studies analysing the environmental effects of CLC of natural gas can be found in literature. Petrakopoulou et al. [30] performed an economic and environmental evaluation of three oxy-fuel plants with CO₂ capture, a plant with CLC and two zero emission plants reaching 100% and 85% CO₂ capture, and compared them with a reference plant without CO₂ capture. They also performed a LCA and concluded that the most environmentally friendly concept is the plant with CLC. In this study, the authors considered that the CLC unit was a black box and assumed a fuel conversion of 98% working with a NiO-based oxygen carrier. Nevertheless, no losses of oxygen carrier were considered. In a later study, these authors conducted an analysis for three natural gas-fired power plants, i.e. a reference plant representing the business as usual scenario, one plant with CO₂ capture based on monoethanolamine (MEA) and one with CLC [31]. They highlighted the major environmental impact of natural gas consumption compared to CO₂ emissions. Recently, a systematic LCA analysis of the environmental sustainability of CLC with natural gas was presented including the evaluation of the influence on the global warming impact (GWI) of the lifetime of the oxygen carrier, global warming potential (GWP), type of oxygen carrier and the energy efficiencies of a CLC power facility [32]. They considered two different oxygen carriers, i.e. a synthetic material based on nickel and a mineral (ilmenite) and estimated from literature their lifetimes in a CLC system. They integrated the CLC unit in a power cycle and considered different thermodynamic performances as a function of the temperature at the air reactor outlet. The LCA performed offered interesting conclusions

regarding the GWI of CLC systems. However, most of the data used in this LCA study were taken from different literature sources that could add significant uncertainties to the conclusions reached. Therefore, in this paper, we present a comprehensive LCA analysis of CLC-based combined cycles (GCLC-CC) based in our own experience in oxygen carrier development and testing in CLC pilot units from 0.5 to 10 kW_{th} and compare the results with those expected with a conventional gas turbine combined cycle without and with CO₂ capture (GTCC and GTCC-Amine). In our study, several aspects are considered for the first time in LCA of the CLC process. First of all, five different oxygen carriers, both synthetic materials and minerals, developed and tested in our research group and based on CuO, NiO and Fe₂O₃ have been included in the analysis. The specific preparation method developed for each of them has been included in the LCA with the corresponding potential emissions. Second, we include estimations for the lifetime of the particles as well as solids inventories in both fuel and air reactors which were obtained in the experiments in our continuous CLC units for each of the materials considered. Third, we estimated the possibility of recovery of the active metal oxide in the elutriated oxygen carrier in order to consider its possible re-use before being sent to landfill. Additionally, we performed an experimental study on the lixiviation potential of each of the oxygen carriers with the aim of classifying them and determining which type of landfill they would require. For that purpose, we followed the current legislation set by the European Union gathered by the Council directive 1999/31/EC and the Council decision 2003/33/EC. These documents set the requirements to dispose waste in landfills by classifying the waste as hazardous, non-hazardous and inert waste. Thus, the conclusions reached in the present study could give a realistic picture of the environmental impacts associated to the development of the CLC technology for electricity production.

3. Methods

3.1 Scope of the study

The LCA presented in this paper follows the recommendations given by the European Platform on Life Cycle Assessment [33]. The present study is focused on a power plant operating with a CLC combined cycle (GCLC-CC). The scope of the study includes all the stages outlined in Figure 3. We have considered the production of 1 MWh_e as functional unit in both GCLC-CC and conventional GTCC. Processes such as oxygen carrier manufacturing, re-use or landfilling, natural gas extraction and combustion and CO₂ capture and storage are considered. It is assumed 8000 h/year of power plant operation.

Figure. 3. Life cycle boundaries (dotted line) of the GCLC-CC process

The software GaBi[®] Pro. 8.7 has been used for the LCA simulation together with the databases associated to this program. If possible, processes located in Spain have been considered. Otherwise, data from the European Union or Germany are considered.

3.2 Life cycle inventory (LCI)

Table 1 presents the main characteristics of the oxygen carriers analyzed in the present work.

