Absorption enhanced reforming of lignite integrated with molten carbonate fuel cell

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Abstract

A technical-economic assessment of an innovative system which integrates absorption enhanced reforming (AER) of lignite with molten carbonate fuel cell (MCFC) for electricity generation is investigated using the ECLIPSE process simulator.

The simulation results show that the proposed system of combining AER with MCFC has the electricity output of 206 kW, with the electrical efficiency of 44.7% (low heating value – LHV) and CO_2 emissions of 751 g/kW h, when fuelled with lignite. The system has a specific investment (SI) of £11 642 and a break even electricity selling price (BESP) 21 p/kW e, compared to the SI of £10 477 and the BESP of 19 p/kW e for the basic case of MCFC fuelled with natural gas.

A sensitivity analysis of the break even selling price (BESP) of electricity and the specific investment (SI) versus the capital cost show that capital costs have a significant effect on BESP and SI. Based on the basic case of capital cost of £2 398 000, when the capital cost of the system reduces 50%, the relevant BESP lowers down to 10.8 p/kW e, the SI also reduces by 50%, to £5864/kW e.

A sensitivity analysis of fuel cost versus BESP show that the fuel cost has a little effect on BESP. For the basic case of the system with the cost of lignite £20/ton, the BESP is 21.1 p/kW e. While the fuel cost reduces by 50%, to £10/ton, the BESP lower down to 20.9 p/kW e, only reduces 0.2 p/kW e, the change is 0.9%.

Although the BESP and SI are high for the AER + MCFC system, there are no nitrogen oxides (NO_x) and sulphur oxides (SO_x) emissions from the system; the CO_2 gas stream produced in the AER process is suitable for subsequent sequestration. Thus the combination system may become a power generation with zero greenhouse gas emissions.
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1. Introduction

The emission of greenhouse gases from the human activities in energy sector and the following effect on the global climate change has caused great concern all over the world. The combustion of fossil fuel in power plants is responsible for the majority of greenhouse gas emissions. Hence it is always the main focus for researchers and engineers to reduce the emission of greenhouse gas worldwide.

Coal is the largest fuel resource to meet the energy requirement in the world [1,2]. It is abundant, safe, secure and affordable; easy to transport and store; and widely available [3–5]. Lignite (brown coal) is a kind of low rank coal. The recoverable reserves of lignite are 157 967 million tons in the world, which is 17.4% of the total reserves of coal in the world in 2003. In the same year, the production of lignite is 886.4 million tons, which is 18.0% of the total world coal production in the year. The outputs of lignite are mostly consumed for the electricity production, especially in European countries [6].

Coal electrical power generation is a well-established and highly reliable technology. Coal produces 39% of the

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world's electricity [2,3], which is twice as much as the next largest source. Coal use in power generation is projected to grow 60% by 2030 [2]. Statistic data show that coal is still the most important energy resources in the world today and in the future.

But as a fuel, coal has its own disadvantages because it is the most carbon intensive fuel for electricity generation. The combustion of coal generates not only carbon dioxide (CO_2), but also the other harmful gases such as carbon monoxide (CO), nitrogen oxides (NO_x) and sulphur oxides (SO_x). This poses challenges for the researchers and engineers to find out new coal-based generation technologies.

Fuel cell generates electricity and heat by electrochemically combining a gaseous fuel (hydrogen) and an oxidant gas (oxygen from the air) through electrodes and across an ion conducting electrolyte [7]. During this process, water is formed at the exhaust. The main characteristic of a fuel cell is its ability to convert chemical energy directly to electrical energy giving much higher conversion efficiencies than any conventional thermo-mechanical system thus extracting more electricity from the same amount of fuel, to operate without combustion and without CO, NO_x, SO_x and CO_2 emissions so they are virtually pollution free. But the current technologies to produce the fuel–hydrogen (H_2) for fuel cell are energy-consuming and expensive. Therefore, it is required to find out an efficient and cheaper method to generate hydrogen.

