

Quantifying the role and value of chemical looping combustion in future electricity systems *via* a retrosynthetic approach

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ABSTRACT

Carbon capture and sequestration of CO₂ from the combustion of fossil fuels in thermal power plants is expected to be important in the mitigation of climate change. Deployment however falls far short of what is required. A key barrier is the perception by developers and investors that these technologies are too inefficient, expensive and risky. To address these issues, we have developed a novel retrosynthetic approach to evaluate technologies and their design based on the demands of the system in which they would operate. We have applied it to chemical looping combustion (CLC), a promising technology, which enables carbon dioxide emissions to be inherently captured from the combustion of fossil fuels. Our approach has provided unique insight into the potential role and value of different CLC variants in future electricity systems and the likely impact of their integration on the optimal capacity mix, the operational and system cost, and dispatch patterns. The three variants investigated could all provide significant value by reducing the total investment and operational cost of a future electricity system. The minimisation of capital cost appears to be key for the attractiveness of CLC, rather than other factors such as higher efficiency or lower oxygen carrier costs.

1. Introduction

Climate change brought about by the anthropogenic emission of greenhouse gases (GHG), in particular CO₂ (Archer, 2005), is one of the greatest challenges of the 21st century (IPCC, 2014). Strategies to reduce emissions of CO₂ include reducing energy consumption, switching to fuels with a higher ratio of hydrogen to carbon in their composition, increasing the use of renewables and nuclear energy, enhancing the natural, biological sequestration of CO₂ and carbon capture and storage (IEA, 2013; IPCC, 2005). Despite having access to a suite of mitigation options (International Energy Agency, 2012; Pacala and Socolow, 2004), progress remains slow, relative to what is known to be required to meet the ambitious targets to which the world has agreed (United Nations Framework Convention on Climate Change, 2015).

A key barrier to deployment is the perception that, in the absence of subsidy, emerging energy technologies are too expensive or inefficient to be attractive to investors. They are also perceived as risky since they are not proven at a commercial scale. This is particularly the case for large-scale technologies such as power plants with carbon capture and

storage (Oxburgh, 2016). Another important problem with electricity systems which affects governments and technology developers is deciding which technologies should be implemented, given that only certain combinations are possible because back-up capacity is needed for intermittent generators and strict carbon constraints must be met. Specifying what is likely to be required of technologies by the system is therefore also difficult since it is dependent on the combination that is installed. Generally, only targets for cost reduction are set (Offshore Wind Programme Board, 2016). This has been sufficient for the case of intermittent renewables that are generally able to feed electricity into the system whenever it is available. In other cases e.g. thermal power plants, it leaves developers hamstrung since further information is important such as load-following requirements and the capacity factors at which the plants are likely to operate.

For the reasons mentioned, many emerging technologies are unable to cross, what is commonly known as the ‘Valley of Death’, the gap between R&D and commercialisation. In this paper we seek to address these issues by using a novel approach, which we call ‘retrosynthetic analysis and technology design’. It is termed ‘retrosynthetic’ since the

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Nomenclature

BECCS	Bio-energy with carbon capture and storage
CAPEX	Capital expenditure
CCGT	Combined cycle gas turbine
CCS	Carbon capture and storage
CLC	Chemical looping combustion
CLOU	Chemical looping with oxygen uncoupling
ESO	Electricity systems optimisation
GenSto	Battery storage
iG	<i>In situ</i> gasification
IGCC	Integrated gasification combined cycle

InterImp	Interconnection (import)
InterSto	Interconnection (storage)
IRR	Internal rate of return
NGCC	Natural gas combined cycle
OC	Oxygen carrier
OCGT	Open cycle gas turbine
OPEX	Operating expenditure
PHSto	Hydro storage
SD	System demand
TL	Transmission losses
TSC	Total system cost

approach is able to guide the synthesis, *i.e.* the design and development, of a technology so that it is optimised based on the requirements and demands of the system within which it would operate.

In our approach, a technology and its design is evaluated in the context of the system in which it might operate. This is first done using typical parameters for the technology to identify the role and value of the base case. Due to the uncertainty in the various parameters of a technology, an analysis follows where certain physical and techno-economic parameters are varied within physically feasible ranges and based on relevant understanding and modelling from the process and sub-process scales. In this way key performance envelopes, sensitivities and normative budget guidelines can be identified. This detailed insight allows the current and future potential of the technology to be understood and so modifications to the design of the technology can be suggested, which would maximise its attractiveness for deployment. The key research activities required to achieve these modifications can also be highlighted. A schematic summarising the retrosynthetic approach is given in Fig. 1.

In this paper, our approach has been applied to a promising technology for the production of low carbon electricity – chemical looping combustion (CLC). Modelling and simulations have been developed and on the experimental side, the CLC process has been proven up to 1 MWth (Ströhle et al., 2014a,b) and 3 MWth for a limestone CLC process (Andrus et al., 2012). While such work gives confidence that CLC technologies could in principle be implemented at full scale, it does not identify constraints arising when the technology is deployed in an electricity system and cannot determine whether deploying CLC in future is worthwhile. This paper represents the first evaluation of this technology in the context of an electricity system. The application of our approach furthermore enables us to address the aforementioned issues and identifies a number of key research areas, apart from those

associated with scale-up (Gauthier et al., 2017; Lyngfelt and Leckner, 2015), to be tackled.

The remainder of this paper is structured as follows. Section 2 introduces chemical looping combustion and reviews the different variants of CLC available for electricity generation. Section 3 describes the electricity system model used and the three variants of CLC investigated. The results of the analysis are presented in Sections 4 and 5 and their implications are discussed in Sections 6 and 7. Conclusions are drawn in Section 8.

2. Chemical looping combustion

Carbon capture and storage (CCS) involves the capture of CO₂ arising from the combustion of fossil fuels and its subsequent permanent storage in a suitable location, *e.g.* depleted oil reservoirs or saline aquifers. If CCS were widely implemented in the power and industrial sectors, carbon capture and storage (CCS) could reduce mitigation costs by 27.5% (Victor et al., 2014), compared to scenarios where it is not used. Three conventional approaches exist for capturing CO₂: post- and pre-combustion capture and oxy-fuel combustion (Boot-Handford et al., 2014; IEA, 2013; MacDowell et al., 2010).

Conventional CCS technologies require the addition of process elements, to conventional thermal power plants, leading to increased capital cost and reduced efficiency. CLC, at a marked change in the mode of combustion over conventional technologies, inherently produces a stream of CO₂ that is suitable for sequestration. The reduction in efficiency is therefore smaller than for other CCS technologies (DOE/NETL, 2014; Petrakopoulou et al., 2011; Zhu et al., 2015). Many authors have suggested that CLC is one of the most economical for CO₂ capture (Adanez et al., 2012; Bhawe et al., 2014; Ekström et al., 2009; Fan et al., 2012; IPCC, 2005; Kerr, 2005), as well as having a lower environmental

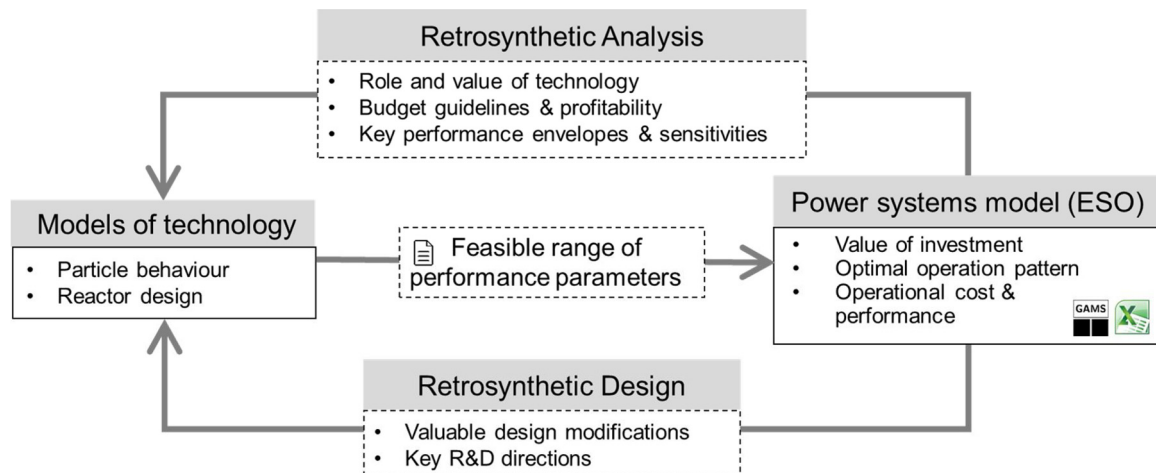
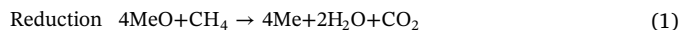


Fig. 1. Schematic diagram of the retrosynthetic approach. The arrows denote the flow of information. The dashed boxes describe the type of information that is passed. The solid boxes describe the outputs of models.

impact, certainly less than solvent-based processes (Petrakopoulou et al., 2011). In particular, NO_x emissions are avoided (Bayham et al., 2013; Ishida and Jin, 1996).

CLC is based on the redox cycling of a transition metal oxide in particulate form, known as an oxygen carrier (OC). The fuel can be in gaseous, liquid or solid form (Adanez et al., 2012; Adánez et al., 2017). The process can be illustrated by the combustion of methane:



where Me represents an appropriate transition metal, such as iron, copper, nickel or manganese (Adanez et al., 2004; Cho et al., 2004). To utilise the reactions, the OCs are brought into contact with the fuel in their oxidised form, so that the lattice oxygen in the solid reacts with the fuel via Reaction (1). The gaseous product, after removal of H₂O, is a pure stream of CO₂, suitable for sequestration. The reduced OC is regenerated by re-oxidising it in air, Reaction (2). Overall, the fuel has been burnt in air (Reaction (3)), so the total enthalpy change is the same as for conventional combustion. In principle, the proportion of carbon captured is 100%, since the separation of CO₂ is inherent to the process. In conventional separation technologies, typically only 85–90% of the carbon dioxide emissions are captured (Petrakopoulou et al., 2011). Some authors have more recently suggested that capture rates of up to 95% are possible without an excessive increase in cost or decrease in efficiency (Dickmeis and Kather, 2014; Flø et al., 2016). Entropic barriers mean that higher levels of capture are technically challenging and unlikely to be economically viable.