Table 1. Main characteristics of the oxygen carriers considered for CLC-CC

Name	Method	MeO (%)	Lifetime (h)		$T_{FR}-T_{AR}$ (°C)		Solids inventory (FR+AR) (kg/MW)		Recovery (%)
Ni18	Impregnation	18	10000	[34]	1200-1200		100 + 100	[35]	80
Ni60	Granulation	40	40000	[36]	1200-1200	[37]	125+ 100	[20, 38]	80
Cu15	Impregnation	15	5000	[16]	800-900	[39]	150 + 350	[40, 41]	80
Fe20	Impregnation	15	1100	[17]	950-950	[42]	230+400	[41]	80
Ilmenite	-	100	700-800	[43]	950-1050	[44]	850 + 250	[45]	0

The table includes, the manufacturing method used in the production of each oxygen carrier, the lifetime estimated in the operation in continuous CLC units burning different fuels, the maximum temperature for operation in fuel and air reactors reported to date and the corresponding needs of solids inventory. More details about the preparation method of the oxygen carrier and the main results obtained in their use in a CLC continuous unit are given as supplementary information. Moreover, previous studies from the authors with elutriated Cu15 oxygen carrier [21] and other synthetic materials in Table 1, indicated that it is possible to recover some of the metal oxide contained in the oxygen carrier. Based on this experience, a recovery factor was included in Table 1 and considered in the LCA, except for the case of ilmenite. After the recovery process, the spent elutriated material should be considered as a residue of the CLC process. In order to analyse the final disposal of these elutriated particles a lixiviation study was performed with samples of the oxygen carriers in Table 1 taken during operation in continuous CLC units and following the standard CEN 12457-3, as it was previously done with Fe-based oxygen carriers used in the research group [46]. As a conclusion of these tests, the oxygen carrier residues could be classified as follows. Ni18 and Ni60 can be considered as non-hazardous wastes and disposed in a municipal landfill; Cu15

and Fe₂O₃ are considered as inert residues and ilmenite as stable non-reactive hazardous waste.

Figures 4 to 8 show the system boundaries and the fluxes and processes considered for each oxygen carrier. In the calculation of the total amount of oxygen carrier used it was considered the lifetime of each of the oxygen carriers summarized in Table 1 together with the solids inventory required, which allowed to estimate the amount of oxygen carrier needed in kg/MWh.

In the present study, two NiO-based oxygen carriers were considered with different NiO content, i.e. 18 and 60 wt%, identified as Ni18 and Ni60, respectively. Figure 4 shows the fluxes and processes considered when Ni18 is used as oxygen carrier. Thermal energy for alumina and oxygen carrier calcination has been calculated considering their specific heats and 90% of thermal insulator. Nitric acid and water used in the oxygen carrier production and recovery, ferro nickel (29%) production and thermal energy from natural gas generation can be modelled with GaBi[®] databases. Alumina manufacture has been modelled with data appeared on last LCA profile of European Aluminium [47]. Natural gas extraction, transport and combustion can also be modelled with GaBi[®] databases. Finally, the depleted oxygen carrier after the recovery process is treated in a municipal landfill.

Ni60 was synthesized from NiO and NiAl₂O₄ by freeze granulation and later calcined at 1300°C for 4 h [48]. Figure 5 presents the system boundaries and the fluxes and processes considered when Ni60 is used as oxygen carrier. The depleted oxygen carrier after the recovery process is also sent to a municipal waste landfill.

The copper oxygen carrier considered in the present work (Cu15) was also developed at ICB-CSIC. Figure 6 summarized the fluxes and processes considered. The operation

with Cu15 imposes a restriction in the temperature to be used in fuel and air reactors in order to maintain the estimated lifetime. These temperatures were estimated as 800 and 900°C for fuel and air reactors, respectively [39]. Copper production by electrolysis process can be modelled with GaBi[®] databases. The lixiviation study performed with samples after operation in our CLC continuous unit indicated that, in this case, the depleted oxygen carrier after the recovery process can be disposed in an inert waste landfill.

The oxygen carrier based on Fe₂O₃ (Fe20) was also obtained at ICB-CSIC using the hot incipient impregnation method [42], similar to those previously described for Ni18 and Cu15. In Figure 7, iron collection and refining process was modelled with GaBi[®] database. Regarding the final disposition of the depleted oxygen carrier after the recovery processes, we also performed a lixiviation study with samples drawn from the continuous CLC unit. According to the results, the depleted oxygen carrier can also be deposited in an inert landfill.