Absorption enhanced reforming (AER) of coal/lignite may become a process that meets the requirement for H_2 production. The AER process combines steam gasification of coal, with the high temperature removal of CO_2 by using high temperature efficient sorbent materials (e.g. limestone or dolomite). The combination of the gasification and the in situ CO_2 capture shifts the reaction towards H_2 production in the gas stream. Experiments with different gas/liquid hydrocarbon fuel and dolomite revealed that hydrogen concentrations from 85% to 95 vol.% can be achieved using this technology [8,9]. The CO_2 laden sorbent material is regenerated in an additional regeneration step – calcination before being recycled back into the gasifier. This calcination step produces reactivated sorbent material and a CO_2 gas stream for subsequent sequestration.
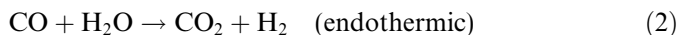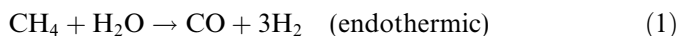
The objective of this study is to carry out a techno-economic analysis on the feasibility/possibility to combine the process of absorption enhanced reforming of lignite for H_2 generation with molten carbonate fuel cell, to realise a cleaner power generation with less greenhouse gas emissions.

2. The technologies of absorption enhanced reforming of fuel and fuel cell

The description of the features of absorption enhanced reforming of fuel (methane, methanol and propane/hexane) can be found in the literatures [8,9]; and the characteristics of fuel cell may be found in literature [7]. The following section is a brief description of AER process and the process of molten carbonate fuel cell (MCFC).

2.1. Absorption enhanced reforming of fuel

The main part of today's hydrogen produced is made via steam methane reforming [10]. This conventional technology for hydrogen generation from methane is characterised by steam reforming at temperatures of about 850–900 °C. The reactions are as following:

$$CH_4 + H_2O \rightarrow CO + 3H_2 \quad \text{(endothermic)} \tag{1}$$

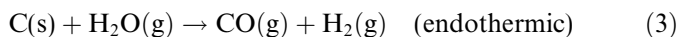$$CO + H_2O \rightarrow CO_2 + H_2 \quad \text{(endothermic)} \tag{2}$$

The hydrogen production process is endothermic. In other words, it is an energy consuming process. That means a large amount of heat must be added to the process in order to maintaining the reaction process.
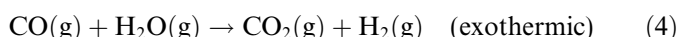
AER process is an alternative new approach with a relative lower temperature range of 600–700 °C for hydrogen generation. One advantage of the AER process represents the integration of the CO_2 absorption and CO shift reaction enthalpy (both exothermic reactions) into the fuel-to-hydrogen conversion process, which is highly endothermic in the conventional process. The high temperature absorbent material removes CO_2 during the fuel reforming process, enhancing the hydrogen production by shifting the reaction equilibrium towards increased hydrogen concentration.

The AER process is shown in Fig. 1. It combines steam-coal reforming, the water gas shift, and CO_2 sorption to produce a synthesis gas with relatively high hydrogen purity and low CO_2 and CO content. The reactions of AER process are as following:
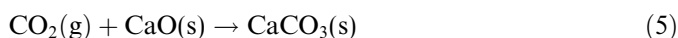
Carbon–H_2O reaction:

$$C(s) + H_2O(g) \rightarrow CO(g) + H_2(g) \quad \text{(endothermic)} \tag{3}$$

Water–gas shift reaction:

$$CO(g) + H_2O(g) \rightarrow CO_2(g) + H_2(g) \quad \text{(exothermic)} \tag{4}$$

In situ CO_2 sorption with sorbents:

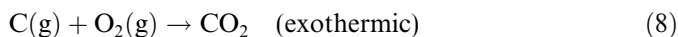$$CO_2(g) + CaO(s) \rightarrow CaCO_3(s) \tag{5}$$

Overall reaction scheme (integrated gasification and CO_2 sorption)
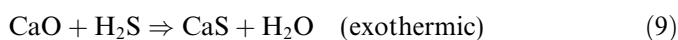
$$C + 2H_2O + CaO \rightarrow 2H_2 + CaCO_3 \quad \text{(exothermic)} \tag{6}$$

An additional calcination process for sorbent regeneration–desorption of $CaCO_3$:

$$CaCO_3 \rightarrow CO_2 + CaO \tag{7}$$

Partial oxidation for heat supply:

$$C(g) + O_2(g) \rightarrow CO_2 \quad \text{(exothermic)} \tag{8}$$

During the AER process, a sub-process of desulfurization reaction using CaO is also happened:

$$CaO + H_2S \Rightarrow CaS + H_2O \quad \text{(exothermic)} \tag{9}$$

$$CaS + \frac{3}{2}O_2 \Rightarrow CaO + SO_2 \quad \text{(exothermic)} \tag{10}$$