The most common configuration for CLC involves two interconnected fluidised bed reactors (Fig. 2), in which the two sub-reactions (Eqs. (1) and (2)) are separated spatially (Lyngfelt et al., 2001). The OC is reduced in the fuel reactor and re-oxidised in the air reactor. The main drawback of a fluidised bed system is the need to transport large volumes of solids, which consumes energy and leads to attrition of the OC. The separation of gas and solids at high pressure is also a challenge. The design of first plants of this type would probably be based on circulating fluidised bed combustors, which are in operation at industrial scale worldwide (Lyngfelt and Leckner, 2015). If the fuel is gaseous, the sub-reactions can instead be separated temporally in packed bed reactors by switching the gas feed periodically between reducing and oxidising conditions (Noorman et al., 2007). This allows particle circulation to be avoided, but temperature control and gas switching at high temperatures and pressures are significant challenges.

Whilst CLC can also be employed for the generation of low carbon steam on chemical plants and refineries, in the case of electricity generation, CLC can be categorised according to the fuel combusted, the mode of combustion and the power cycle it is integrated with, as depicted in Fig. 3 (Nandy et al., 2016).

2.1. Generation of electricity using gaseous fuels

For the combustion of gaseous fuels (primarily natural gas), the arrangement proposed most commonly is CLC coupled to a combined cycle (NGCC-CLC), as depicted in Fig. 4. Compressed air from a gas turbine compressor is fed to the air reactor and air depleted of oxygen leaving the air reactor is expanded in a gas turbine followed by a heat recovery steam generator (HRSG) to generate steam for a steam turbine. The CO₂-rich stream from the fuel reactor is also sent to a HRSG. Typical temperatures in the air and fuel reactors are between 850 and 1200 °C, depending on the OC material, with pressures proposed in the range 0–20 barg.

Petrakopoulou et al. (2011) modelled a NGCC-CLC power plant and compared it to natural gas combined cycle (NGCC) power plants with capture using amine scrubbing and without CO₂ capture. In the NGCC-

CLC plant, the turbine inlet temperature (TIT) was 1200 °C and the OC material was NiO, employed for its high rate of reaction and durability. The efficiencies were 51.3, 45.8–48.2 and 56.3% (LHV) respectively. The levelised costs of electricity (LCOE) of the NGCC-CLC plant and the NGCC plant with capture using amine scrubbing were similar, at €92/MWh and €92/MWh–€96/MWh (2011 prices) respectively. In comparison, a conventional NGCC plant without capture had a LCOE of €74/MWh (2011 prices). Ekström et al. (2009) predicted a thermal efficiency of 51–52% (LHV) for an NGCC-CLC plant, compared to 56.2% for NGCC without carbon capture. The LCOE was predicted to be 26% higher than without capture. CLC is typically found to be approximately 4 percentage points less efficient than a NGCC power plant without CO₂ capture, compared to a 6–8 percentage point drop for a plant with post-combustion capture by amine scrubbing of flue gases.

The TIT has a significant effect on the thermal efficiency of a combined cycle (Consonni et al., 2006). When such a cycle is coupled to CLC, the TIT is limited by the maximum temperature that the OC material can withstand. The melting points of Fe and Ni are 1538 and 1455 °C respectively. Cu-based materials are limited to lower temperatures (typically 900 °C). The melting point of Cu is only 1085 °C. Generally the metal oxides have higher melting points e.g. Fe₂O₃ and Fe₃O₄, the metal oxides that an iron-based material would cycle between due to thermodynamic limitations, have melting points of 1565 °C and 1597 °C respectively. A number of studies have investigated the effect of using Fe or Ni. The effect on efficiency was found to be no more than 0.5 percentage points and was mainly due to differences in oxygen carrying capacity and heats of reaction in the air and fuel reactors. The effect on efficiency is small compared to the effect of changing the TIT (Mantripragada and Rubin, 2016; Wolf et al., 2005). As a result, cost, durability and non-toxicity are likely to dictate the choice of transition metal.

Porrazzo et al. (2016) investigated the impact of the cost and lifetime of the oxygen carrier material (Ni-based) and the cost of fuel (CH₄) on the LCOE of CLC. They found that, above a carrier lifetime of 1000 h, its cost made only a small contribution to the LCOE. As with all thermal plants, the cost of the fuel had a significant impact on the LCOE, ranging from \$80/MWh to \$114/MWh (2015 prices) for fuel costs ranging from \$3.8/GJ to \$8.8/GJ. Linderholm et al. (2008) have reported lifetimes of 4000 h for Ni-based particles with methane as fuel.

Naqvi et al. (2007) conducted a part-load analysis of a NGCC-CLC power plant and found that it showed better relative part-load efficiency compared to conventional combined cycles. At 60% load, the drop was 2.6 percentage points from a peak efficiency of 52.2% at full load. An advanced control strategy was necessary.

Other power cycles are also possible. Petriz-Prieto et al. (2016) considered 15 combinations of CLC systems, power cycles and OCs. A conventional CLC arrangement (two, interconnected fluidised beds) was found to be the most favourable. CLC with a steam cycle gave the lowest efficiency (47.5%), followed by one with a steam injected gas turbine (48.1%). The highest efficiency was achieved by CLC with a

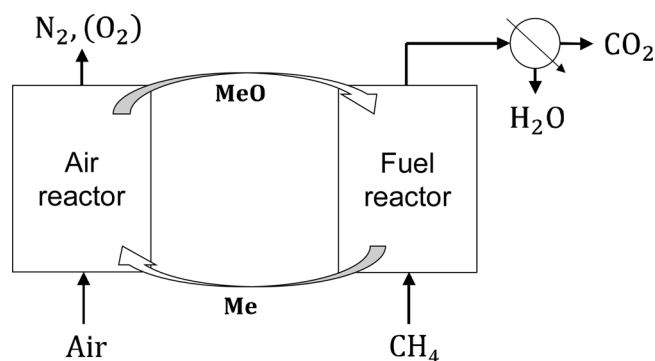


Fig. 2. CLC with two interconnected fluidised bed reactors.

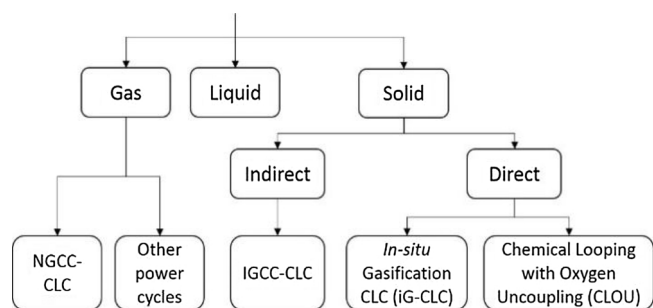


Fig. 3. Classification of CLC according to the fuel used and the combustion process.

humid air turbine cycle (HAT) (53.8%), a concept originally suggested by Ishida and Jin, (1994) to increase efficiencies. The efficiencies quoted are averages of the nine different pressure (10, 20, 30 bar) and temperature (1020, 1050, 1350 °C) combinations used by the authors. An increase in reactor pressure and temperature generally led to an increase in efficiency.

2.2. Generation of electricity using solid fuels

Solid fuels can be combusted directly in the fuel reactor, *in situ* gasification (iG-CLC) or indirectly by gasifying them *ex-situ* and combusting the resulting syngas, integrated gasification combined cycle (IGCC-CLC). IGCC-CLC is therefore similar to NGCC-CLC, except that the fuel is syngas rather than methane (Fig. 4). A typical scheme for iG-CLC is depicted in Fig. 5.

When CLC is applied to an IGCC plant with carbon capture, it replaces the water-gas shift reactor, the physical absorption process and the syngas combustor. Instead syngas, after removal of sulfur and ash, is combusted directly in the fuel reactor. The exhaust gases are at high temperature and pressure and are used in a combined cycle to generate electricity, similar to NGCC-CLC. Rezvani et al. (2009) assumed a Ni-based carrier in their study. They estimated the efficiency to be 35.2% (LHV) and the breakeven cost of electricity to be €86/MWh (2009 prices). Zhu et al. (2015) compared IGCC using CLC with a Ni-based carrier (60 wt% NiO on alumina) to a calcium looping process, physical absorption-based system and one with no capture and obtained efficiencies of 39.8, 37.7, 36.6 and 44.1% (LHV) respectively. Cormos (2015) investigated an ilmenite-based system with 20% biomass and coal as fuel. By combusting biomass in CLC, negative carbon dioxide emissions could be achieved. The efficiency was 38.0% (LHV) and the LCOE was €77/MWh (2015). For comparison, without carbon capture the efficiency was 46.1% (LHV) and the LCOE was €54.1/MWh. With post-combustion carbon capture using amine solvents, the efficiency

was 36.9% (LHV) and the LCOE was €77/MWh.

Hamers et al. (2014) compared the operation of IGCC-CLC with *ex-situ* gasification using either two interconnected fluidised bed reactors or dynamically-operated packed bed reactors. They concluded that the efficiency was the same in both configurations (42% LHV) and that the choice therefore depends on the relative availability, operability and cost of the two different reactor systems. Spallina et al. (2013) reported a similar efficiency for IGCC-CLC using packed bed reactors.

A high TIT is important for the efficiency of an IGCC-CLC system (Erlach et al., 2011; Jiménez Álvaro et al., 2015). Jiménez Álvaro et al. (2015) also found that 20 bar was the optimal pressure for the CLC reactor system. Mukherjee et al. (2015) compared different OC materials in an IGCC-CLC process. They found that OCs with a higher enthalpy of reaction for oxidation with air and higher melting temperatures were preferred and gave slightly higher efficiencies (up to 0.7 percentage points). The absolute efficiencies were quite low (maximum of 34.3%) since the temperatures were limited to 750 and 950 °C in the fuel and air reactors respectively. This was to allow OCs with a low melting point, e.g. Cu-based, to be included in the comparison. The Fe-based OC came out as most favourable, followed closely by the Ni-based one.