Finally, the mineral ilmenite has been considered as an oxygen carrier (see Figure 8). Ilmenite mineral production process is not in GaBi[®] database and has been modelled with Ecoinvent database [49]. In this case, no recovery process for the oxygen carrier used in the process has been proposed since it is a mineral available in large amounts and at low cost.

Together with an adequate description of the oxygen carrier manufacturing and disposal, another important parameter to be taken into account in the LCA assessment is the efficiency of the GCLC-CC process considering the oxygen carrier used. Depending on the material used as oxygen carrier, a different maximum temperature at the air reactor outlet can be reached and this can be considered as the temperature at the inlet of the gas turbine (TIT). A comprehensive literature review was carried out in order to find

345 the better estimation for each material and results are summarized in Table 2. As it is
346 shown in Table 2, the values used in the present study are an averaged estimation of
347 those found in previous studies for different oxygen carriers [6, 7, 50]. In the
348 calculation of the total amount of natural gas supplied, the efficiencies of the GCLC-CC
349 process in Table 2 were considered.

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356 **Figure 4.** GCLC-CC system with Ni18 as oxygen carrier and system boundaries (dotted line) of the LCA (Functional Unit: 500 MW_e during 25
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365 **Figure 5.** GCLC-CC system with Ni60 as oxygen carrier and system boundaries (dotted line) of the LCA (Functional Unit: 500 MW_e during 25
366 years)

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372 **Figure 6.** GCLC-CC system with Cu15 as oxygen carrier and system boundaries (dotted line) of the LCA (Functional Unit: 500 MW_e during 25
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379 **Figure. 7.** GCLC-CC system with Fe₂O₃ as oxygen carrier and system boundaries (dotted line) of the LCA (Functional Unit: 500 MW_e during 25
380 years)

Figure 8. GCLC-CC system with ilmenite as oxygen carrier and system boundaries (dotted line) of the LCA (Functional Unit: 500 MW_e during 25 years)

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390 **Table 2.** Electric efficiencies for the GTCC, GTCC-Amine and GCLC-CC processes for different oxygen carriers

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	CO ₂ capture (%)	Wolf et al. [7]			Navqi and Olland [6]		Zerobin and Pröll [50]		In this study	
		T (°C)	Pressure bar	Efficiency (%)	Pressure bar	Efficiency (%)	Pressure bar	Efficiency (%)	Pressure bar	Efficiency (%)
CuO	100	900				45.2	5 6.7	41- 44.3	10	45
Fe ₂ O ₃	100	950				46			10	46
Ilmenite	100	1000	9	49		48		48.2	10	49
		1050				49				
		1100				50.5		51.3		
NiO	100	1150				51			10	52
		1200	13	52		52		53.9		
Reference plant without CO ₂ capture	0	1400				56.6				
Reference plant with CO ₂ capture (amine scrubbing)	90	1400				48.6				

The values of electric efficiency considered for the other two process compared with the GCLC-CC are also included in Table 2, namely natural gas combustion in gas turbine without CO₂ capture (GTCC) and with CO₂ capture (GTCC-Amine). Moreover, some values were taken from bibliography for the calculation of the corresponding indicators of the GTCC-Amine process. Monoethanolamine (MEA) has been considered as amine solvent and its production process by the reaction between ethylene oxide and ammonia at 100 °C, assuming a molar ratio ammonia/ ethylene oxide of 10:1 [51]. All data have been modelled using GaBi[®] databases. The solvent solution used in the chemical absorption of CO₂ schematized in Figure 1(b) was assumed to be composed of 30% monoethanolamine (MEA) [52]. The energy required in the CO₂ desorption process was calculated as 3.8 GJ/ton CO₂ captured and the needs for new amine solution as 0.35-2 kg solution/ ton CO₂ captured [9]. Regarding the disposal of the used MEA, it is considered to be burned in an incineration plant.