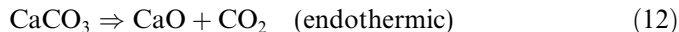$$CaO + SO_2 + \frac{1}{2}O_2 \Rightarrow CaSO_4 \quad \text{(exothermic)} \tag{11}$$
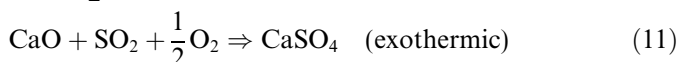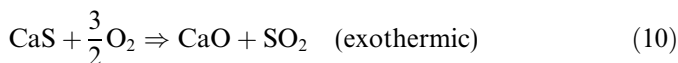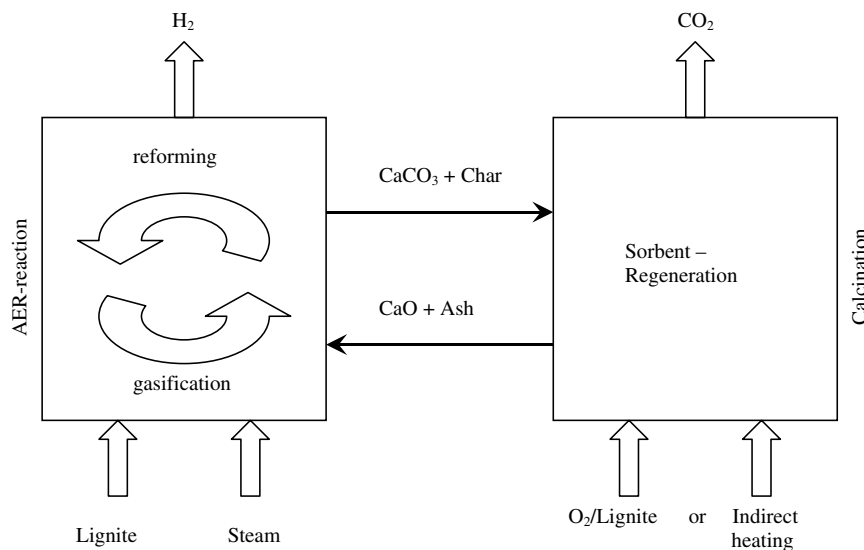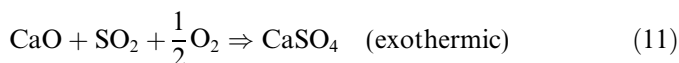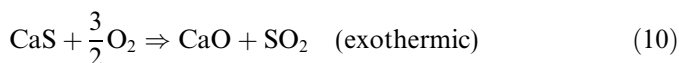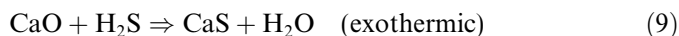
Fig. 1. Absorption enhanced reforming (AER) process.

In this process, H_2S in the produced gas is fixed as CaS, which is oxidized into gypsum in the oxidizer; thus sulphur is discharged from the system in the form of stable gypsum.
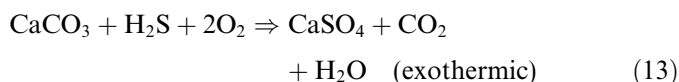
In calcinator, using limestone ($CaCO_3$), H_2S in the produced gas is also fixed as CaS, which is oxidized into gypsum in the oxidizer; thus sulfur is also discharged from the system in the form of stable gypsum. The reactions are

$$CaCO_3 \Rightarrow CaO + CO_2 \quad \text{(endothermic)} \tag{12}$$

and then, similarly as that in AER process,

$$CaO + H_2S \Rightarrow CaS + H_2O \quad \text{(exothermic)} \tag{9}$$

$$CaS + \frac{3}{2}O_2 \Rightarrow CaO + SO_2 \quad \text{(exothermic)} \tag{10}$$

$$CaO + SO_2 + \frac{1}{2}O_2 \Rightarrow CaSO_4 \quad \text{(exothermic)} \tag{11}$$

The overall reaction is

$$CaCO_3 + H_2S + 2O_2 \Rightarrow CaSO_4 + CO_2$$
$$+ H_2O \quad \text{(exothermic)} \tag{13}$$

2.2. Molten carbonate fuel cell

Molten carbonate fuel cell (MCFC) is developed in 1960s. MCFC uses an electrolyte composed of a molten mixture of carbonate salts. Two mixtures are used: lithium carbonate and potassium carbonate, or lithium carbonate and sodium carbonate. It operates at a high temperature (600–700 °C) to melt the carbonate salts and achieve high ion mobility through the electrolyte.