Where the solid is combusted in the fuel reactor, after the initial volatiles release, the process involves gasifying the solid fuel *in-situ* (iG-CLC) so that the resulting syngas reacts with the lattice oxygen in the OC. The rate of reaction is usually limited by the slow rate of gasification. Some OCs e.g. Cu-based, can release gas-phase oxygen, which can react directly with the solid fuel, eliminating the need for the slow gasification step. This is known as chemical looping with oxygen uncoupling (CLOU). In the literature most work has considered the iG-CLC and CLOU processes with the reactors at ambient pressure, coupled to a steam cycle. These processes can be conducted at elevated pressure (Xiao et al., 2010). Hazardous materials e.g. Ni should be avoided since the OC material will contaminate the ash.

Ekström et al. (2009) studied CLC in a circulating fluidised bed (CFB) with a steam cycle with bituminous coal and petroleum coke. They predicted efficiencies of 41.6–41.7% (LHV) compared to 45% for a conventional pulverised coal power plant. The efficiency was the same as that obtained by Authier and Le Moullec (2013), also with a steam power cycle. The cost of generation was predicted to be 19 and 9% higher, for coal and petroleum coke respectively, than for a conventional pulverised fuel power plant with no carbon capture. In terms of operation, they found that the load change characteristics of a CLC-based system should be similar to a conventional CFB combustor, with minimum loads of 45–50% compared to 40% for a conventional CFB and slightly longer start-up times. The National Energy Technology Laboratory (NETL) presented a CLC reference plant design and sensitivity study (DOE/NETL, 2014). The study was for a coal-fired CLC

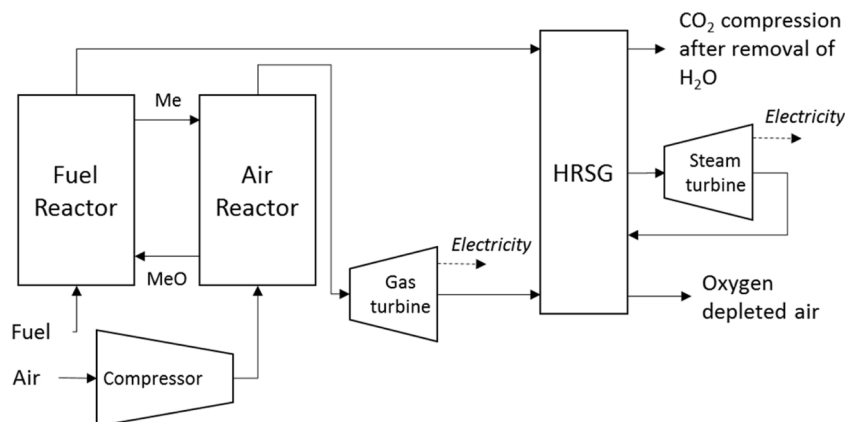


Fig. 4. Typical NGCC-CLC power plant scheme. In some arrangements there is a CO₂ turbine before the stream from the fuel reactor enters the HRSG. IGCC-CLC schemes are similar, except that the fuel is syngas from gasification of solid fuel rather than methane.

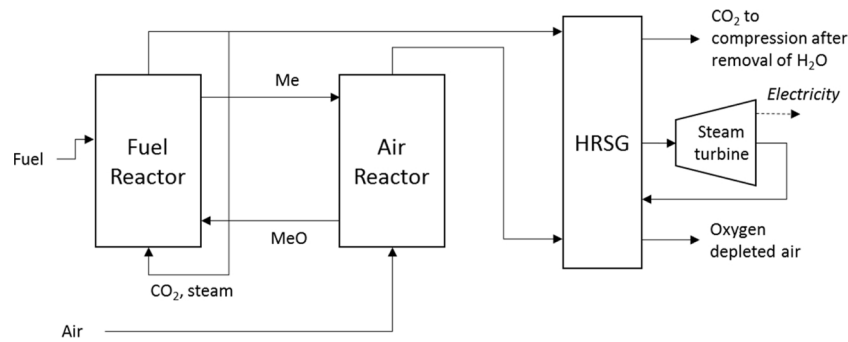


Fig. 5. Typical iG-CLC or CLOU power plant scheme.

power plant based on a CFB design coupled to a steam cycle. The OC was synthetic and Fe-based. The predicted efficiency was 36.4% (LHV coal) and the LCOE was \$115/MWh (2011 prices). For comparison, a reference plant design for a pulverised coal (PC) with carbon capture by amine scrubbing had an efficiency of only 29.5% (LHV coal) and a LCOE of \$137/MWh.

Spinelli et al. (2016) studied the integration of CLOU in a coal-fired power plant with a Cu-based OC. They obtained an efficiency of 42% (LHV), 2.5 percentage points below the reference plant with no carbon capture, but 5 percentage points higher than an oxy-fuel CFB plant.

A number of advanced options allowing the co-production of electricity and H₂ have also been suggested (Cormos et al., 2014; Fan et al., 2012). For example, Cormos (2015) proposed an ilmenite-based system to produce 400–500 MWe and up to 200 MWh H₂ using biomass. To enable co-production, a third reactor was added to the system, where the carrier was partially re-oxidised with steam before the oxidation was completed in the air reactor. This plant concept is very flexible, since it can operate continuously at full load, but diversify the generated energy vectors depending on the grid demand.

3. Modelling

For the electricity system model, a mixed-integer linear program was developed to determine simultaneously the optimal structure and dispatch schedule of a power system on a national scale. The model formulation simultaneously addresses capacity expansion of the power system and unit commitment. It focussed on a detailed representation of power plant operation while including system operability, reliability, and carbon emission constraints. The high temporal granularity made it possible to get an insight into the expected operational patterns and interactions between power generation and storage technologies in a future mix. A detailed mathematical formulation and validation of this Electricity Systems Optimisation (ESO) model has been published

(Heuberger et al., 2017a,b, 2016), and, for completeness, the governing equations are provided in the Appendix A of this paper. All data inputs are available for download at <https://www.imperial.ac.uk/a-z-research/clean-fossil-and-bioenergy/downloadable-content/>.

At a design level, the optimal capacity mix of power generation and storage technologies to meet system-wide electricity, ancillary service, and emission requirements was determined. The cost-optimal operation of conventional thermal, abated thermal, intermittent renewable, and energy storage units was evaluated on a coupled hourly basis. The operation of the power plants was represented in three discrete modes, “off”, “start-up”, and “on”. When operating in the latter two modes continuous ramping between a lower and upper power output range was possible. In each mode there was a minimum number of hours during which a power plant had to remain in that mode. This was called the “stay time” and is a key characteristic of a power plant’s flexibility along with the minimum stable generation point. Energy storage was represented by the charging and discharging behaviour of pumped-hydro units (PHydro, each 300 MW, 8 h storage duration) and lead-acid units (GenSto, each 50 MW, 4 h storage duration). The international interconnection capacity of the UK power system was modelled in part as large storage capacity (InterSto, 1 GW in 2015), and in part as electricity import (InterImp, 3 GW in 2015). The hourly availability profile of solar, onshore, and offshore wind power plants was accounted for using underlying data from the literature (Pfenninger and Staffell, 2016; Staffell and Pfenninger, 2016).

The model objective was the minimisation of the total cost of the system. The total system cost (TSC) accounted for annualised investment costs (CAPEX) including interest of 7.5% over the construction period of plants. The construction period was specific to each technology. Operational expenses (OPEX) were also accounted for and these consisted of start-up cost, fixed (“no load”) cost, fuel cost, variable cost and cost of CO₂ per emitted tonne, including transport and storage as they occurred in the hourly operational schedule for each power unit.

Table 1

Input data for different CLC variants for the electricity system model; ^a overnight CAPEX, interest during construction is added subsequently at 7.5% over a 5 year construction period ^b fixed operating cost, ^c fuel, carbon tax, and carbon transport and storage cost, ^d switching time from “off” to “start-up” mode, “start-up” to “on”, “on” to “off” in hours, ^e inertia is defined as the amount of kinetic energy stored in rotating mass, e.g., in motors and generators.

Parameter	Unit	NGCC-CLC (Consonni et al., 2006; Porrazzo et al., 2016)	IGCC-CLC (Cormos, 2015)	iG-CLC (DOE/NETL, 2014)
Unit size	MW	500	500	500
Efficiency	%HHV	0.468	0.342	0.351
CAPEX ^a	£(2016)/kW	1066	2285	1962
fixed OPEX ^b	£(2016)/MWh	2.46	20.73	21.2
OPEX ^c	£(2016)/MWh or h	37.8	23.9	23.6
Lifetime	years	30	25	30
Carbon capture	%	100	99.5	95.8
Annual availability	%	90	85	85
Minimum stable generation	%	40	70	45
Stay time (off, start-up, on) ^d	h	3,1,1	4,3,2	4,2,2
Potential to provide inertia ^e service	MWs	10	10	10
Potential to provide reserve capacity	%	100	100	100

Application of the ESO model allowed the value of a technology on the level of an electricity system to be determined, while considering detailed operational performance parameters. The value of a technology to the electricity system was defined as the percentage reduction in TSC caused by the deployment of the technology. The value of a technology is not a constant value, but a function of the capacity installation level and the constraints of the electricity system it is operating within.

The three main variants of CLC were considered: CLC with natural gas coupled to a combined cycle (NGCC-CLC), integrated gasification of solid fuel combined cycle with CLC (IGCC-CLC) and CLC with *in situ* gasification and combustion of solid fuel (*iG*-CLC). For the latter two processes, coal was the fuel.

