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Life cycle global warming impact of CO_2 capture by in-situ gasification chemical looping combustion using ilmenite oxygen carriers

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ABSTRACT

In-situ gasification chemical looping combustion (iG-CLC), which has been tested on pilot plant level, is regarded as an advanced carbon capture and storage (CCS) technology for reducing CO_2 emissions. A life cycle global warming impact (GWI) analysis is performed to consider the lifetime emissions of the low-carbon iG-CLC technology. Herein, the capacity is considered to be 610 MW_e using natural ilmenite as oxygen carrier and steam as gasification agent. At the condition of operational pressure of 15 atm and air reactor temperature of 1050 °C, the net power efficiency of 37.7% for achieving 93.5% inherent CO_2 capture is obtained in simulations with thermodynamically optimum condition. The life cycle GWI is calculated equal to be 160.3 kg CO_2-equivalent/MW h. The effects of several essential parameters, including steam to carbon ratio (S/C), oxygen carrier to fuel ratio (ϕ), different oxygen carriers and lifetime of oxygen carriers, on the lifecycle GWI have been analyzed and discussed to meet the potential possibility for further reducing greenhouse gas (GHG) emissions. To obtain sufficient carbon capture efficiency, the S/C ratio and ϕ are suggested to be 1.3 and 1.2 in this study, respectively. The life cycle GWI is heavily dependent on lifetime of ferrum (Fe) when it is less than 2000 h, beyond that range, the GWI is decreasing, but very slowly.

1. Introduction

Increasing concerns associated with irreversible climate change have led policy-makers to design climate policies to reduce the anthropogenic Greenhouse gas (GHG) emissions. For example, the Chinese government committed to reduce its CO_2 emissions per unit of GDP by 40%–45% of 2005 levels by 2020 (Yi et al., 2011). Significant R&D efforts are underway worldwide to mitigate GHG emissions, including (1) switching to low-carbon fossil fuels, such as biofuels, natural gas or hydrogen; (2) decarbonisation of fuel or flue gas and then carbon sequestration, i.e. carbon capture and storage (CCS); (3) accelerating the use of renewable energy (such as bioenergy, direct solar and wind energy). The share of low-carbon electricity supply (including renewable energy, nuclear and CCS) is expected to be increased from currently 30% (2014) to more than 80% by 2050 (IPCC, 2014).

Yet still in a very early stage of technical demonstration, the cost of renewable energy is not currently competitive; the current CCS technology incorporating into a power plant has also not reached large-scale commercial maturity owing to the efficiency losses (e.g. CO_2 capture by amine scrubbing reduces the power output by 20–30% for a typical coal-fired power plant) as well as cost enhancement on CCS. Though numerous power plants can be incorporated with CCS activity, very limited demonstration plants have gained any measure of acceptance from an industrial viewpoint as more than 20 large-scale CCS projects have been cancelled in 2010–2016 owing to the sharply fluctuated policy and financial support (IEA, 2016). Up-to-date information related to CCS can be found elsewhere (Boot-Handford et al., 2014; Macdowell et al., 2010).

Chemical looping combustion (CLC) can obtain inherent separation of CO_2 in redox reactions without extra energy penalty. CLC is regarded as one of the most promising options for a long-term implementation for CO_2 capture (Boothandford et al., 2014). The name of CLC was first proposed by Ishida and Jin in 1987 (Ishida et al., 1987), followed by significant research effort in the last three decades from "proof of concept" to small test-rigs. A couple of comprehensive reviews covering the development statue of CLC

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including the oxygen carrier development and the operation experience have been reported by Adanez et al. (Adánez et al., 2018; Adanez et al., 2012). Recently the up-to-date progress of CLC and hydrothermal processes for carbon conversion for future cleaner energy production was summarized by Demirel et al. (2015). Generally, CLC allows for combustion of gaseous fuel, liquid fuel and solid fuel (Hoteit et al., 2011; Lyngfelt, 2014; Mattisson et al., 2001). Historically, development of CLC at early stage focused on gaseous fuel feedstocks (e.g. natural gas), and today considerable attentions have been paid to solid-fuel CLC, which is regarded as a key dimension for CLC development. Two options are taken into consideration for direct solid fuel CLC operation, i.e. in-situ gasification chemical looping combustion (iG-CLC) (Cao and Pan, 2006) and chemical looping with oxygen uncoupling (CLOU) (Mattisson et al., 2009). The difference of two options relies on distinct reaction mechanisms, since in terms of iG-CLC process the gasified syngas (from char gasification) as well as volatiles is oxidized by oxygen carrier to raise a stream of CO_2 and H_2O, different from that of CLOU concept-direct oxidation of coal char by O_2 released from specific oxygen carrier (Adanez et al., 2012). In this study, the former option is focused on because of its mature. Up to now, two bigger iG-CLC pilots have been erected, i.e. a 1 MW_{th} unit at University of Darmstadt and a 3 MW_{th} unit at Alstom Power Plant. The oxygen demand required for fully oxidizing the unreacted gas in 1 MW_{th} unit was varied between 12 and 17 mol O_2/s and oxygen injection for full oxidization was proven to be successful (Ströhle et al., 2014). Autothermal operation was achieved for Alstom plant. These preliminary test results seemed promising (Lyngfelt, 2014).

The reactions happened in the FR include: first the coal is gasified into syngas via gasification agent, e.g. steam (mainly $C + H_2O{=}CO + H_2$); secondly the syngas reacts with the oxidized oxygen carrier (OC), i.e. MeO, to obtain a mixture of CO_2 and H_2O ($CO + MeO = CO_2{+}Me$ and $H_2{+}MeO = H_2O + Me$). To maintain cyclic redox, the reduced OC, i.e. Me, is re-oxidized in the air reactor (AR) with air ($Me{+}1/2O_2 = MeO$). The outlet gas from the FR mainly consists of CO_2 and H_2O, and by condensing water vapor, almost pure CO_2 can be gained. The captured CO_2 can be transported to the storage site or used for enhanced oil recovery (EOR). With repeated cycling, OC strength is reduced because of its structure change, thus leading to decreased attrition rate and lower lifetime (Adánez et al., 2009; García-Labiano et al., 2009; Ksepko et al., 2010; Shen et al., 2010). To compensate such loss, fresh OC is continuously directed to the FR. And the spent OC is disposal of in landfill.