MCFC has its own advantages: they are more resistant to impurities than other fuel cell types; they are capable of internal reforming of carbon monoxide and methane; and they can be made resistant to impurities such as sulphur and particulates that result from converting coal process of AER.

The work principle is shown in Fig. 2. When heated to a temperature of around 650°C, these salts melt and become conductive to carbonate ions (CO_3^{2-}). These ions flow from the cathode to the anode where they combine with hydrogen to give water, carbon dioxide and electrons. These
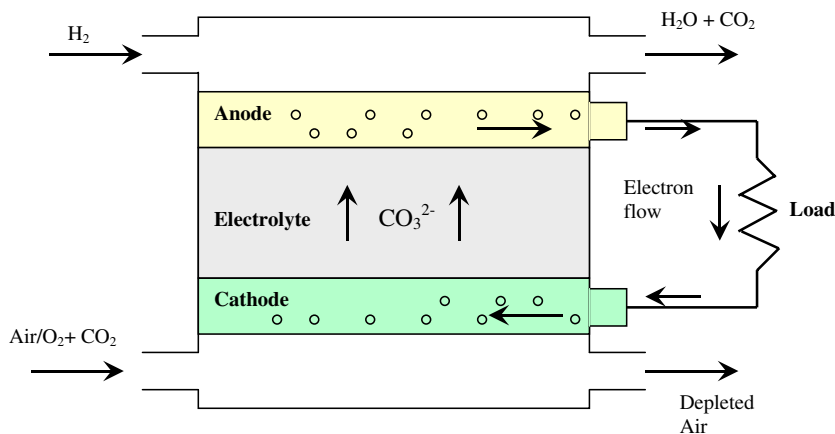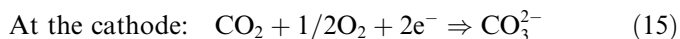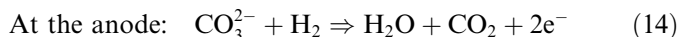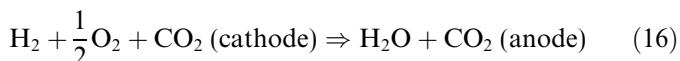

Fig. 2. Molten carbonate fuel cell.

electrons are routed through an external circuit back to the cathode, generating electricity and by-product heat.

The electrochemical reactions occurring in the cell are

$$\text{At the anode:} \quad CO_3^{2-} + H_2 \Rightarrow H_2O + CO_2 + 2e^- \tag{14}$$

$$\text{At the cathode:} \quad CO_2 + 1/2O_2 + 2e^- \Rightarrow CO_3^{2-} \tag{15}$$

With the overall cell reaction

$$H_2 + \frac{1}{2}O_2 + CO_2 \text{ (cathode)} \Rightarrow H_2O + CO_2 \text{ (anode)} \tag{16}$$

The CO_2 generated at the anode is recycled to the cathode where it is consumed.

3. The proposed combination system of AER with MCFC and simulation

3.1. The process of the combination system of AER with MCFC

The work process of a combination system of AER with MCFC is shown in Fig. 3. Lignite and steam are supplied to AER process reactor. In the reactor, lignite and steam react with calcium oxide (CaO) at high temperature (600–700 °C). Then, hydrogen, $CaCO_3$ and char are formed. Hydrogen is sent to fuel MCFC. $CaCO_3$ and char are sent to calcinator. In the calcinator, char is burnt together with the supply of lignite and oxygen to generate the heat required for $CaCO_3$ regeneration at high temperature (800–900 °C). $CaCO_3$ is decomposed to CaO and CO_2. CaO is delivered back to the AER reactor; high purity of CO_2 is released which depends on the purity of the oxygen input to the calcination process. The released CO_2 may then be collected by an auxiliary system and be delivered/pumped into underground to store.