Finally, the captured CO₂ has been considered to be transported and injected into an underground deposit [53]. Electric power used for the CO₂ compression is $4.99 \cdot 10^{-3}$ kWh/CO₂ captured, and the electric power used for the injection is $6.99 \cdot 10^{-3}$ kWh/ kg CO₂ captured. Finally, during CO₂ transport there are $1.701 \cdot 10^{-3}$ kg of CO₂ emission/kg CO₂ captured

3.3. Life Cycle Impact Assessment (LCIA)

GaBi 8.7[®] Pro allowed performing the LCIA study through the calculation of 14 Environmental Impact Indicators (EIIs) and following the recommendations made by the European Research Center [33]. Table S1 in Supporting Information (SI) presents the values obtained for all the EIIs used and the procedure for their estimation together with their corresponding units in three categories: level “I” (recommended and satisfactory), level “II” (recommended but in need of some improvements) or level “III”

(recommended, but to be applied with caution). The values of the indicators allow the comparison for the different process analyzed (GTCC, GTCC-Amine and GCLC-CC). In all the cases studied it is considered that the CLC process with the corresponding oxygen carrier is integrated with a combined cycle operating at its optimum pressure.

4. Results

4.1 Results obtained with the different oxygen carriers

First of all, comparisons between the conventional natural gas combined cycle without (GTCC) or with CO₂ capture (GTCC-Amine) and the GCLC-CC process is addressed. To perform the comparison, only one indicator has been selected. This indicator is the Global Warming Potential (GWP), as it is the most interesting regarding the effect of incorporating CO₂ capture in the natural gas combustion conventional process. As it can be seen in Figure 9 and Table S2, the value of GWP indicator is 40% reduced when the capture of CO₂ with amine scrubbing is incorporated to the conventional process (GTCC), which clearly indicates the advantage of incorporating CO₂ capture. Nevertheless, differences are also observed between the CO₂ capture technologies analysed, i.e. GTCC-Amine and GCLC-CC. Independently of the oxygen carrier considered in the GCLC-CC process, the value of the GWP indicator was approximately two times larger for the GTCC-Amine process. It has to be indicated here that the actual CO₂ capture efficiency of the processes based on amine solvents is 90% while in the GCLC-CC process, all the CO₂ produced in combustion is captured, as the capture is inherent to the process. This would explain the lower values of GWP obtained for GCLC-CC.

Focusing on the GCLC-CC process, the lower values of GWP are obtained for the NiO-based oxygen carriers. Those oxygen carriers are the only carriers that can operate at temperatures in the air reactor of 1200°C and therefore, the electric efficiency of the GCLC-CC process is higher (see Table 2). A high efficiency represents a lower gas natural consumption and therefore a lower impact derived from the extraction and transport of natural gas. Moreover, when the GWP values obtained for the different oxygen carriers are compared, they follow the same sequence as the electric efficiencies. The GWP values increase in the order $\text{Ni}_{18}=\text{Ni}_{60} < \text{Ilmenite} < \text{Fe}_{20} < \text{Cu}_{15}$, which is the same order for the decrease in the electric efficiency. This fact indicates an important contribution of the impact of natural gas extraction and transport to the global value of GWP. Actually, the contribution of oxygen carrier manufacturing to the value of GWP was always lower than 1%, as it is represented in Figure 9 and Table S2. Previous studies on natural gas combustion using the chemical looping technology also pointed to this conclusion [32]. The confirmation of this result is advantageous for the CLC technology, since it reveals that the environmental impact of manufacturing the oxygen carrier particles is not as important as the extraction and transport of natural gas. Therefore, the CLC technology could be thought as a good alternative to the conventional GTCC from the environmental point of view.

Despite the good results obtained with the NiO-based oxygen carriers, in the last years, toxic and environmental considerations have questioned the use of this type of oxygen carriers. Nickel has water and cancer and non-cancer human toxicity [54] and thus its use should be avoided. To date, there is rising interest on finding less costly/toxic and environmentally friendly materials. Thus, in view of the results obtained in Figure 9, alternatives should be evaluated in order to replace NiO-based oxygen carriers. Those alternatives will be investigated in the following section.