Life cycle assessment (LCA) is aimed at evaluating the environmental impact throughout a product's life cycle by taking into consideration of all emissions from construction and plant operation to final decommissioning. Table 1 presents the representative LCA works on first-generation CCS (pre-combustion, post-combustion and oxy-fuel) and advanced carbon capture (e.g. calcium looping) power generation technologies. Odeh et al. have examined the life-cycle GHG emissions of coal-fired pre-combustion and post-combustion CCS through LCA (Odeh and Cockerill, 2008b). Additionally, a similar LCA assessment has been conducted for oxy-fuel combustion (Gładysz and Ziębik, 2015). For comparison purpose, LCA assessments of the first-generation CCS technologies have been performed on the same basis (Cuéllar-Franca and Azapagic, 2015). Furthermore, to demonstrate the superiority of second-generation calcium looping technologies, Hurst et al. and Kursun et al. have made efforts to justify the carbon reduction potential of coal-fired calcium looping process using LCA (Hurst et al., 2012; Kursun et al., 2014). Despite all previous efforts, a full chain GHG emission assessment related to an iG-CLC power plant with ilmenite as oxygen carrier is still absent in the literature to the best of our knowledge. The motivation to perform such study is the necessity to examine CO_2 emissions of solid fuel-based CLC process from its lifetime view against other capture technologies, in which iG-CLC technology is relatively mature (with successful demonstration plants) and broadly studied experience in comparison with CLOU technology, and the use of ilmenite as suitable OC relies on its cheap price, reasonable reactivity and wide use for iG-CLC technology. Though a preliminary study associated with the LCA of natural gas-based three-stage chemical looping process for hydrogen production was conducted by Petrescu et al. (2014), our study focuses on coal (instead of natural gas)-feed iG-CLC process for power generation (rather than hydrogen production). It is clear that iG-CLC process is beneficial for reducing carbon footprint during operation, but doubters concern if it qualifies to reduce GHG emissions from the life cycle view because of the additional emissions resulting from OC manufacturing for make-up due to the reactivity losses during redox reactions as well as the different configurations and operating conditions against other capture technologies. Concerns are generally focused on the effects of OC types, conversion rate of OC with coal and the lifetime of OC on life cycle GHG emissions. In this study, we therefore conduct a LCA to examine the GHG missions in the lifetime of iG-CLC plant for the first time to address these concerns and to provide recommendations for future research needs.

2. Methodology

2.1. Life cycle assessment method

To address the complexity of LCA, the International Organization for Standardization (ISO) built a methodological framework (ISO life cycle assessment-requirements and guidelines) to carry out a complete LCA, which contains four main phases, namely the goal and scope definition, the inventory analysis, the impact assessment and the interpretation (Organization, 2006).

2.2. Goal

The aim of this study is to examine the life cycle GHG emissions of the iG-CLC power plant. The intended audiences are inclined to be researchers working on CLC as well as carbon capture, furthermore the reported results can be used as preliminary data for policy-makers to compare among the multi-carbon-capture options and design effective carbon-capture policy.

2.3. Scope

2.3.1. iG-CLC power plant

The in-detail description of the iG-CLC power plant is reported in our previous study as well as its detailed simulation methodology (Fan et al., 2017b). IG-CLC process as presented in Fig. 1 should integrate downstream gas turbine combined cycle for power generation. The function of iG-CLC unit is to achieve inherent separation of CO_2 with the aid of intermediate oxygen carriers as well as to generate two combustion flue gas streams, i.e. AR-Flue gas mainly composed by N_2 and few O_2, and FR-Flue gas mainly composed by CO_2 and H_2O. Notably, a carbon stripper is employed to facilitate char conversion efficiency by recycling unconverted char back to the FR. The pressurized high-temperature flue gases are directed into combined cycle for power generation. Notably, a fraction of middle-pressure steam (at approximately 15 bar) is extracted from the heat recovery steam generation unit to the FR as gasification agent. The cooled FR-Flue gas is mainly composed by CO_2 and H_2O, and water vapor condensation benefits separation of H_2O from CO_2. Through compression, the captured CO_2 is ready for further transport.

Table 1
Representative work on LCA of power plant with CCS.

	Source	Main aim
First-generation	Odeh and Cockerill, Pehnt and Henkel (Odeh and Cockerill, 2008b; Pehnt and Henkel, 2009)	Evaluating life cycle GHG emissions from coal-fired power plant with CCS (pre-combustion and post-combustion capture)
	Gładyszand and Ziębik, Koiwanit et al. (Gładysz and Ziębik, 2015; Koiwanit et al., 2014)	Presenting LCA for an integrated oxygen-fuel combustion power plant with CCS
	Cuéllar-Franca et al. and Singhet al. (Cuéllar-Franca and Azapagic, 2015; Singh et al., 2011)	Comparing life cycle GHG emissions of all first-generation CCS technologies
	Lidia and Nie et al. (Lombardi, 2003; Nie et al., 2011)	Reducing life-cycle CO_2 emissions strategies in power plant
	Singh et al. and Piewkhaow (Piewkhaow, 2016; Singh et al., 2011)	Conducting life-cycle assessment of electricity generation from coal based CCS technologies including pipeline CO_2 transport and geological storage
Advanced technologies	Hurst et al. (Hurst et al., 2012)	Examining life cycle GHG emissions of a 500 MW_e pulverized coal-fired power plant with calcium looping
	Kursun et al. (Kursun et al., 2014)	Demonstrating the environmental benefits of a coal-derived syngas calcium looping plant to capture halides, sulfur and CO_2 using LCA
	Petrescu et al. (Petrescu et al., 2014)	Examining life cycle GHG emissions of a three-stage iron-based chemical looping combustion process for hydrogen production

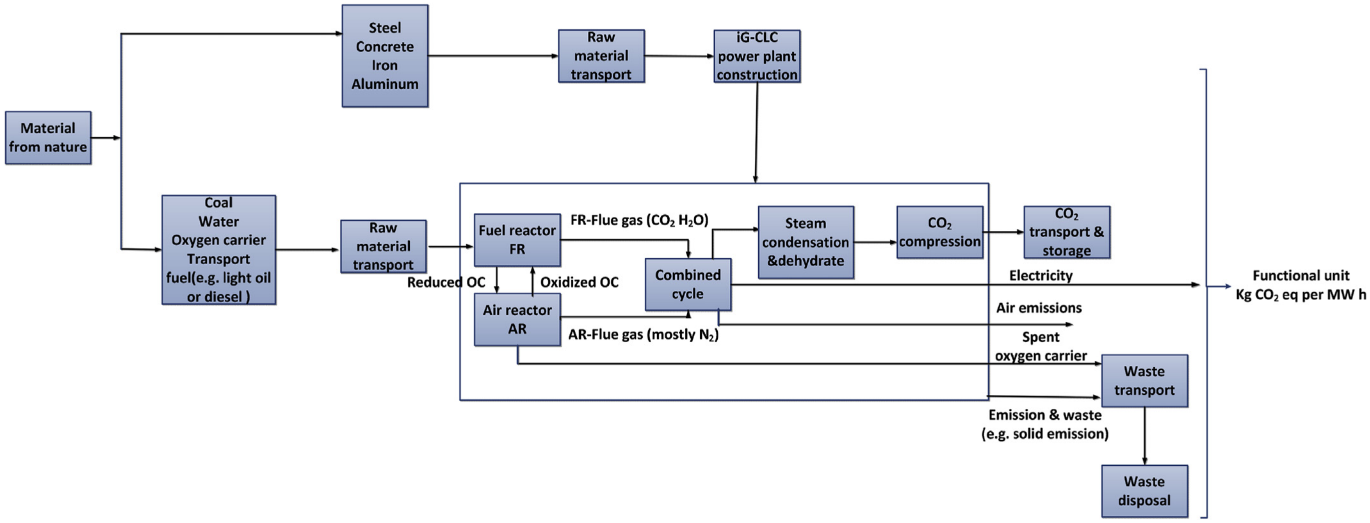

Fig. 1. Boundaries for the iG-CLC power plant.