In the MCFC, the hydrogen is reacted with the oxygen (either pure or in the air), to generate electricity and at the same time, to produce water. The depleted air is exhausted back to the atmosphere.

With the CO_2 released from the AER process captured, the combination system of AER with MCFC may be a feasible power generation with no or less greenhouse gases emissions.

3.2. The process simulator – a brief introduction of ECLIPSE

To provide a consistent basis for evaluation and comparison, the system analysed above are modelled using the ECLIPSE process simulation package [11–13]. ECLIPSE was developed for the European Commission and has been used by the Northern Ireland Centre for Energy Research and Technology at the University of Ulster since 1986 [14,15]. ECLIPSE was successfully used for many European and international projects to implement techno-economic analysis of power systems.

ECLIPSE is a personal-computer-based package containing all of the program modules necessary to complete rapid and reliable step-by-step technical, environmental and economic evaluations of chemical and allied processes. ECLIPSE uses generic chemical engineering equations and formulae and includes a high-accuracy steam–water thermodynamics package for steam cycle analysis. It has its own chemical industry capital costing program covering over 100 equipment types. The chemical compound properties database and the plant cost database can both be modified to allow new or conceptual processes to be evaluated. A techno-economic assessment study is carried out in stages; initially a process flow diagram is prepared, technical design data can then be added and a mass and energy balance completed. Consequently, the system's environmental impact is assessed, capital and operating costs are estimated and an economic analysis performed. Whilst every effort is made to validate the capital cost estimation data, using published information and actual quotations from equipment vendors, the absolute accuracy of this type of capital cost estimation procedure has been estimated at about 25–30%. However, as the comparative capital cost estimates are based on the accurate calculation by the mass and energy balance program of differences in basic design, families of similar technologies composed of similar types of equipment can be compared on a consistent basis.
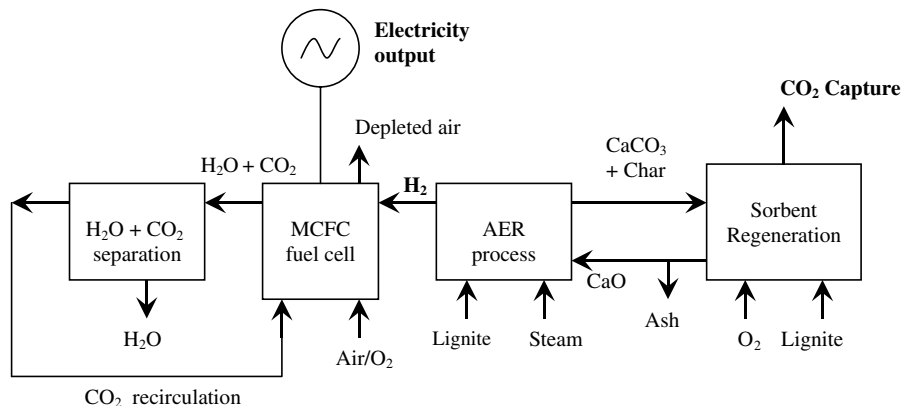

Fig. 3. The combination system of AER with MCFC.

4. Simulation results and discussion

4.1. The design conditions of MCFC and AER process system

The evaluated system is based on the case study of MCFC completed in the Energy Research Centre at the University of Ulster and described in literature [16]. The information of the costs of MCFC and AER process gasifier is estimated from sources in the literature [17,18]. Table 1 shows the design conditions of MCFC and AER process used in ECLIPSE simulation.

4.2. The results from the basic case study

A summary of technical, environmental and economic results is given in Table 2 for the 200 kW e MCFC plus AER process gasifier configuration. The results show that the original external MCFC case has an electrical efficiency of 53.1% (LHV), with the net electrical output of 206 kW and CO_2 emissions of 367 g/kW h, while fuelled with natural gas. For the combination of AER with MCFC, it has the same electricity output of 206 kW, but with an electrical efficiency of 44.7% (LHV) and CO_2 emissions of 751 g/kW h, while fuelled with lignite. The efficiency of AER with MCFC is lower and the CO_2 emission is higher than the original case, due to two reasons: one is that the lignite has a high carbon content than that of natural gas, which