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468 **Figure. 9.** GWP indicator for conventional natural gas combined cycle (GTCC) and the
469 two processes with CO₂ capture (GTCC-Amine and GCLC-CC). Five different oxygen
470 carrier materials are considered in the GCLC-CC process. In the case of the GCLC-CC
471 process, the contribution to the GWP indicator value of natural gas extraction and
472 oxygen carrier extraction and manufacturing is indicated

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474 **4.2 Improvement of CLC combustion with different oxygen carriers**

475 **4.2.1 Temperature of operation at the air reactor**

476 In view of the results obtained in Figure 9, one of the first approaches to replace the
477 NiO-based oxygen carriers would be to increase the temperature of operation in the air
478 reactor, as this is the temperature considered at the entrance of the turbine (TIT). Thus,

1200°C is set as temperature at the air reactor outlet for the rest of the oxygen carriers. Some of them should be nevertheless discarded under this assumption. Cu₁₅ could present agglomeration problems at temperatures higher than 900 °C and the lifetime sharply decreases at high temperature [39]. Besides, it is known that ilmenite can also agglomerate at temperatures higher than 1050 °C. Therefore, only Fe₂₀ is considered to operate at 1200°C in the air reactor. As a consequence, the electric efficiency considered using Fe₂₀ will also increase to a value close to that of NiO-based materials. Results of this analysis can be also observed in Figure 9. In the figure, the GWP value obtained for Fe₂₀ under the assumptions above is comparable to those obtained with Ni₁₈ and Ni₆₀. Despite the differences in lifetime values and solids inventories between NiO-based oxygen carriers and Fe₂₀ it is again confirmed that the major contribution to the GWP value is natural gas extraction. More research work would be needed in order to assess if the Fe₂₀ oxygen carrier could withstand the operation at temperatures as high as 1200 °C.

4.2.2 Average lifetime of the oxygen carrier

The lifetime of an oxygen carrier could be one of the most important parameters to be considered in the CLC process [55]. During CLC combustion, losses of oxygen carrier are expected due to the attrition suffered by the oxygen carrier particles. The extent of these losses can greatly affect the value of the different indicators in the life cycle assessment. The NiO oxygen carriers used in the present work demonstrated a high lifetime (see Table 1), between 9-35 times that found for the Fe₂₀ oxygen carrier. If we assume that both NiO oxygen carriers and Fe₂₀ can operate at 1200 °C in the air reactor, then it would be interesting to know which is the lifetime that the Fe₂₀ material should have to reach a similar GWP value to that obtained for Ni₁₈ and Ni₆₀. Figure 10 shows the results obtained for a wide range of lifetime values.

Figure. 10. Influence of the average lifetime of the oxygen carrier on the GWP value ($T_{AR} = 1200$ °C for Ni18, Ni60 and Fe20; $T_{AR} = 900$ °C for Cu15; $T_{AR} = 1050$ °C for ilmenite)

It can be seen that, for lifetimes lower than 2000 h, the GWP value is higher for Fe20 compared to Ni18 and Ni60. In this interval the contribution of the oxygen carrier to the global GWP value is higher. But for lifetimes higher than 2000 h, the GWP values for the three oxygen carriers are similar and barely change with the lifetime. As a conclusion, it can be said that if the lifetime of the Fe20 material could be improved to values around 2000 h, the Fe20 oxygen carrier could be a promising material to replace Ni18 and Ni60. The evolution of the GWP value with the average lifetime was also calculated for the rest of oxygen carriers analyzed in this work, i.e. Cu15 and ilmenite, and included in Figure 10. In all the cases, the values of GWP obtained for Cu15 and ilmenite were larger than those obtained with Ni18, Ni60 and Fe20 at $T_{AR} = 1200$ °C.

Nevertheless, the trend observed was similar: a decrease of the GWP value up to lifetimes of 1000-2000 h and then then the GWP is almost not affected by the lifetime considered.

4.2.3 Recovery of the oxygen carrier

In order to further reduce the impact of the oxygen carrier on the GWP value, it would be possible to recover part of the metal oxide used in the manufacturing process. As mentioned before, this recovery process was evaluated by the authors experimentally for some of the oxygen carriers used [21]. All the calculations presented until now assume an 80% recovery of the oxygen carrier except for ilmenite (see Table S3 in supporting information). The analysis of the environmental impact of the recovery degree was extended to other indicators used in the life cycle assessment and not only to the value of GWP since previous studies in literature indicated that the incorporation of CO₂ capture in the natural gas GTCC could negatively affect other environmental parameters. With this consideration in mind, Figure 11 presents an analysis applied to 14 indicators on the influence of the recovery percentage in the value of the indicator. In Figure 11, the values obtained for the GTCC process without CO₂ capture are considered as reference and results for both the GTCC-Amine and the GCLC-CC process are shown as an increment/reduction of this reference value. All the symbols in the figure represent results assuming 80% recovery percentage (except ilmenite). Results corresponding to recovery between 65-90% are indicated by the error bars. This percentage interval was selected since it was wide enough to evaluate the environmental impacts of the recovery degree.