The whole process is simulated using commercial Aspen Plus, which has been demonstrated as a powerful tool for process development and modelling. The PR-BM method uses the Peng-Robinson cubic equation of state with the Boston-Mathias alpha function for prediction of the thermodynamic data and phase behavior of a material stream, and it is recommend for gas processing application by the Aspen Property method selection guider (Fan et al., 2016). Therefore, PR-BM is chosen as global property method in this study. Besides, several assumptions are considered including: (1) negligible pressure and heat losses in all simulations; (2) steady-state operation; (3) consideration of inert ash component (no reactions between ash and oxygen carriers); (4) assuming phase and reaction equilibrium, i.e. thermodynamically best performance. However it should be noted in practice the char gasification step in iG-CLC process is more inclined to be kinetics-controlling, restricting by reaction residence time and reaction rate in the FR. In case of a 100 kW iG-CLC unit coupled with a carbon stripper, the carbon capture efficiency was more than 90% in 20 h of experimental results (similar to thermodynamic results), however the selectivity of coal oxidization to CO_2 and H_2O was much lower than that obtained in the thermodynamic results (nearly full selectivity), and the downstream pure oxygen polish step was necessary with an oxygen demand (i.e. the fraction of oxygen lacking to achieve a complete combustion of the gases leaving the fuel reactor) of 16% (Markström et al., 2013). It is possible to be close to thermodynamic equilibrium conditions in future improvement by means of improving separation efficiency of the carbon stripper, prolonging reactor length and usage of catalysts for accelerating reaction rate. The main assumptions in this analysis are presented below:

(1) The pressurized operation of CLC process is considered in this study in spite of current inability to realize a real pressurized CLC demonstration plant with fluidized bed, with two major reasons: (1) it has been recognized that for the energy generation sector, pressurized iG-CLC has been identified as the most interesting option to increase the system efficiency of power system (Adánez et al., 2018); (2) this thermodynamic performance is intended to present the best performances of iG-CLC power system by considering the pressurized operation, in line with the future demands of solid fuel iG-CLC power plant. Therefore the best potential of greenhouse gas emission reductions for such plant can be estimated and determined. The operational pressure of CLC

reactors is chosen to be 15 bar in this study to meet the optimum power efficiency in according with the AR reactor temperature (Fan et al., 2017b).

(2) Though the coal-N and coal-S would lead to SO_x and NO_x emissions in air reactor and fuel reactor, the removal of these impurities is not considered due to the focus of greenhouse gas emissions in this LCA. Also the downstream oxygen polishing step is not considered because of the thermodynamic assumption of this simulation.

Table 2 presents the simulation specification of the iG-CLC power plant.

The plant capacity is assumed to be 610 MW (net output) with 100% capacity, with corresponds to constant coal consumption of 8640 t/d. And the power efficiency is calculated to be 37.3%. The plant lifetime is considered to be 30 years with preliminary 3 years for construction (Spath et al., 1999). Though the lifetime of power plant has certain influence on GWI (global warming impact), these influences are very slight especially for when the lifetime of plant is high, which mainly stem from influences of plant construction; since lower lifetime of plant leads to higher GWI (less than 10 years) but it is not realistic. And the compositions of coal used is presented in Table 3.

In regards to a CLC process, oxygen carrier is acted as the most important role working as intermediates for completing redox reactions. For all the possible options, ilmenite-based oxygen carrier has received continuous attentions worldwide due to its enough reactivity as well as its cheap price. For example, a 100 kW_{th} ilmenite-based iG-CLC plant was successfully run in CHALMERS (Markström et al., 2013). Following the plant capacity was scaled up to 1 MW_{th} (Ströhle et al., 2015). The composition of ilmenite employed in this study is composed by 22.0 wt % Fe_2O_3, 38.5 wt % Fe_2TiO_3, 39.5 wt % TiO_2 (Abad et al., 2011; Cuadrat et al., 2012a). The attrition rate of ilmenite is assumed to be 0.076%, with corresponds to a lifetime of 1315 h (Cuadrat et al., 2012b).

2.3.2. Functional unit

The functional unit aims to provide a reference to which the inputs and outputs are normalized (Organization, 2006). For comparison purpose and easy understanding, in current study the functional unit is set as 1 MW h of electricity output from the power plant, which is in line with previous LCA studies concerning on power plants.

2.3.3. System boundary

The system boundaries for the current study are shown in Fig. 1. The system boundaries should be drawn as broadly as possible (Spath et al., 1999); all of the major processes necessary to generate electricity by means of iG-CLC including the upstream emissions, downstream emissions and emissions during plant operation have been totally involved. In terms of upstream process, construction material manufacturing (i.e. steel, concrete, iron and aluminum) related emissions should be counted as well as emissions associated with transportation of construction materials to the iG-CLC power plant site. Besides, underground coal mining (including mining equipment manufacturing, methane emissions and on-site electricity consumption and fuel consumption), and coal transport integrated with oxygen carrier transport have been taken into consideration. For downstream processes, wastes transport and disposal of them in a near-by landfill are included. In an iG-CLC power plant, the wastes refer to spent oxygen carriers and solid ashes. Additionally, emissions related to pipeline manufacturing for CO_2 transport to the storage site conjunction with re-compression of CO_2 along the pipeline have also been involved.

Notably, sulfur-removal process is not considered in present iG-CLC power plant. In conventional coal power plant, the desulfurizing unit is necessary to remove sulfur compounds contained in the combustion flue gas. By contrast, the coal-S compounds are generally contained in the FR-flue gas (mostly H_2S and SO_2) in the iG-CLC power plant. In light of our recent research, the concentrations of H_2S and SO_2 in the FR-flue gas are approximately 20 times more than recommendations (recommendation: $H_2S + SO_2 < 300$ ppm (Cormos and Cormos, 2014; De Visser et al., 2008)) (Fan et al., 2017b). Therefore, to meet the need for safely transporting and storing CO_2 for future iG-CLC power plant, removal of sulfur compounds is of necessity. However suitable wet-scrubbing solvents for capturing few amount of sulfur compounds

Table 3
Proximate and ultimate analysis (wet basis) and lower heating value of coal (Cuadrat et al., 2012c).