The base-case parameters used in the model for the base cases are given in Table 1. The unit size was assumed to be 500 MW for all. The efficiency, CAPEX, OPEX, lifetime, carbon capture rate, and annual availability are based on the literature (Consonni et al., 2006; Cormos, 2015; DOE/NETL, 2014; Porrazzo et al., 2016). Minimum stable generation and stay times for NGCC-CLC, IGCC-CLC and *iG*-CLC were based on the literature, where it was available (Ekström et al., 2009; Naqvi et al., 2007) and otherwise were assumed to be similar to NGCC, IGCC and pulverised coal with post-combustion capture respectively. The potential to provide inertia was assumed to be 10 MWs and the potential to provide reserve capacity was assumed to be 100%. The input parameters on future cost and performance are subject to uncertainty. A detailed sensitivity analysis of the CAPEX, OPEX and efficiency parameters of the CLC variants aims to alleviate these shortcomings. They are varied within physically feasible regions and based on relevant understanding and modelling from the process and sub-process scales. The electricity system was based on that of the United Kingdom. The parameters used, corresponding to scenarios for 2030 and 2050, are given in Table 2 (National Grid, 2014; Ofgem, 2015; Powernet, 2014; Royal Academy of Engineering, 2013; UK Department of Energy and Climate Change, 2015). While the numerical results are therefore scenario specific, we believe that the qualitative conclusions are both plausible and insightful.

Slight variations on these three CLC processes included in the ESO model, such as CLOU or the use of fixed beds systems, were concluded to be insufficiently distinct in terms of the parameters of the model for it to be worthwhile to include them separately. A CLOU power plant is likely to be very similar to an *iG*-CLC facility (Spinelli et al., 2016) and fixed bed systems are generally reported to be similar to fluidised bed systems (Hamers et al., 2014). The only exception may be the CAPEX for which there is very little information, but this factor was already varied in the sensitivity analysis.

4. Results

The CLC variants (NGCC-CLC, IGCC-CLC and *iG*-CLC) were analysed in the ESO model for the UK in the 2030 and 2050 scenarios, both as single technologies and with all being available and competing with each other. The availability of CLC capacity was gradually increased whilst the upper bound of capacity for the remaining technologies was capped, at the capacity mix when no CLC was deployed. As the deployment of NGCC-CLC was increased, both the TSC and the total capacity decreased. The optimal mix of technologies also evolved. This can be seen in Fig. 6. The point at which the addition of capacity of a technology gave no further reduction in TSC, was called the 'maximum level of economic deployment', a useful way to benchmark technologies. For NGCC-CLC, it was 35.5 GW in 2030 and 40.0 GW in 2050. The corresponding reductions in TSC were 59.8% and 59.5%, respectively, compared to the base case with no NGCC-CLC.

The capacity factor of a power plant is the average power generated compared to the rated power. For NGCC-CLC, the maximum is ~ 0.9 . In the results it was lower, since the model did not impose base-load operation on any technologies. This is high compared to the capacity

factors of intermittent renewables, since those technologies generate power intermittently in time and location. In the 2030 and 2050 scenarios described here, the average capacity factors of onshore and offshore wind were 0.29 and 0.34 respectively. The addition of NGCC-CLC capacity up to the maximum level of economic deployment decreased the total capacity installed in the electricity system significantly, as shown in Fig. 6. This is because deploying CLC plants significantly reduced the requirement for balancing and back-up capacity.

Similar trends in capacity displacements occurred when IGCC-CLC or *iG*-CLC were deployed, as seen in Figs. 7 and 8, although the reductions in TSC were less pronounced than with NGCC-CLC. While the maximum levels of economic deployment of IGCC-CLC and *iG*-CLC were lower in 2030 compared to NGCC-CLC, they exceeded 40 GW in 2050. The lower maximum capacity factor of 0.85 and lower operational flexibility of IGCC-CLC and *iG*-CLC meant that their optimal deployment levels were higher to satisfy operational and emission requirements.

Regardless of which CLC variants were integrated in the 2030 scenarios, thermal power plants with post combustion capture by amine scrubbing did not play a significant role. In other words, the availability of CLC effectively displaced this competitor technology, with the zero residual emissions of CLC found to be an important source of value. In 2050, however, due to more ambitious emission targets, CCGT plants with post combustion capture did play a part in the least-cost capacity mix under the given input assumptions. Coal-fired power plants with conventional post combustion capture were installed at low deployment levels of CLC capacity, but were seen to become uneconomical when 30 GW of any CLC technology was available. Conventional CCGT and open cycle gas turbine (OCGT) plants were always part of the mix.

Fig. 9 summarises the reductions in TSC as the capacity of each CLC variant was increased up to the maximum level of economic deployment. NGCC-CLC was the most favourable variant, with *iG*-CLC marginally more favourable than IGCC-CLC. Conventional NGCC with post-combustion capture by amine scrubbing is also shown for comparison. The reduction in TSC was much lower.

To investigate the relative value of the different CLC variants further, the optimal capacity mix when all variants were available at increasing levels of deployment in 2030 and 2050 scenarios was determined. This is shown in Fig. 10. When each CLC technology was constrained to a deployment of 10 GW, NGCC-CLC, IGCC-CLC and *iG*-CLC were all deployed at this maximum level in the 2030 and the 2050 scenarios. When the constraint was increased to 30 GW for each CLC technology, only NGCC-CLC was deployed in the 2030 scenario. This

Table 2

Input parameters for the UK electricity system in the 2030 and 2050 scenarios. Values are from UK Department of Energy and Climate Change (2015), National Grid (2014), the Royal Academy of Engineering (2013), Ofgem (2015) and Powernet (2014); ^a value of UK Department of Energy and Climate Change (2015) estimate from 2035, equivalent to an 88% reduction to 2014 levels), ^b to reduce sources of uncertainty an estimate in price increase for imported electricity is not included, ^c minimum additional de-rated capacity above peak demand necessary to maintain power grid stability (UK Department of Energy and Climate Change, 2015), ^d minimum additional reserve capacity from dispatchable power generation or storage technologies to balance intermittent operation from wind and solar, ^e Value taken from National Grid (2014).

Parameter	Unit	2030	2050
Annual demand	TWh	337	377
Emission target	Mt _{CO2}	26.64	16.6 ^a
Carbon tax	£/t _{CO2}	76.2	100
Avg. elec. import price	£/MWh	26.9	26.9 ^b
Capacity margin ^c	%-MW	4	4
Wind reserve margin ^d	%-MW	15	15
Avg. transmission losses	%-MWh	0.077	0.077
System inertia threshold ^e	GW.s	100	100

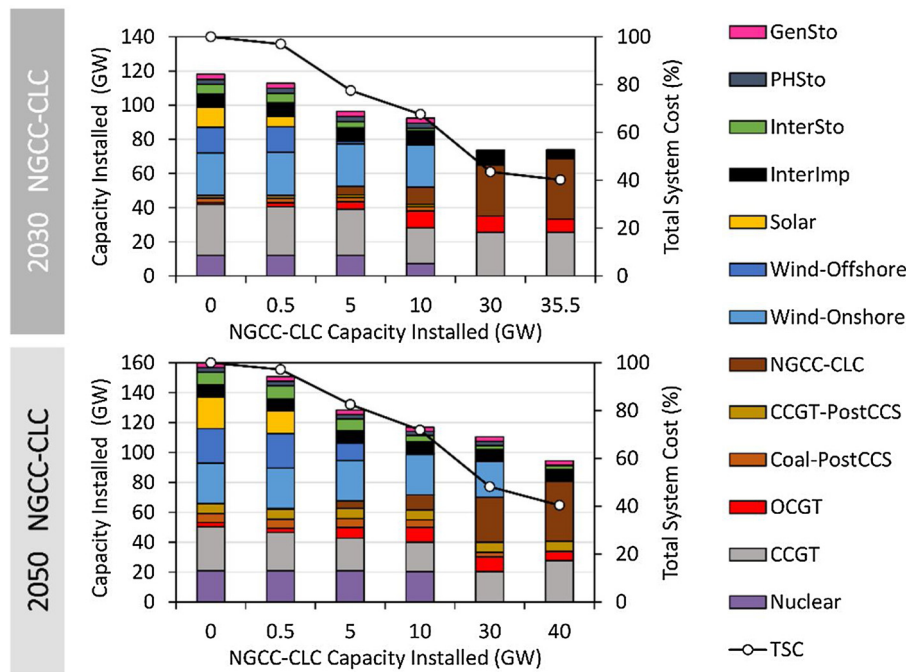


Fig. 6. Installed capacity and total system cost when NGCC-CLC was available in 2030 and 2050.

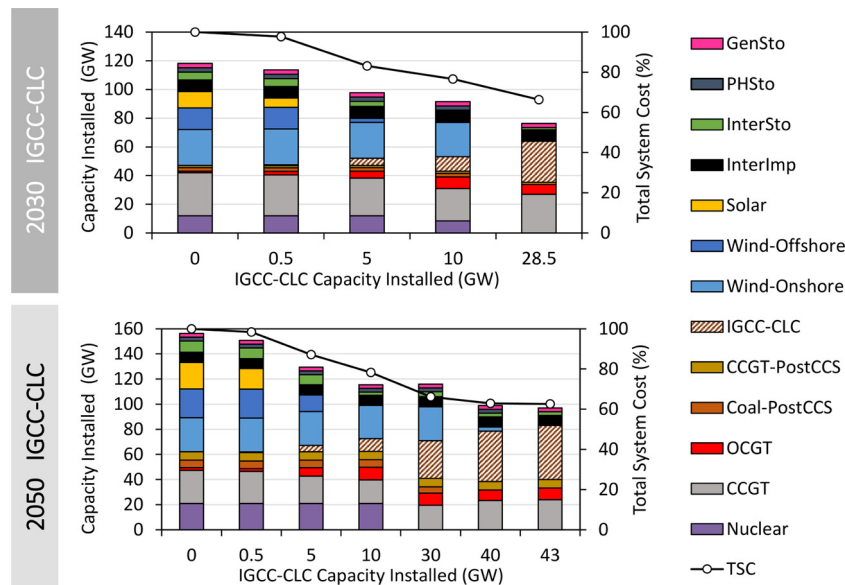


Fig. 7. Installed capacity and total system cost when IGCC-CLC was available in 2030 and 2050.

was supplemented with 10 GW of iG-CLC in the 2050 scenario. These trends are in agreement with those presented in Fig. 9, where NGCC-CLC was found to be the most valuable technology, followed by iG-CLC.