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542 **Figure.11.** Variation percentage for the 14 indicators used in the life cycle assessment of the GCLC-CC process for different oxygen carriers

543 referenced to the base case (GTCC without ant with CO₂ capture). Results for recovery percentages of the oxygen carrier of 65 % and 90 %

544 indicated by the error bars

One of the first findings in Figure 11 is that the GTCC-Amine process represents an increment over the reference value in all the indicators considered except GWP. However in the case of the GCLC-CC process and independently of the oxygen carrier considered, the values of the indicators Global Warming Potential (GWP), Photochemical Ozone Formation (POF), Acidification Potencial (AC), Eutrophication Terrestrial (EUT) and Eutrophication Marine (EUM) are lower than the reference, i.e. decrease with respect to those calculated for the conventional process (GTCC). This decrease is mainly associated to the avoidance of NO_x/amine emissions in GLCC-CC. Moreover, this is a quite remarkable advantage of the GCLC-CC process when compared to GTCC-Amine, since it indicated that is possible to introduce CO₂ capture in the GTCC process without increasing other environmental impacts. Nevertheless, in the GCLC-CC process there is a slight increase respect to reference case for the indicators: Particulate Matter (PMP), Ionization Radiation (IR), Eutrophication Freshwater (EUF), Human Toxicity Cancer (HTC), Human Toxicity Non-Cancer (HTNC), Ecotoxicity Freshwater (ECFW) and Resources Water Depletion (RDW). The increase is even larger for the indicator Resources Mineral Depletion (RDM) in the case of Cu15 and ilmenite. These trends are also common to all oxygen carriers, although the values of the indicators were lower for Ni18 and Ni60, mainly due to the long lifetime of these materials.

Another fact to highlight in Figure 11 is that the recovery percentage does not affect significantly the value of the indicators for any of the oxygen carriers considered, except for the case of Resources Mineral Depletion (RDM) for Cu15. The RDM indicator is also especially high for ilmenite, since no recovery has been assumed for this oxygen carrier. If ilmenite could be used for its original utility (TiO₂ production), the value of this indicator would be notably decreased. Finally, it should be noted that human and

water toxicity indicators values are not very high for nickel oxygen carriers although it has been said previously that nickel has high toxic potential. This is because in this study it has been considered that nickel dust is not released during Ni18 and Ni60 oxygen carriers manufacture and use due to the difficulty of quantifying these amounts. Finally, another interesting conclusion that can be drawn from Figure 11 is related to the comparison of the CO₂ capture processes (i.e. GTCC-Amine and GCLC-CC). In almost all of the indicators considered and for almost all oxygen carriers the values obtained are lower for the GCLC-CC process. Therefore, the implementation of the GCLC-CC process in natural gas combustion is a more advantageous option than the commonly used GTCC-Amine. This is an additional advantage that could be also considered together with the cost of both CO₂ capture technologies. Since the CO₂ capture process is the most costly in the CCS chain, a comparison between CCS technologies could be focused on the levelized cost of electricity (LCOE). CLC of natural gas seems to be clearly advantageous when compared to the GTCC-Amine process. A recent study [56] estimated the LCOE for a natural gas CLC integrated with a supercritical CO₂ cycle as 40.5 €/MWh while LCOE for GTCC-Amine was calculated as 48 €/MWh [57].