	Lignite
Proximate analysis	
Moisture (wt.%)	12.5
Volatile matter	28.7
Fixed carbon	33.6
Ash	25.2
Ultimate analysis	
C	45.4
H	2.5
N	0.5
S	5.2
O	8.6
LHV (kJ/kg)	16250

Table 2
Simulation specification of the iG-CLC power plant.

Unit	Simulation	Specification
FR	Three reactors, i.e. Ryield, RGibbs, and RGibbs embedded in Aspen plus, in series to simulate in-situ gasification chemical looping reduction process with function of simulating coal pyrolysis, coal gasification by steam and syngas oxidation with oxygen carriers, respectively.	Feedstock: lignite coal; FR operating temperature: 866 °C; FR operating pressure: 15 atm (Fan et al., 2017b); Steam to carbon ratio (S/C): 1.3; Oxygen carrier: ilmenite.
AR	One reactor, i.e. RGibbs built-in Aspen plus, is used to simulate re-oxidation of reduced oxygen carriers.	Air composition: 21 vol% O_2 and 79 vol% N_2; Air inlet temperature: 25 °C; AR operating temperature: 1050 °C (Ridha et al., 2016); AR operating pressure: 15 atm (Fan et al., 2017a).
Carbon stripper	An embedded Sep model is employed to perform solid separations.	Char separation efficiency: 95% (Ströhle et al., 2014)
GT/ST	A Compr model is used to simulate gas/steam expansion for power generation	Isentropic efficiency: 0.88 (Fan and Zhu, 2015) Mechanical efficiency: 0.98 (Fan and Zhu, 2015; Zhu and Fan, 2015)
HRSG	MheatX model is employed for simulation of multi-stage heating and cooling heat exchanging, which is formulated restricted by the minimum temperature difference (pinch temperature)	Steam pressure level: 80 atm/15 atm/3 atm (Zhu and Fan, 2015) Steam temperature: 450 °C/450 °C/450 °C

in concentrated CO_2 stream (>95 vol %) are still under research and deserve future investigations to improve sulfur-capture selectivity. Therefore sulfur removal process is not considered in this study because of the uncertainties of future solvents and its trace influence on GWI.

2.4. Life cycle inventory (LCI) analysis

2.4.1. Data collection

2.4.1.1. Construction material requirement. Several types of building materials are required to build a power plant, largely relying on concrete, steel, aluminum and iron. Due to lack of data, fundamental industrial material requirements are taken from an average value of these required in a coal fired plant and a natural gas fired plant, as tabulated in Table 4, and the CO_2 emissions during these materials manufacturing are presented in Table 5. The GHG emissions during plant construction stand for very limited percentages in terms of cradle-to-grave emissions in a conventional power plant (Spath and Mann, 2000; Spath et al., 1999). And based on the calculated results, this percentage is demonstrated to be very low in an iG-CLC power plant (less than 1% as shown in Fig. 2). Therefore for analyses, this assumption is valid for an iG-CLC power plant. The emissions associated with plant decommissioning are not considered because of its trace share to life cycle GHG emissions (the share of plant construction is even less than 1% including construction material manufacturing and transport). Furthermore recycling the construction materials after decommissioning is not considered.

2.4.1.2. Coal mining. Depending on geological conditions, coal can be obtained from either surface mining or underground mining (Spath et al., 1999). In China, surface coal resources only represent for less than 10% of total coal resources; the remains belong to underground coal resources. In this study, the coal is assumed to be mined from underground. Equipment associated with longwall mining include longwall unit, continuous miner, shuttle car, etc. Table 6 summarizes the steel requirements for coal-mining equipment manufacturing as well as energy requirement during mining. Besides, during coal mining, methane leakage has a significant influence on life cycle GHG emissions because methane belongs to category of GHG. The amount of CH_4 emissions are transformed into CO_2 equivalents using a global warming potential factor of 21 over a 100-year horizon (Change, 1996). The methane emission is considered to be 5.01 kg/ton coal from the institute of coal chemistry in China (Xia et al., 2010). The emissions during coal cleaning are negligible and have been rejected from consideration in this study (Spath et al., 1999).

2.4.1.3. Raw material transportation. We assume construction materials, coal and oxygen carriers are obtained from the same place. Transport distance is assumed to be 100 km near plant site within the economic-interest distance (Odeh and Cockerill, 2008a). The trucks using diesel fuel are considered for transport, with a dead weight capacity of 10 ton per truck and diesel fuel consumption of 17 L/100 km.

2.4.1.4. Landfilling of coal ash and spent oxygen carrier. The unconverted char, coal ash and spent oxygen carriers should be disposed by landfill. All waste materials are assumed to be transported via diesel-fuelled truck and the distance between plant site and landfill destination is assumed to be 10 km, following the national standard of China to direct the distance for waste landfill (GB 16889–2008). The GHG emissions from materials manufacturing of landfilling equipment are neglected (Spath et al., 1999) owing to the relatively small amount of solid wastes.

2.4.1.5. CO_2 transport and storage. As assumed, the captured CO_2 is compressed to 110 bar and transported via a 300-km pipeline for storage by injecting in gas filed as a representative transport distance for six pipelines constructed in North American (Vandeginste and Piessens, 2008). The pipeline's diameter is considered to be around 320 mm by taking an example located in Bravo Dome to Guymon with the similar capacity (around 4 Mton CO_2/year) (Vandeginste and Piessens, 2008), and the final designed diameter is set to be 355.6 mm with a thickness of 24 mm, with corresponds to a weight of 196.5 kg/m, following the national standard of GB 5310–2008 in China. The electricity requirement for CO_2 recompression along the pipeline has been considered by consuming 3 kW of electricity per km of CO_2 pipeline (Spath and Mann, 2004). The energy requirement for CO_2 injection to the reservoir is negligible owing to the high-pressure CO_2, and the CO_2 leakage is also assumed to be negligible with safe operation.

2.4.2. Data calculation

The life cycle GHG emissions can be calculated according to the following equation:

$$T = T_{pc} + \int_0^n \left(T_{rp} + T_{rt} + T_{cr} + T_{wt}\right) dt + \int_0^n \frac{C_{coal} y_c}{12} * 44 * \left(1 - \eta_{cap}\right) dt$$

where

$$\eta_{cap} = \frac{m_{CO_2}}{\frac{C_{coal} y_c}{12} * 44} \tag{1}$$

where T denotes total emissions, kg; T_{pc} denotes emissions during plant construction, including iG-CLC power plant construction and CO_2 pipeline construction, kg; T_{rp} denotes emissions during raw material production, namely coal mining and oxygen carrier manufacturing, kg/y; T_{rt} denotes emissions during raw material transport, kg/y; T_{cr} denotes emissions resulting from recompressing CO_2, kg/y; T_{wt} denotes emissions due to waste transport, kg/y; C_{coal} denotes coal consumption, kg/y; y_c denotes carbon fraction in coal, wet basis; η_{cap} denotes carbon capture efficiency, which is defined as the mass flow ratio between the captured CO_2 and CO_2 emissions from coal direct combustion (without capture); n denotes operating lifetime, y; m_{CO_2} denotes the mass flow rate of CO_2 in the captured CO_2 stream.