Fig. 10(a) shows the share of electricity by generation source when 5 GW of NGCC-CLC capacity was available over a period of two, non-consecutive, sample days in 2030. Fig. 10(b) shows the reserve provided by source over the same time period. Reserve is the additional capacity held by generators or energy storage technologies which does not serve the instantaneous electricity demand but can be called upon if necessary. Over the two sample days shown, the available level exceeded the minimum reserve requirement during all time periods. Reserve requirements increased with the level of intermittent power generation (hours 32–44). When battery storage was utilised (e.g. hours 25 and 42) the reserve excess became less pronounced. NGCC-CLC provided firm reserve capacity and with an annual average of 17% in

2030 and 11% in 2050 of the total reserve provision, it provided the third largest reserve component, after storage technologies and conventional CCGT, at this level of deployment (5 GW).

CLC power plants also provided essential ancillary services in these scenarios such as frequency control to the power system, in a similar manner to conventional thermal power plants.

5. Retrosynthetic analysis

The impact at the system level of key parameters associated with CLC technologies and their design was investigated. The focus was on the impact of capital cost, oxygen carrier cost and thermal efficiency. This analysis enables the current and potential future role and value of CLC to be identified, and how the parameters could be optimised. This is discussed in Section 6 and the key research directions and design

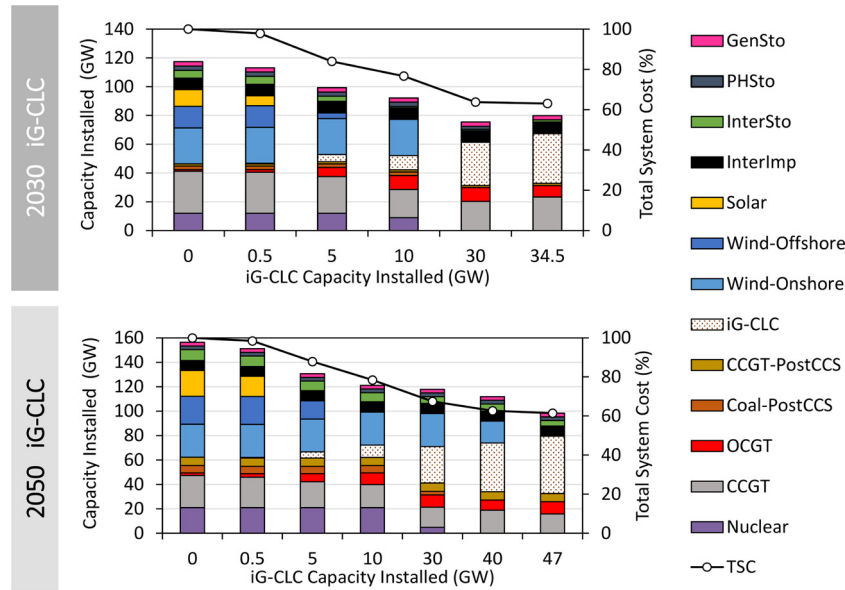


Fig. 8. Installed capacity and total system cost when iG-CLC was available in 2030 and 2050.

modifications are discussed in Section 7 (Fig. 11).

5.1. Sensitivity to capital cost

There is uncertainty in the costs associated with CLC, in particular the estimates for capital expenditure (CAPEX), because no full-scale plants have yet been built. Fig. 12 shows the maximum level of economic deployment of CLC in 2030 as a function of CAPEX. For all CLC technologies, the optimal level of economic deployment followed a similar, non-linear trend as the CAPEX was increased. For NGCC-CLC, the optimal level of economic deployment dropped gradually to 30 GW as the CAPEX was increased to £2000/kW since its competitiveness with conventional thermal power plants with post combustion capture diminished. As the CAPEX was increased further, and the thresholds corresponding to the CAPEX of onshore wind (light grey) and of nuclear capacity (dark grey bars) were passed, the optimal level of NGCC-CLC deployment experienced discrete drops in value. For onshore wind, the CAPEX shown is availability-scaled to account for the significant backup capacity which must also be installed since the capacity factor of technologies that generate electricity intermittently is low. IGCC-CLC and iG-CLC technologies followed a similar trend. The fact that all CLC technologies remained part of the optimal capacity mix in 2030 at high CAPEX, underlines their value as low-carbon and dispatchable power generators. The results at high CAPEX were strongly influenced by the fixed upper bound of deployment that was set for conventional technologies in the mix, *i.e.* considering reasonable capacity build-rates, it

was assumed that the deployment of each technology could not exceed certain capacities in line with estimates by DECC (UK Department of Energy and Climate Change, 2015).

5.2. Sensitivity to oxygen carrier cost

One factor that could affect the operating expenditure (OPEX) of CLC plants is the cost of the OC material. For NGCC-CLC and IGCC-CLC plants, the contribution of the OC cost to the OPEX appears to be low, provided that it has a reasonable lifetime (Porrizzo et al., 2016). On the other hand, in the iG-CLC plant, reported by the National Energy Technology Laboratory (NETL) (DOE/NETL, 2014) and used in this study, the Fe-based OC which was used accounted for 73% of the variable OPEX, excluding fuel. The sensitivity of the value of iG-CLC in an electricity system to a 75% and 50% decrease and 50% increase in the variable OPEX was investigated. This corresponded, respectively, to 29% and 19% reductions and a 21% increase in the overall OPEX, which included fuel, carbon tax, transport and storage. The optimal mix of capacity remained constant across the range of OPEX investigated. Operational performance parameters were marginally affected by the cost of the carrier. As the OPEX increased, CLC utilisation decreased, while the utilisation of other power plants increased to achieve the lowest TSC. Fig. 13 visualises these performance dependences, when 2.5 GW of iG-CLC was deployed in a 2030 scenario.

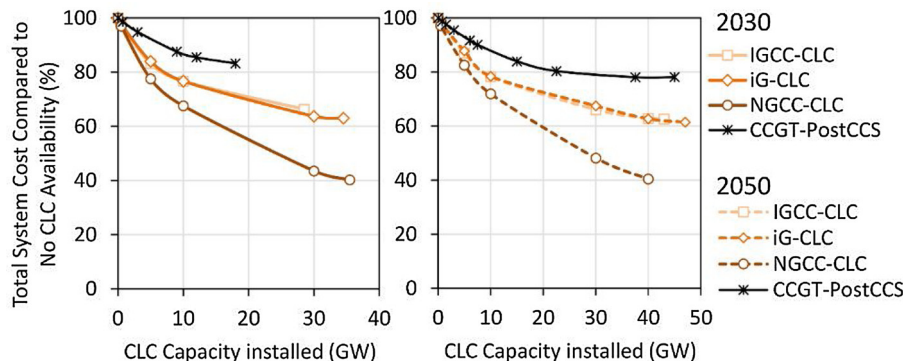


Fig. 9. Value of CLC technologies for reducing total system cost at different levels of deployment in 2030 and 2050.

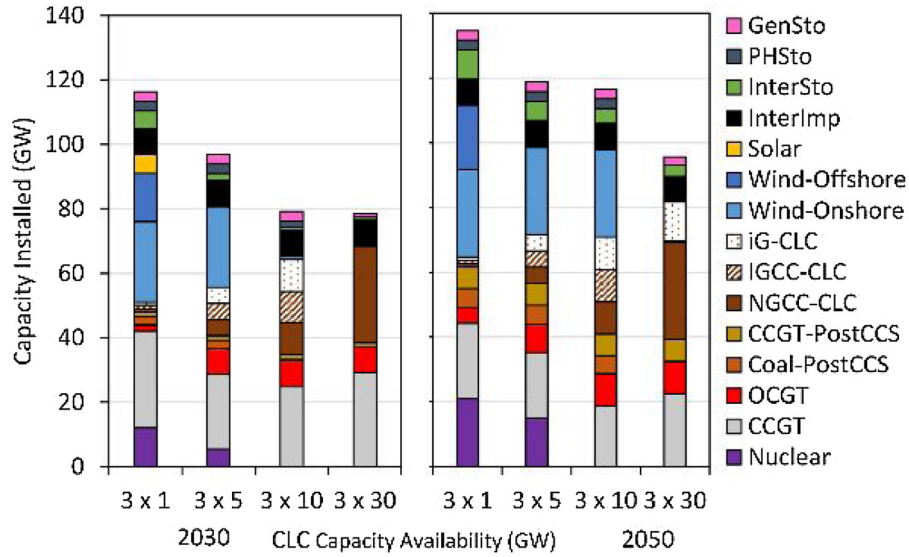


Fig. 10. Installed capacity with all three CLC technologies available in 2030 and 2050. The x axis gives the maximum capacity that can be deployed for each technology.