4.3 Uncertainties in the LCA assessment

In order to evaluate if LCA results are significantly affected by uncertainties on inventory data, a Monte Carlo analysis involving 100 simulations has been performed [58], estimating uncertainty variations of ± 10 % in the main inputs of the processes which are the oxygen carriers and natural gas consumption. Results for the coefficients of variation (CV) for the EIs are shown in Table S4 and they give an indication of the variability associated to the risk parameters [59]. In Table S4, only the increase in CV higher than 0.3% is considered. Regarding Ni oxygen carriers it can be seen that natural

gas inputs are the only data that introduce high uncertainty. Natural gas amounts for each CL-CLC process have been calculated using electric efficiencies for the CL-CC (Table 2) and can be considered reliable. For Fe₂O₃ oxygen carrier main uncertainty is again introduced by natural gas input. For Cu₂O oxygen carrier main uncertainty is also introduced by natural gas input (Table 2). For ODP indicator some uncertainty is introduced by alumina and Cu(NO₃)₂ inputs. Amounts of each fluxes has been extracted from OC data used in the CLC experimental plant [40, 41]. Cu(NO₃)₂ manufacture has been simulated using nitric acid and a copper manufacture processes that appear in GaBi[®] database. One source of uncertainty appears for Resource Depletion Mineral (RDM) indicator when a variation is introduced in the amount of Cu(NO₃)₂ recovered after OC use in CL-CLC system. However, this uncertainty does not vary the main conclusions of this study and it can be considered reliable. Finally, for ilmenite OC the main uncertainty is also introduced by natural gas input (Table 2). In summary the main sources of uncertainty of these studies comes from the electric efficiency and OC inputs data used for each simulation. However, the LCA study can be considered robust and the main conclusions reliable because these data have been extracted from experimental CLC plants.

4.4 LCA in a CO₂-emission constrained world

According to the data presented above, the main environmental impact in the combustion of natural gas using the three different options outlined in the present work comes from natural gas extraction and transportation. From the data supplied in the software GaBi[®], it can be estimated that about 0.0156 kg CO₂ eq. per MJ electricity produced are emitted in these processes and not related to the combustion process itself. Moreover, some methane leakages are also acknowledged, contributing with 0.0086 kg

CO₂ eq/ MJ electricity. Nevertheless, the software does not specify which the origin of the CO₂ emissions outside combustion is. It could be thought that most of them would come from the natural gas cleaning process before being commercialized. In any case, in a CO₂ constrained world as that envisaged in the Paris Agreement [1], this CO₂ emitted outside the combustion process could be also avoided. Actually, there is already some experience in CO₂ capturing and storing in the natural gas extraction processes [2]. Therefore, the environmental impact of the processes studied in this paper could be significantly modified in the next future. Based on the figures previously mentioned, an estimation of the possible GWP values attained is also shown in Figure. 9 (green bars). The GWP values for the CLC process can be reduced by 70%, approximately.

5. Conclusions

The GCLC-CC process has been analysed from a life cycle assessment perspective in order to evaluate its environmental impact compared to different processes: natural gas combustion in a combined cycle without CO₂ capture (GTCC) and with CO₂ capture with amine scrubbing (GTCC-Amine). An effort to adapt the results obtained to the current state-of-the art was made by using data obtained for oxygen carriers already tested in the CLC continuous unit now in operation. Two of the oxygen carriers were based on NiO (Ni18 and Ni60), one on iron oxide (Fe20), one on CuO (Cu15) and the last was a mineral (ilmenite).

The most interesting conclusions obtained is that the CLC process does not add an important negative environmental impact to those already observed in natural gas combustion by other technologies. The impact associated to the use of oxygen carriers of different nature was always lower than that associated to natural gas extraction and transportation. Moreover, the values of the indicators GWP, POF, AC, EUT and EUM

were lower than those obtained for GTCC. When GCLC-CC and GTCC-Amine were compared, the environmental indicators were always lower for GCLC-CC, which highlights the potential of GCLC-CC as an alternative technology with CO₂ capture.

Regarding the type of oxygen carrier to be used, the NiO-based oxygen carriers were those with lower values of the GWP indicator. The electric efficiency of the GCLC-CC process reached using this type of oxygen carriers was the highest since they can operate at the highest temperature (1200 °C). However, their use is no more recommended due to their toxicity. Several alternatives to their use have been presented in this paper. Fe₂O₃ seems to be the best candidate for their replacement although it requires previous improvement of both its chemical and mechanical properties. It should be possible to operate at higher temperatures than those reported until now and a lifetime higher than 2000 h should be reached, which is almost the double than that reported to date.

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