The emissions during plant construction (T_{PC}) can be calculated through the following equation.

Table 4
Plant material requirements (kg/MW plant capacity).

Material	Source (Spath et al., 1999)	Source (Spath and Mann, 2000)	In this work
Concrete	158758	97749	128253
Steel	50721	31030	40875
Aluminum	419	408	413
Iron	619	204	411

Table 5
CO_2 emissions form manufacturing construction materials.

Material	Concrete	Steel	Aluminum	Iron
CO_2 emission (kg/kg)	0.81 (Hendriks et al., 2004)	1.16 (Burchart-Korol, 2013)	22.4 (Norgate et al., 2007)	1.16 (Wang et al., 2010)

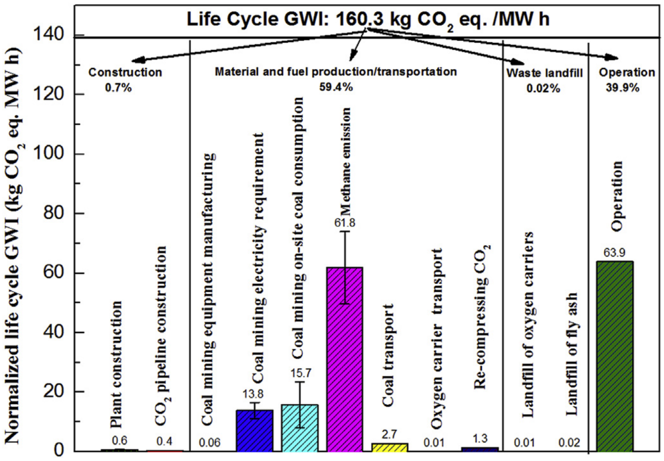

Fig. 2. Results for life cycle GWI analysis of the studied iG-CLC power plant.

$$T_{pc} = \sum_i P_e m_i e_i + m_{pipe,steel} * L_{pipe} * e_{steel} \tag{2}$$

where m_i denotes the requirement of construction materials, kg/MW, including concrete, steel, aluminum and iron; P_e denotes electricity output from the iG-CLC power plant, MW; e_i denotes the CO_2 emissions during manufacturing per kg of construction materials, kg/kg; $m_{pipe,\ steel}$, L_{pipe} and e_{steel} denote the steel requirement for pipe manufacturing (kg/m), length of pipeline (m) and CO_2 emissions during manufacturing per kg of steel for pipeline (kg/kg), respectively.

The emissions during raw material production (T_{rp}) can be expected as follows:

$$\begin{aligned} T_{rp} = {} & m_{cm,steel} e_{steel} + e_{MW\ h}\left(C_{e,cm} * C_{coal} * 10^{-6}\right) \\ & + e_{coal}\left(C_{coal,cm} * C_{coal} * 10^{-6}\right) \\ & + GWP_{methane}\left(e_{methane,\ cm} * C_{coal} * 10^{-3}\right) + \left(e_{oc} * C_{oc}\right) \end{aligned} \tag{3}$$

where $m_{cm,steel}$ denotes the steel requirement during coal mining equipment manufacturing, kg; $C_{e,cm}$ denotes electricity consumption per ton of coal mined, kW h/ton coal; $e_{MW\ h}$ denotes CO_2 emissions per MW h electricity produced for conventional coal-driven PC plant, assumed to be 876 kg/MW h(Cuéllar-Franca and Azapagic, 2015); $C_{coal,cm}$ denotes on-site coal consumption during coal mining, kg/ton coal; e_{coal} denotes CO_2 emissions per ton of coal consumed, recommend as 2.4 t/t_{ce} coal (Xia et al., 2010); $e_{methane,\ cm}$ denotes methane emissions during coal mining, kg/ton coal; $GWP_{methane}$ denotes global warming potential of methane; e_{oc} denotes emissions from manufacturing oxygen carriers for CLC, kg/kg; C_{oc} denotes oxygen carriers consumption, kg/y.

In terms of the emissions related to raw materials transport (T_{rt}), it is associated with both coal transport and oxygen carriers transport, and can be obtained via Equation (4).

$$T_{rt} = e_{transport}\, L_{transport}\left(C_{coal} + \frac{m_{oc} * Q_{LHV}}{t_{oc}} * t_{operation} * 10^{-3}\right) \tag{4}$$

where $e_{transport}$ denotes CO_2 emissions caused by truck vehicle per ton/kilometer, kg/(ton • km); $L_{transport}$ denotes the transport length, km; m_{oc} denotes the oxygen carriers inventory, kg/MW (heat input); Q_{LHV} denotes the heat input, MW; t_{oc} denotes the lifetime of oxygen carrier, h; $t_{operation}$ denotes the operational time of current iG-CLC power plant, h.

In relation to the emissions concerning on re-compression of CO_2 (T_{cr}) along the pipeline, it can be determined by the following equation.

$$T_{cr} = e_{MW\ h} * W_{cr} * 10^{-3} * t_{operation} \tag{5}$$

where W_{cr} denotes the work requirement for recompressing CO_2 along the pipeline, kW.

For emissions during spent oxygen carrier transport as well as fly ash (T_{wt}), these emissions can be calculated via:

$$\begin{aligned} T_{wt} = {} & e_{transport}\, L_{transport}\Bigg[C_{ash} + \bigg(\frac{m_{oc} * Q_{LHV}}{t_{oc}} * t_{operation} \\ & - m_{oc} * Q_{LHV}\bigg) * 10^{-3}\Bigg] \end{aligned} \tag{6}$$

where C_{ash} denotes the mass flow rate of fly ash, ton/y.

2.4.3. Limitation and uncertainties

Though the LCA is performed with the high quality data at the time of study, a number of data limitations and uncertainties are identified, mainly including the lack of up-to-date data sources and use of multiple types of data sources. Due to the nonexistence of large-scale iG-CLC demonstration plants by far, construction material requirements for LCA are taken from traditional coal power plant, resulting in uncertainties. The data sources from different countries further increase the uncertainties. Unlike traditional coal power plant, which concentrates influences on plant operational step, iG-CLC power plant may be sensitive to upstream and downstream-related emissions, particularly with the emissions resulting from OC manufacturing. This probably leads to uncertainties applied to different countries because of the distinct OC

Table 6
Emission factors during coal mining.