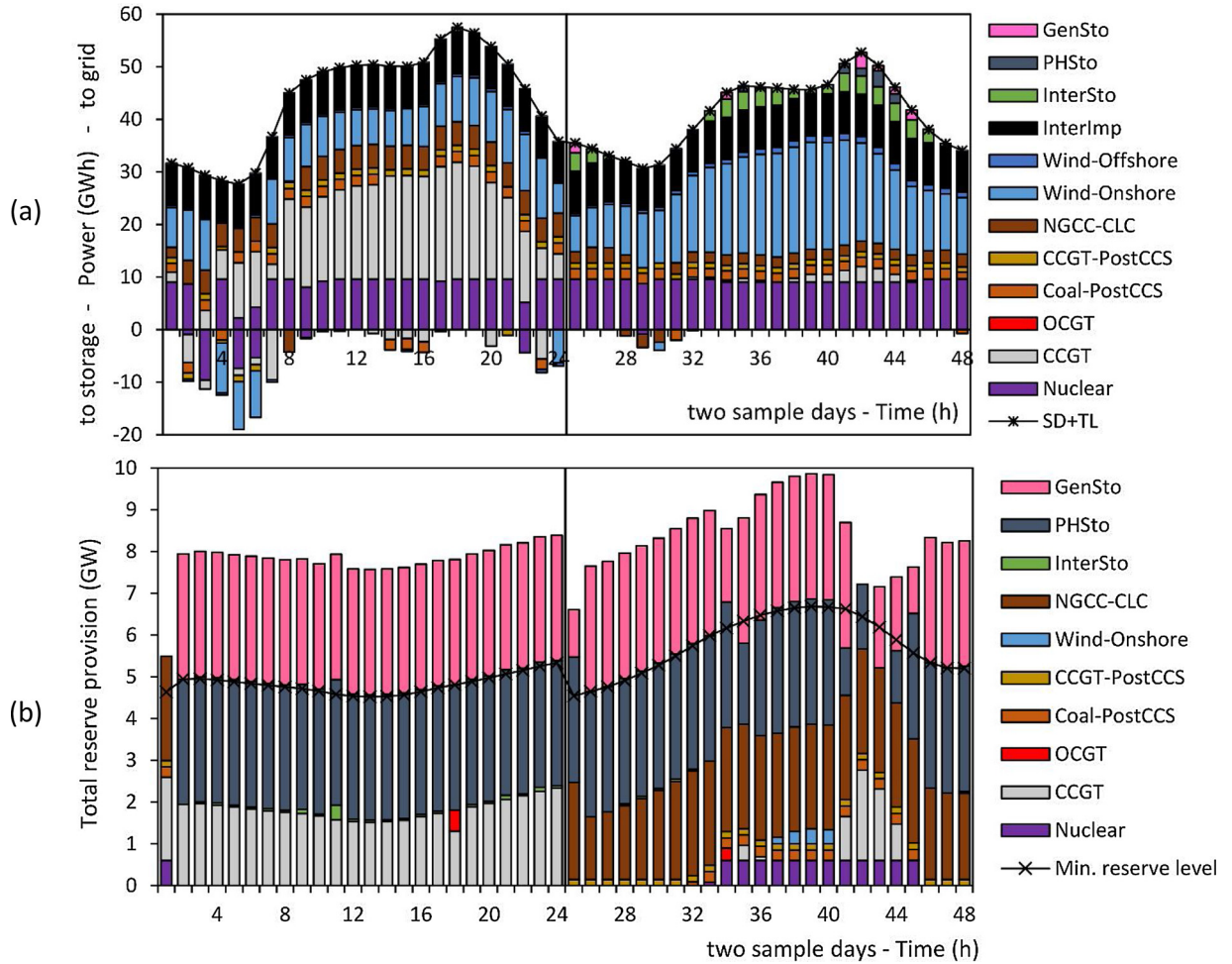


Fig. 11. (a) Hourly power generation and storage operation and (b) reserve provision during two sample days in 2030 when NGCC-CLC was deployed at 5 GW capacity level. Total power provision had to meet grid-level electricity demand (SD) while accounting for transmission losses (TL).

5.3. Sensitivity to efficiency

There is a broad consensus in the literature regarding the efficiencies of the different CLC variants included in the ESO model. The

sensitivity of the electricity system and individual power plants to the efficiency of NGCC-CLC plants was nevertheless investigated since there is scope for these to improve in future. An increase or decrease in efficiency of NGCC-CLC power plants by 10%, corresponding to a range

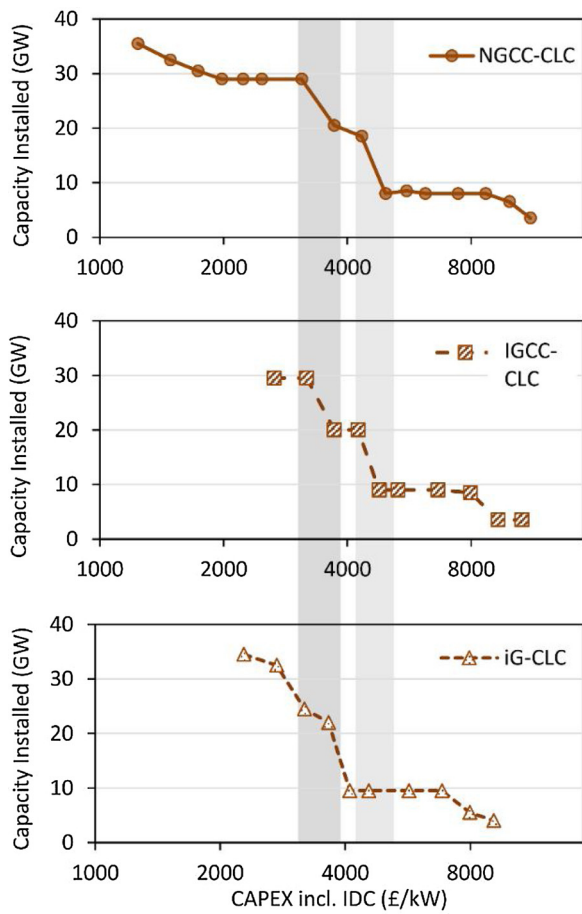


Fig. 12. Optimal NGCC-CLC capacity deployment in 2030 as a function of the CAPEX including interest during construction (IDC) of 7.5% over a 5 year construction period. The leftmost point in each case indicates the maximum level of economic deployment with the base case CAPEX. The grey shaded areas indicate the CAPEX of availability-scaled (nominal CAPEX/Capacity Factor *i.e.* £1150/kW/0.3) onshore wind (first, dark), and the CAPEX of nuclear capacity (second, light).

of 42.2–51.5% (HHV) did not affect the optimal capacity mix, when 10 GW was deployed in a 2050 scenario. The competitiveness and the dispatch merit order were affected and in turn influenced the system-

wide costs and the hourly operation. A reduction in efficiency of NGCC-CLC power plants caused their average annual utilisation to drop from 78.5% to 77.5%. Conversely, the annual average utilisation of CCGT plants with post combustion capture (CCGT-CCS) increased from 68% to 69%. Fig. 14 shows the effect of a 10% increase and decrease in the efficiency of NGCC-CLC on the annual running and start-up OPEX of different power plants. The technologies where the costs were affected the most are shown in terms of percentage changes from the base case in Fig. 6. In the base case, the OPEX of NGCC-CLC was lower than for CCGT-CCS, so NGCC-CLC plants ran as base load, resulting in no cost associated with start-up. CCGT-CCS plants and especially open cycle gas turbine (OCGT) units were forced to switch on and off and to ramp their power output to satisfy demand during peak hours.

A reduction in efficiency of NGCC-CLC plants by 10% (orange bars) increased the NGCC-CLC fuel cost such that CCGT-CCS and NGCC-CLC both operated with roughly the same running costs. On an annual basis, this reduced NGCC-CLC utilisation by 1%, from 78.5% to 77.5%. To compensate, CCGT and CCGT-CCS power plants increased their power output, and their annual running costs increased as shown in Fig. 14. OCGT power plants were more heavily used at peak times, while the frequency of start-ups and shutdowns of CCGT-CCS units decreased.

With an increase in efficiency of 10% (green bars), NGCC-CLC power plants were only marginally below conventional unabated CCGT power plants, modelled with an efficiency of 52.7%. This decreased the running cost associated with fuel consumption. The utilisation of NGCC-CLC power plants increased only marginally (0.1%). Since base load operation had already been achieved with the original efficiency, there was little scope to increase utilisation further. The frequency of start-up and shutdown of OCGT power plants decreased. Accordingly, the annual OCGT start-up cost decreased while the running costs increased.

The TSC was virtually unaffected by changes in the efficiency of NGCC-CLC plants. A 10% increase in efficiency to 51.5% decreased the TSC by only 0.2%, while a 10% decrease increased the TSC by 0.5%. The internal rate of return (IRR) of a NGCC-CLC power plant with the base case efficiency (46.8%) was 5.4%. The IRR was determined with a discount rate of 5%, a project lifetime of 50 years, construction time of 5 years. An average electricity price of £80/MWh in the 2050 UK power system (Mac Dowell and Staffell, 2016) The correlation between CAPEX and efficiency was modelled corresponding to pulverised unabated coal-fired power plants (International Energy Agency, 2016), such that the NGCC-CLC CAPEX for 42.2% efficiency was determined to be £681.2/kW, and £1450.8/kW for 51.5%. A power plant with an efficiency 10% lower than the base case, but with reduced CAPEX,

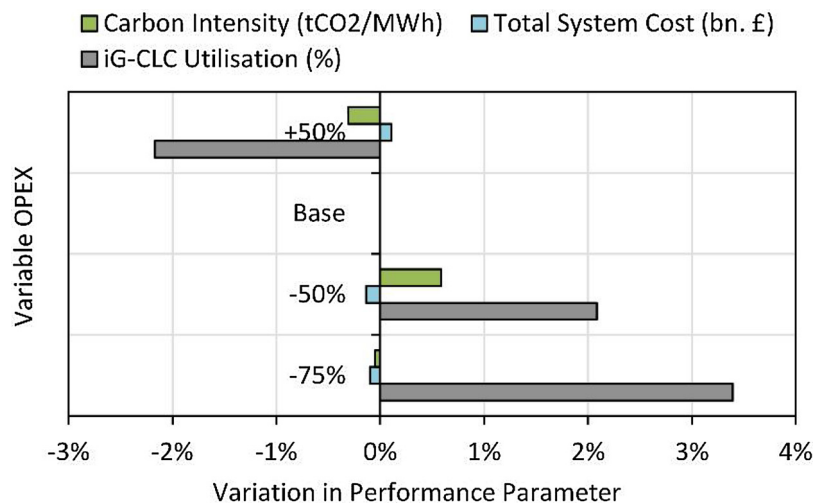


Fig. 13. The sensitivity of different performance parameters to a change in the variable OPEX. For the case of 2.5 GW iG-CLC in the 2030 electricity system, the variable OPEX was dominated by the cost of the oxygen carrier.