Item	Value
Steel requirement for equipment manufacturing (kg/tone coal per year)	2.755×10^{-3} (Katell and Hemingway, 1974)
Electricity requirement (kW h/tone coal)	26.7–34.4 (Xia et al., 2010; Zhuang and Jiang, 2009)
On-site coal consumption (kg/tone coal)	10.9–26.7 (Xia et al., 2010; Zhuang and Jiang, 2009)
Methane emissions (kg CH_4/tone coal)	4.23–5.01 (Katell and Hemingway, 1974; Xia et al., 2010)

manufacturing processes. Furthermore the data of global warming potential of GHG taking from old version (2nd) of IPCC report are consistent with previous studies associated with examining GWI in conventional coal power plants to make results comparable (as presented in Section 4.1).

2.5. Life cycle impact assessment (LCIA)

LCIA examines the environmental and human health effects with a range of categories, such as ozone depletion, GWI, and radionuclides. As a preliminary step to carry out a LCA for an iG-CLC power plant, GWI is solely considered in this study. CO_2, CH_4 and N_2O are three representative GHG to examine GWI, however very few NO_x formation was observed in CLC process by our previous study. In terms of lignite based iG-CLC process, NO_2 and NO emissions account to be 0.18 kg/MW h and 3.86 kg/MW h, respectively, and N_2O emissions are rarely observed (Fan et al., 2017b). Therefore only CO_2 and CH_4 emissions are considered herein, and the common characterization factor is set to be CO_2-equivalent for the GWI analysis.

3. Results

3.1. Thermodynamic performance

The process diagram and the detailed stream properties are provided in the Supporting Information. Table 7 presents the performance results of the studied iG-CLC power plant. It can obtain a net efficiency of 37.7% with 93.5% CO_2 capture efficiency as presented in Section 2.3.1. It should be noted that higher CO_2 capture efficiency of iG-CLC plant (e.g. exceeding 95%) is possible if the operating conditions are favored to improve carbon conversion rate such as increase in FR operating temperature (most significantly) or increase in carbon stripper efficiency (Pérez-Vega et al., 2016). However in this study, we consider the feasibility of auto-thermal operation of iG-CLC process; the FR temperature is determined by the sensible heat of the heated ilmenite and no supplementary heat is provided. Auto-thermal operation is hard to achieve as an example of the 1 MW_{th} iG-CLC rig erected at Technical University of Darmstadt was failed to meet the FR operating temperature goal of 900 °C by auto-thermal operation (Ströhle et al., 2015). Therefore the carbon capture efficiency obtained in this study is not maximized, but the highest at auto-thermal operation. Besides it should be noted that this optimistic performances of iG-CLC plant are obtained at thermodynamically optimum condition (i.e. the best case).

3.2. GWI results

Fig. 2 presents the life cycle GWI analysis results. Notably, the normalized life cycle GWI is achieved by functional unit (shown above) that enables to obtain the greenhouse gas emissions within 1 MW h of electricity output from the power plant. Clearly, the life cycle GHG emission is equal to be 160.3 kg CO_2-equivalent/MW h, of which the main contributor comes from plant operation process and material and fuel production/transportation process, leading to 39.9% and 59.4% of life cycle emissions, respectively. The emissions resulting from plant operation stem from burning unreacted char in AR and work required for recompression of captured CO_2 along the pipeline transport, leading to CO_2 emissions to atmosphere. However it should be noted reduction of CO_2 emissions in AR is possible by adjusting system parameters as discussed below. The emissions from material production process mainly result from methane emission, contributing to 61.8 kg CO_2 eq./MW h. In terms of construction process, emissions are negligible. The transport process remains a moderate contribution to the total emissions, mainly assigning by raw material transport and re-compression of CO_2 along the pipeline. Landfill of spent oxygen carriers as well as fly ash accounts for only 0.02% of life cycle GHG emissions. For low-carbon iG-CLC technology, majority of the emissions result from upstream operation (e.g. coal preparation) and unit operation, depending on the operational performance, such as carbon conversion efficiency and lifetime of OC, discussed below. In this study, the operation-resulted emissions rank the highest, followed by the methane emissions during coal mining. Therefore it presents greatest potential to reduce emissions during operation step, such as increasing the carbon stripper separation efficiency, elevating fuel reactor temperature, etc. In parallel, collection of released methane from coal mining process has been deemed as an effective method to reduce GHG emissions though it faces difficulties in coal mining field.

Notably, the error bars shown in this figure (calculating the errors between the maximum and minimum data input shown in Table 6) present that the methane emission during coal mining, is most sensitive to uncertainties, followed by coal mining on-site coal consumption and electricity requirement.

Table 7
Performance of the studied iG-CLC power plant.

Variable	Units	Value
Fuel input (LHV)	MW	1625
Energy penalties		
Air compression	MW	655.5
Water pumping	MW	1.7
CO_2 compression	MW	63.67
CO_2 re-compression	MW	0.9
CO_2 capture capacity	ton/h	556.9
CO_2 capture efficiency	%	93.5
CO_2 emissions (operation)	kg/MW h	63.9
Ash production	ton/h	90.7

4. Discussion

In this Section, we first present a comparison of life-cycle GWI between conventional coal-driven plants with and without carbon capture and iG-CLC plant in order to demonstrate the superiority of CLC process. Then we discuss the effects of operating conditions towards lowering down life-cycle GWI. Generally, the parameters taken into consideration should involve steam-to-carbon (S/C) and OC-to-fuel (ϕ) ratios (influencing FR operating temperature by autocrat-thermal operation) that affect char conversion. However as highlighted by Raúl et al. (Pérez-Vega et al., 2016), solid inventory in FR was an another important parameter as fundamentals of CLC to affect the whole carbon capture efficiency by determining the residence time of particles in FR even though this factor had been experimentally demonstrated to be less influential than other parameters (such as FR temperature). In this study, because of the consideration of both phase and reaction equilibrium conditions (i.e. the reaction time is long enough to meet reaction equilibrium), the effect of solid inventory in FR on life-cycle GWI is not reachable.

4.1. Comparison with other coal-based power plants

For further study, comparisons in terms of life cycle GWI between iG-CLC and other coal-based power plants are conducted, as shown in Fig. 3.

The life cycle GWI for other coal-based power plants (including pulverized coal (PC), oxy-fuel, integrated gasification combined cycle (IGCC)) are the range value reported in different studies (with

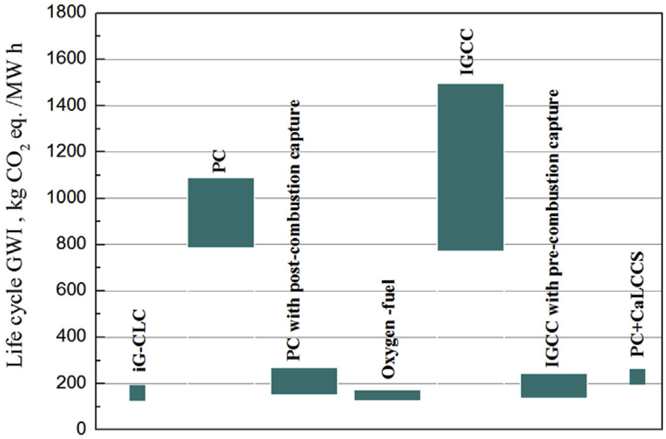

Fig. 3. Comparison with other coal-based power plant; data taken from (Cuéllar-Franca and Azapagic, 2015; Hurst et al., 2012).

different operating conditions and scales) as summarized in Ref. (Cuéllar-Franca and Azapagic, 2015). The system boundaries of these studies are consistent with this study including extraction and supply of fossil fuels to power plant, power generation, and CCS. Another reference system is PC coupled with post calcium looping for CCS (PC + CaLCCS). Obviously, the life cycle GWI of iG-CLC plant obtained in this study (thermodynamically optimum case) ranks the lowest (160.3 kg CO_2 eq./MW h) except for oxy-fuel combustion (151 kg CO_2 eq./MW h).

Though PC plants with carbon capture are almost competitive with iG-CLC power plant to obtain lower GWI, huge energy penalty is paid for carbon capture. While few energy penalty (energy penalty for rising steam as gasification agent) is expected in iG-CLC power plant for carbon capture with a net power efficiency of 37.7% in this study. For example, the net power efficiency drops from 43.7% without capture in a PC plant (advanced super-critical steam cycle) to 33.1% with post-capture (Tola and Pettinau, 2014). And approximately 17% of energy penalty is required in an oxy-fuel combustion process to obtain almost pure oxygen from air separation (Vasudevan et al., 2016). Yet PC + CaLCCS process is emerged as a good option for carbon mitigation, the energy penalty associated with calcium sorbent regeneration significantly reduces the net power efficiency to approximately 31% (Hurst et al., 2012). Besides the cost of electricity (COE) by adopting these capture methods has been proved to be higher than CLC (Rubin et al., 2015).

4.2. *Effect of steam to carbon ratio (S/C)*

The effect of S/C (defined in Equation (7)) determines the carbon conversion, and its influence on life cycle GWI has been shown in Fig. 4.

$$\frac{S}{C} = \frac{additional\ steam\ (molar\ basis) + water\ \ content\ in\ coal\ (molar\ basis)}{carbon\ content\ in\ coal\ (molar\ basis)} \tag{7}$$

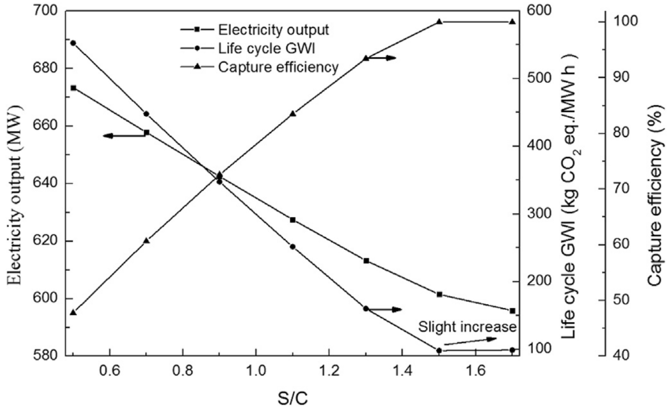

Fig. 4. Effect of S/C on life cycle GWI.

Obviously, increase in S/C benefits carbon conversion, as the capture efficiency increases from 47.8% at S/C = 0.5 to about 100% at S/C = 1.5, where the char is fully converted into in-situ syngas. Therefore further increase in S/C results in flat change of life-cycle GWI and carbon capture efficiency. On the other hand, the net electricity output is decreased with S/C owing to the supplementary energy requirement for steam generation. The life cycle GWI first decreases with S/C to a minimum value of 98.3 kg CO_2 eq./MW h and then increases slightly because of the further reduction of electricity output. Thermodynamically, complete carbon conversion can be achieved in this study when S/C > 1.5. However, S/C is chosen to be 1.3 herein, corresponding to capture efficiency of 93.5% and life cycle GWI of 160.3 kg CO_2 eq./MW h, because of the following reasons: (1) Capture efficiency at this point is close or even higher compared with current “first-generation” CCS plants, e.g. MEA (Monoethanolamine)-scrubbing PC plant with capture efficiency of 85–90% (Davison, 2007; Rubin et al., 2007); (2) Further increase in S/C reduces net electricity output (the decrease becomes less steep).

4.3. *Effect of oxygen carrier to fuel ratio (ϕ)*

The parameter ϕ has a determined influence on FR operating temperature owing to the FR is operated at endothermic condition and the required heat is supplied by sensible heat of OC. It is defined as the availability of oxygen in the flow of oxygen carrier divided by the oxygen requested to completely convert the coal to CO_2 and H_2O (Abad et al., 2013).

$$\varnothing = \frac{R_{o,ilm}F_{OC}}{\Omega_{coal}} \tag{9}$$

where $R_{o,ilm}$ is the oxygen transporting capacity of ilmenite, F_{OC} is the flow rate of ilmenite, Ω_{coal} is the oxygen demanding of coal.

The oxygen demanding of coal, Ω_{coal}, is defined as follows:

$$\Omega_{coal} = M_o\left(2\frac{[C]_{coal}}{M_c} + 0.5\frac{[H]_{coal}}{M_H} - \frac{[o]_{coal}}{M_o}\right) \times m_{coal} \tag{10}$$

where $[C]_{coal}$, $[H]_{coal}$ and $[o]_{coal}$ correspond to the carbon, hydrogen and oxygen fraction in the coal, which are obtained from ultimate analysis.