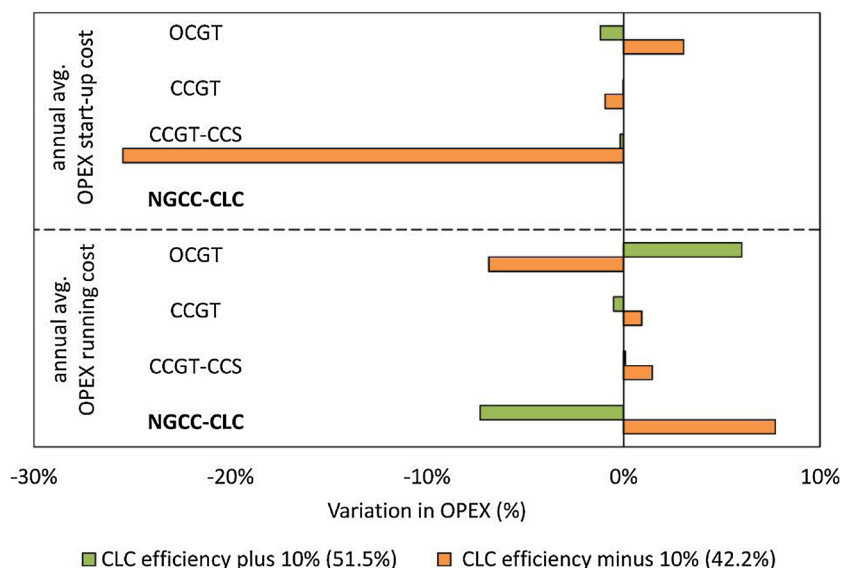


Fig. 14. The sensitivity of annual average OPEX running cost, £/MWh (including fuel cost, variable, fixed cost, carbon tax, CO₂ transport and storage cost where applicable) and OPEX start-up cost, £/MW of OCGT, CCGT, CCGT-CCS, and NGCC-CLC power plants to the efficiency of NGCC-CLC power plants at an installation level of 10 GW in 2050. The x axis shows the change in the annual avg. OPEX associated with start-up and OPEX associated with running (i.e., combination of OPEX and fixed OPEX from Table 1) in percent for different technologies when the NGCC-CLC efficiency was varied by $\pm 10\%$ (i.e., 51.5%, 42.2%, compared to 46.8% base case).

sacrificed only 1% of utilisation, and gave a higher IRR of 6.8%. A NGCC-CLC power plant with a 10% higher efficiency, gave a marginal increase in utilisation, but due to the greater CAPEX, it resulted in a lower IRR of 4.3%.

6. Discussion

All three CLC variants (NGCC-CLC, IGCC-CLC and iG-CLC) could be valuable in a future electricity system and further investment would therefore be worthwhile. The maximum levels of economic deployment and the associated reductions in TSC, of all the variants, are significantly higher than for NGCC with post-combustion capture, the current benchmark. While such high deployments may be difficult to envisage, regardless of the level at which CLC technologies are deployed, they lead to reductions in total installed capacity and, more importantly, in the TSC. This is because they are able to substitute conventional unabated and abated thermal, nuclear, wind and solar generating capacity economically. The value of intermittent renewables, especially at higher levels of deployment, was significantly lowered by the availability of CLC since it is dispatchable as well as being low carbon. This would be one likely feature of having energy policies that do not favour any particular technology, but rather are directed at satisfying targets at the system level e.g. carbon constraints and least cost. This policy framework was an assumption of the ESO model that was used in this work.

Amongst the various CLC technologies, NGCC-CLC would be the most favourable, followed by iG-CLC and then by IGCC-CLC. NGCC-CLC was the most attractive CLC technology in the scenarios since the lower capital expenditure (CAPEX) and higher efficiency outweighs the greater cost of gaseous fuel compared to solid fuel. In addition it is likely to have good load-following characteristics. The iG-CLC and IGCC-CLC variants have very similar potential to reduce TSC. The former has slightly higher efficiency and lower CAPEX than the latter. OPEX and operational flexibility are very similar. Owing to uncertainty in the CAPEX and OPEX, the relative value of iG-CLC and IGCC-CLC could change. Across the world, the cost of gas and coal varies. This could impact the relative value of all the different CLC variants.

In terms of scale-up, a significant variation amongst NGCC-CLC, IGCC-CLC and iG-CLC is that it has been assumed that the reactors in the first two variants would operate at elevated pressure to recover energy in a topping cycle. Systems operating at atmospheric pressure are, certainly initially, likely to be similar in design to circulating fluidised bed combustors (CFBCs) (Lyngfelt and Leckner, 2015). Circulating fluidised bed combustors are widely used for power generation

across the world, with the largest plant rated at 460 MWe (Hotta, 2009; Nowak and Mirek, 2013). Operation at pressure has additional challenges and so the technological risk is higher, for example because particle separation prior to the gas turbine becomes essential. There is some experience of performing this and also of constructing turbines that can withstand some level of particulates, since this was necessary to enable the construction and operation of demonstration and commercial pressurised fluidised bed combustors (Asai et al., 2004; Japan Coal Center, 2007; Komatsu et al., 2001; Shimizu, 2013). Maintaining stable circulation between pressurised reactors is another challenge, that has been achieved at the pilot scale (Xiao et al., 2012), but has yet to be demonstrated at full scale. Dynamically operated packed bed reactors would overcome the circulation problem, but instead temperature control and valve switching at high temperature and pressure would be the technological risk. The choice depends on the relative cost, operability and availability of the two systems (Hamers et al., 2014), which is at present not certain.

For reasons of technological risk, an iG-CLC plant is likely to be the most attractive as a first full-scale demonstration of CLC technology. The fact that NGCC-CLC was most attractive in terms of value, would indicate that, for that technology, taking the step from operating at atmospheric pressure to an elevated pressure of up to 10 bar is likely to be worthwhile. The value of IGCC-CLC was similar to iG-CLC, but it is associated with greater technological risk. It is therefore unlikely to be attractive for implementation.

At low deployment levels (< 10 GW), the ability of CLC power plants to ramp their output made them attractive in the scenarios, since it allowed more intermittent generators to be integrated. In this respect, NGCC-CLC power plants outperformed IGCC-CLC and iG-CLC since they have greater turn-down ratios and shorter mode switching times. For example at 5 GW of CLC deployment in the 2030 scenario, the operational ability of NGCC-CLC plants to turn down their power output resulted on average in a higher annual utilisation of onshore wind of 29% compared to 27.5% in the case of IGCC-CLC integration. The utilisation rate of NGCC-CLC plants was 76% while that of IGCC-CLC plants was 78.7%. The overall cost of the system was therefore reduced as a larger share of wind power generation reduced overall operational cost. By 'making room' for renewable power generators, which are typically intermittent, NGCC-CLC power plants utilised the grid-level energy storage systems more heavily, contributing 20% to annual charging to storage technologies. In comparison, at 5 GW deployment level, IGCC-CLC plants contributed 8% and iG-CLC plants 10%.

At high deployment levels (> 10 GW), the annual availability of dispatchable and low-carbon power generation, alongside operational

cost and efficiency are crucial to the value of a technology. Among the technologies included in the scenarios, CLC plants were unique, along with conventional CCS options, in being able to provide this. NGCC-CLC outperformed IGCC-CLC and iG-CLC in these aspects. In the 2030 scenario, battery and hydro energy storage units (GenSto, PHSto) were entirely displaced at high deployment levels of NGCC-CLC, because of the ability of NGCC-CLC to load-follow. This also reduced the operational cost of the power plant because it reduced the frequency of shutdown. IGCC-CLC and iG-CLC plants are less flexible and so had to shut down and start up frequently. Regardless of the level at which CLC might be deployed, a detailed understanding of the load-varying behaviour and how it could be improved is very important.

The fact that CLC technologies are dispatchable means that they would be well-suited to providing reserve capacity (Fig. 10(b)). They could also provide ancillary services, e.g., inertia which are important for frequency control. Intermittent sources, e.g., wind, solar are unable to provide this and their increased deployment, combined with the decommissioning of many unabated thermal plants, which are inertia-rich, could jeopardise the operability of electricity systems (National Grid, 2014).

6.1. Sensitivities

The CAPEX of the different CLC plants are, at present, uncertain so the sensitivity to this parameter is important. Two threshold values were identified; the availability-scaled CAPEX of onshore wind (~£3000/kW) and the CAPEX of nuclear capacity (~£4400/kW). At these values there were discrete drops in the maximum economic level of deployment. For CLC to be widely deployed in the electricity system of the UK, the CAPEX would need to be at least comparable to the availability-scaled CAPEX of onshore wind, and preferably lower. Globally, this could be generalised to the availability-scaled CAPEX of the most economical renewable, which in some energy systems could instead be solar e.g. in areas of the USA or China.

The most significant contribution to OPEX was the cost of fuel, in particular for NGCC-CLC, owing to the higher cost of natural gas compared to coal. For NGCC-CLC and IGCC-CLC, the cost of the OC material has a small effect on the OPEX, because its lifetime is long as a result of only coming into contact with clean natural gas (NGCC-CLC) or syngas (IGCC-CLC). For the case of a synthetic Ni-based carrier in NGCC-CLC, Porrazzo et al. (2016) found, that lifetimes greater than 1000 h, resulted in the carrier having a small impact on costs. A synthetic Ni-based carrier at \$15,300/t (2015 prices) was used, since natural mineral ores are typically unsuitable with methane because the reduction kinetics are slow and combustion may not be complete (Leion et al., 2008). In the literature, a number of studies have estimated lifetimes for synthetic OC particles to be significantly greater than 1000 h (Hallberg et al., 2016; Linderholm et al., 2008; Lyngfelt and Thunman, 2005). The reactivity of ores with CO and H₂, the constituents of syngas, is often similar to that of synthetic carriers. A natural ilmenite ore was therefore used in the IGCC-CLC variant included in this study (Cormos, 2015). The shorter lifetime that would be expected due to the lower attrition resistance of ores compared to synthetic particles is more than compensated for by the significantly lower cost at €80/t (2015 prices).

OC lifetimes in an iG-CLC application are likely to be shorter in NGCC-CLC or IGCC-CLC because the carrier will be lost due to the need to separate the solid fuel and the OC (Jerndal et al., 2011; Lyngfelt, 2014). It is not surprising therefore that the cost of the Fe-based OC (2000 \$/t) used in the NETL study (DOE/NETL, 2014) was a significant proportion of the variable OPEX (73%). A significantly cheaper natural ore could be used instead, since the main product of gasification of solid fuel is syngas rather than methane. Although a reduction in cost would be possible, it did not have a significant impact on the maximum economic level of deployment of iG-CLC and only increased the utilisation of the technology slightly.

The sensitivity to the cost of the OC in all the CLC variants was therefore found to be small, provided reasonable lifetimes can be achieved. Ongoing work should not be focussed on reducing cost, but rather on increasing reactivity and metal loading, which could reduce the size of reactor need to accommodate the inventory of OC and therefore the CAPEX of the CLC system.