The life cycle GWI as a function of ϕ is shown in Fig. 5. Clearly,

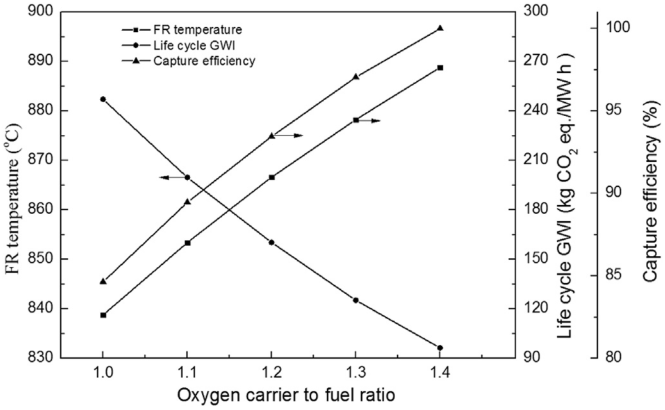

Fig. 5. Effect of ϕ on life cycle GWI.

the higher ϕ is, the higher FR operating temperature results. With the increase in FR temperature, carbon is favorably converted assigning to the endothermic carbon gasification reaction ($C + H_2O{=}CO + H_2$). Consequently both capture efficiency and life cycle GWI are promoted towards their positive direction, as capture efficiency increases from 84.6% to 100% and GWI decreases from 247.0 to 96.4 kg CO_2 eq./MW h. The ϕ is recommend to be 1.2 in this study to avoid cost imposed on OC transport between two reactors, and is also suggested by Cuadrat et al. (2012d). Definitely, further increase in ϕ is beneficial to rise FR temperature, and therefore reduces the CO_2 emissions in AR. Notably, as illustrated previously, because of the consideration of reaction equilibrium (without consideration of residence time) and the strong effect of FR temperature on carbon conversion rate (lowering down carbon emissions as temperature rises), the increase in ϕ leads to improvement of carbon conversion rate. This phenomena is against the experimental results obtained from (Pérez-Vega et al., 2016) that carbon capture efficiency is reduced as a fact of increasing ϕ reduces the residence time of particles in FR with constant operating temperature.

4.4. Effect of oxygen carriers

Several types of oxygen carriers have been screened based on Ni, Cu, Fe, Mn, Co, as well as other mixed oxides and low cost materials (e.g. natural ilmenite). The choice of OCs suitable for iG-CLC process requires reasonable reactivity, enough attrition rate and resistance to impurities in coal (fuel-N and fuel-S). Nickel-based OC is not considered because its reaction with fuel-S to form Ni_3S_2, consequently resulting in SO_2 release in the AR (García-Labiano et al., 2009). In this study, Fe-based oxygen carrier is chosen to determine the effect of other OCs on life cycle GWI, taken ilmenite-based case as reference. Unlike naturally existed ilmenite, Fe-based OC is artificially synthesized. The global warming potential of manufacturing metal Fe is predicted to be 1.16 kg CO_2 eq./kg (Burchart-Korol, 2013). The Fe-based OC is composed by Fe_2O_3 40 wt % with Al_2O_3 as inert supporter and its lifetime is considered to be 1600 h depending on the preliminarily experimental results (Wu et al., 2010). In this comparison, the operating temperature of AR is remained to be consistent with that in ilmenite-based case, and the results are shown in Fig. 6 (left). The power efficiency in both cases remains very close, which is realized by same turbine inlet temperature (i.e. AR operating temperature). Though manufacturing Fe-based OC brings extra emissions during OC manufacturing, the life cycle GWI has been reduced by approximately 39%, mainly attributed by the increase in carbon conversion benefited from higher FR temperature since use of Fe_2O_3/Al_2O_3 is easier to achieve auto-thermal operation (the heat capacity of Al_2O_3 is high).

To further analyze the influence of Fe-based OC manufacturing on life cycle GWI, its lifetime is studied, and the sensitive analysis result is shown in Fig. 6 (right). Clearly, the life cycle GWI is dependent on lifetime of Fe when it is less than 2000 h, beyond that range, the GWI is decreasing, but very slowly. Indeed the relationship between lifetime of OC and GWI obeys the inverse function. As shown clearly, with less lifetime of OC (<1000 h), the OC-manufacturing-related emissions are significant. Within the interval of 1000–2000 h, decreasing tendency has been narrowed down in comparison with previous decrement. The hint here is, in terms of decreasing GWI, the lifetime of OC should be initially reaching the aim of above 1000 h to avoid huge OC-manufacturing-related emissions, then pursuing the goal of 2000 h to reach the moderate point. Meanwhile the results obtained here suggest that further improvement of the lifetime of Fe-based OC adds no significant reduction on life cycle GWI. Fe-based OC instead of ilmenite provides reduction on life cycle GWI, but nowadays much attention is paid on using ilmenite as promising OC because of its naturally-existence and cheap price characters.

5. Conclusion and further research needs

The life cycle GWI assessment of iG-CLC power plant offers a first attempt at examining the GWI of this advanced carbon capture technology through its lifetime. The main conclusions rising from

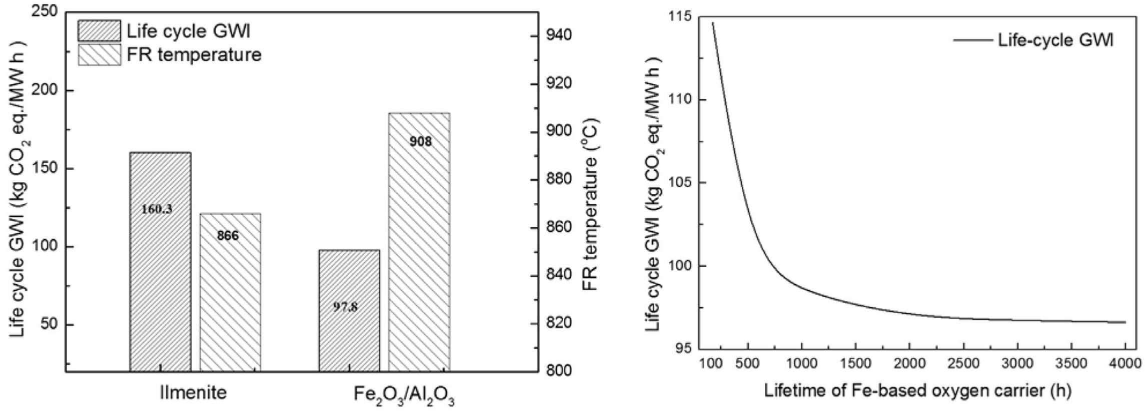

Fig. 6. Effect of different oxygen carriers (left) and lifetime of Fe-based oxygen carrier (right) on life cycle GWI.

this study include: (1) Improvement of coal char conversion is essential to reduce life-cycle GWI, and in order to achieve this goal, increasing steam-to-carbon ratio and OC-to-fuel ratio have been observed to be efficient under auto-thermal operation of iG-CLC process; (2) The thermodynamic performance of iG-CLC plant has certain effect on life-cycle GWI and it turns out to be higher power efficiency leads to lower overall carbon emissions; (3) The lifetime of artificially manufactured Fe exhibits influences on life-cycle GWI until it reaches 2000 h in this study.

Based on the assumptions and the results obtained in this study, we emphasize below two recommendations for further research needs: (1) development of consistent data sources from one nation to perform LCA is benefitted to reduce uncertainties; (2) assessments of a large variety of environmental impacts for iG-CLC power plant are necessary to support future energy policy-making.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2019.06.082.

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