6.2. Future prospects

There is scope for technological improvement to the CLC variants included in this paper. For example, for gaseous fuels, CLC could be coupled to a more advanced power cycle, leading to higher efficiency (Brandvoll and Bolland, 2004; Petriz-Prieto et al., 2016). The efficiency could also be improved by increasing the TIT above 1200 °C through optimising the high temperature performance of the OC particles. Improving the efficiency would have an almost negligible effect on the utilisation of NGCC-CLC plants and reduce the total system cost only marginally. These plants would however be associated with higher CAPEX and so on balance the IRR would probably be lower, than for plants with a lower efficiency. Therefore given the opportunity to reduce CAPEX or increase efficiency, the former would be more attractive. In an electricity system characterised by high penetration of renewables, efficiency gains are outweighed by reduced capacity factor, and thus reducing capital cost is more important.

There is also the potential to reduce the size of the CLC section in a power plant. As mentioned in the previous section, one way to achieve this would be to increase the loading and reaction kinetics of the OC material. It has also been suggested that tuning the residence time distribution of the OC particles in the reactors would allow the inventory and circulation rate of oxygen carrier to be decreased (Schnellmann et al., 2018). This would reduce both the CAPEX and the OPEX. Especially the former could increase the attractiveness of CLC technologies. Porrazzo et al. (2016) estimated that the cost of the reactors accounts for 64% of the cost of an NGCC-CLC plant. For the IGCC-CLC case it was 10% (Cormos, 2015) and for the iG-CLC case it was 30.6% (DOE/NETL, 2014).

As for other thermal power plants with carbon capture, it would be possible to reduce fossil fuel use and achieve negative CO₂ emissions with CLC if biomass were used as fuel (Bio-energy with CCS or BECCS). In fact, it is becoming increasingly apparent that BECCS could play a vital role in achieving the necessary cuts in carbon dioxide emissions to limit warming of the planet to 2 °C (Fuss et al., 2014; Victor et al., 2014). A variety of biomass fuels such as sawdust and wood char have been used in continuous CLC units up to 100 kWth (Gu et al., 2011; Knutsson and Linderholm, 2015; Mendiara et al., 2016). CLOU and iG-CLC processes have been investigated, with both natural mineral ores and synthetic particles as oxygen carrier. For implementation, adjustments to the CLC reactor designs would be necessary, optimal oxygen carriers would need to be identified and other parameters such as fuel size would require optimisation due to the different properties of biomass compared to coal. In a recent techno-economic study, CLC with biomass performed favourably when compared to other BECCS options (Bhave et al., 2014).

Other CLC processes, for example that allow the co-production of electricity and H₂ by the addition of a third reactor (Cormos, 2015), could also be of interest, particularly if a 'Hydrogen Economy' were desired. In such a design where the energy vector could be diversified to enable a plant to produce electricity flexibly, the production of hydrogen would be greatest at times when intermittent renewable sources (e.g. wind, solar) are plentiful. This is not expected to be a problem since storage of hydrogen at large-scale, for example in salt caverns is relatively well understood (Lord, 2009; Ozarslan, 2012).

7. Retrosynthetic technology design and key research directions

In this section key research directions that would make CLC more

attractive for implementation are summarised. They are based on the retrosynthetic analysis conducted in Section 5, and draw on a number of design modifications that were suggested in the discussion in Section 6.

Since CLC has been identified as attractive in future electricity systems, it is worthwhile to invest in its further development and scale-up. Apart from scale-up, since the capital cost of a CLC plant was found to be critical for its competitiveness, research should be aimed at minimising this cost. A reduction in CAPEX was found to be attractive even at the expense of a reduced efficiency. Strategies could include modifying the particles or the reactor design. In terms of reactor design, it has been shown that adjusting the residence time distribution of the particles could reduce the required inventory and circulation rate of solids by almost 20% (Schnellmann et al., 2018). Work to improve the efficiency is unlikely to have a significant effect on the attractiveness of CLC for implementation.

The cost of the oxygen carrier was found to be less important than commonly assumed, so certainly for NGCC-CLC, the use of synthetic particles should not be dismissed. In terms of the design of the particles, characteristics other than cost such as the metal loading and the reaction kinetics were found to be important since they could contribute to reductions in capital cost. Increasing the metal loading of particles would reduce the required circulation rate of solids and an improvement in their reaction kinetics would reduce the required inventory, provided that the rate of reaction is not limited by mass transfer. Further research on these aspects would therefore be valuable.

Finally, a better understanding of the load following behaviour of CLC plants is needed and to what extent designs could be adapted to improve it. The ability of power plants based on CLC technology to adjust their power output depending on demand has not seen significant attention.

8. Conclusions

A novel retrosynthetic analysis and design approach has been presented to evaluate technologies and suggest how they can be modified to optimise them based on the system in which they might operate. It involved investigating their operation in the system, within physically feasible ranges of key parameters. From this the current and potential future role and value of the technology could be determined. Key research directions and a number of design modifications could be

identified to improve its attractiveness for implementation could be identified. For this reason it was termed ‘retrosynthetic’ since it is able to guide the synthesis, *i.e.* the design and development, of a technology so that it is optimised based on the requirements and demands of the system within which it would operate. In this paper, our approach was applied to chemical looping combustion, which is a promising technology for generating low-carbon electricity.

The evaluation showed clearly that chemical looping combustion (CLC) technologies could have a role and could provide significant value to future electricity systems by reducing the total cost of the system. CLC would be more favourable than competing carbon capture technologies since it has higher efficiency and comparable or slightly lower capital and operating costs. Further investment would therefore be worthwhile. Since CLC technologies are dispatchable as well as zero-carbon they would reduce the value of intermittent generators such as wind or solar, especially if energy policies were directed at meeting targets at the system level, such as carbon constraints, rather than being directed at certain technologies.

In terms of sensitivities, an important capital expenditure threshold was identified. To be implemented widely, it would be necessary for the capital cost of CLC technologies to be at least comparable to the availability-scaled capital cost of the appropriate renewable technology in that location, *e.g.* wind, solar. The sensitivity of the role and value of CLC to the cost of the oxygen carrier material was found to be small, although other characteristics such as reactivity or metal loading could be important. Work has been carried out to date directed at improving the efficiency of CLC technologies and further improvements in future are possible. Research and development should rather prioritise the minimisation of capital cost, even if this comes at the expense of reduced efficiency. This is likely to become a feature of the development of emerging technologies due to the increasing diversification of the energy system in the 21st century.

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Appendix A

The governing constraints and the objective function of the Electricity Systems Optimisation (ESO) modelling framework, underpinning the analysis in Sections 3, 4, and 5 is summarised here. A detailed model description can be found elsewhere (Heuberger et al., 2017a,b).

The objective function is the minimisation of total annual cost of the power system (*tsc*) and is described in Eq. (4). The *tsc* is comprised of the capital expenditure for each capacity unit of technology *i*, and the operational cost. Units are modelled to operate in three different modes *m*, start-up (*su*), online with continuous power output adjustment (*inc*), and off (*off*). For the duration of operation (hours per year) *t*, of any unit in any of these modes, operational cost associated with the mode (start-up cost, fuel and maintenance cost) is incurred. There are different technology types *icI* = {*ic*, *ig*, *is*, *ir*}. The index *ic* refers to conventional dispatchable power plants, *ig* to power generation, *is* to energy storage technologies and *ir* refers to intermittent renewables such as onshore wind, offshore wind, and solar. The technology categories are not exclusive. A CCCGT-CLC power plant for example, is a dispatchable *ic* and power generation *ig* type of technology.

$$\begin{aligned} \min\{tsc\} = & \sum_{icI} CAPEX_i \quad d_i \quad Des_i + \sum_{icI, m=[su], m'=[off], t \in T} \left(\frac{OPEX_{ic, m} n_{ic, m, t}}{StayT_{ic, m', m}} \right) + \sum_{ig \in I, m \in [su, inc], t \in T} OPEX_{NL_{ig}} n_{ig, m, t} + \sum_{ig \in I, m=[inc], t \in T} OPEX_{ig, m} p_{ig, m, t} \\ & + \sum_{is \in I, m=[inc], t \in T} OPEX_{is, m} o_{is, t} \end{aligned} \quad (4)$$

A set of system-wide constraints ensures the adequacy, reliability, and operability of the capacity of the power system as well as compliance with CO₂ emission targets. Constraint 5, guarantees electricity demand *SD_t* to be met at any time *t* by power provision from generation technologies *p2d_{ig, m, t}* and power from energy storages *s2d_{is, t}*.

$$\sum_{ig \in I, m \in M} p2d_{ig, m, t} + \sum_{is \in I} s2d_{is, t} = SD_t \quad \forall t \quad (5)$$

To ensure an adequate amount of capacity available to handle sudden outages or variations in power supply from intermittent sources (*ir*),

constraint 6 requires capacity reserves ($r_{ig,m,t}$ and $s2r_{is,t}$) to be greater or equal to the system reserve margin (peak load PL and reserve margin RM) and a percentage of the intermittent power output.

$$\sum_{ig \in I, m \in M} r_{ig,m,t} + \sum_{is \in I} s2r_{is,t} \geq PL \cdot RM + \sum_{ir \in I, m \in M} p_{ir,m,t} WR \quad \forall t \quad (6)$$

Compliance with system operability requirements are provided by constraint 7, which ensures a minimal level of system inertia SI_t at all times t by operating technologies ($n_{ig,m,t}$) and their potential to provide such services ($IP_{ig,m}$).

$$\sum_{ig \in I, m \in M} n_{ig,m,t} Des_{ig} IP_{ig,m} \geq SI_t \quad \forall t \quad (7)$$

Constraint 8 limits total operational CO₂ emissions to an annual emissions target SE .

$$\sum_{i \in I, m \in M, t \in T} e_{i,m,t} \leq SE \quad (8)$$

Additional constraints describing the detailed operation of the different technology types (ic, ig, is, ir) in the different operational modes m , as well as the account for CO₂ emissions, and energy storage charging and discharging on an hourly time discretisation are vital parts of the ESO model. A detailed account of the model formulation, solution approaches, and data inputs can be found in Heuberger et al. (2017a,b) and at <https://www.imperial.ac.uk/a-z-research/clean-fossil-and-bioenergy/downloadable-content/>.

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