Table 1
The process design conditions

MCFC	
Reformer type	External
Fuel feedstock	Natural gas
Sulphur removal technology	ZnO
CO_2 sequestration technology	None
Nominal electricity output (kW)	200.0
Anode inlet temperature (°C)	677
Cathode outlet temperature (°C)	745
Cathode exhaust recycle (%)	69.0
Operating pressure (Bar Abs)	1.04
Fuel utilisation (%)	95.0
Oxidant utilisation (%)	85.0
Inverter efficiency (%)	96.0
Operating temperature (°C)	650
Costs	
Package cost of MCFC (£/kW)	2353
Installed cost (£/kW)	2705
O&M cost (£/kW)	0.022
AER process	
Steam carbon ratio	2.5:1
Operating temperature (°C)	800–900
Sulphur removal technology	Flue gas desulphurization by limestone (FGD)
Costs	
Package cost of AER gasifier (£/kW)	1200
Installed cost (£/kW)	120
O&M cost (£/kW)	0.038

Table 2
Technical, environmental and economic results

System type	MCFC	AER + MCFC
Process details		
Reformer type	External	None
Fuel feedstock	Natural gas	Lignite
Sulphur removal technology	ZnO	FGD
CO_2 sequestration technology	None	None
Cathode exhaust recycle (%)	69.0	69.0
Operating temperature	865 °C	650 °C
CO shifter	No	No
Gasifier type	–	AER
Fuel input (kg/h)	–	43
Thermal input (kW, HHV)	427	478
Thermal input (kW, LHV)	388	460
Fuel cell power output (kW e DC)	220	220
Fuel cell power output (kW e AC)	211	211
Auxiliary power usage (kW e)	5	5
Net electrical output (kW e)	206	206
Electrical efficiency (%, HHV)	48.2	43.1
Electrical efficiency (%, LHV)	53.1	44.7
Gaseous emissions		
CO_2 (g/kW h)	367	751
SO_x (g/kW h)	0	0
NO_x (g/kW h)	–	–
Flue gas details		
MCFC		
Flow (kg/h)	517	723
Temperature (°C)	120	199
Composition (% w/w)		
N_2 + argon	76.2	75.5
CO_2	14.3	0.0
O_2	2.6	11.4
Water	6.9	13.1
	100.0	100.0
AER gasifier		
Flow (kg/h)		246
Temperature (°C)		304
Composition (% w/w)		
Water (from the high water/carbon ratio)		37.0
CO_2		63.0
		100.0
Economic analysis		
Capital cost (£)	2 158 000	2 398 000
Specific investment (£/kW e)	10 477	11 642
Break even electricity selling price (p/kW)	19	21

is 73.9%; another is that the AER process gasifier needs energy/heat input from the extra input of lignite to generate hydrogen.

The results of economic analysis for the basic case are shown in Table 2, with the package cost of MCFC £2353/kW e, the package cost of AER process gasifier £1200/kW e, an installed cost of £2705/kW e for the fuel cell, an installed cost of £120/kW e for the AER process gasifier, the lignite cost of 20 UK£/ton and the discount cash flowrate (DCF) of 10%. The cost of AER gasifier is obtained by scaling the values taken from literatures [18]. The MCFC has a 5 years' lifetime [17]; the remainder of the system is assumed to have a lifetime of 20 years. The

total specific investment (SI) for the system depends on the values for the lifetime of the fuel cell and its installed cost.

From the simulation result, the specific investment (SI) of AER + MCFC is found to be £11 642; the SI of MCFC is found to be £10 477 only, when fuelled with natural gas. The break even electricity selling price (BESP) is also shown in Table 2. For AER + MCFC, the BESP is 21 p/kW e; for MCFC, the BESP is 19 p/kW e.

4.3. Sensitivity analysis of the effect on the costs of the above systems

Figs. 4 and 5 show the sensitivity of BESP and SI versus the costs of AER + MCFC. The discussion is based on the basic case of the system shown in Table 2. The figures show that capital costs have a significant effect on BESP. Based on the basic case of capital cost of £2 398 000, BESP of £211/MW e (21 p/kW e), SI of £11 642/kW e, if the capital

cost of the system reduces 50%, the relevant BESP will lower down nearly 50% to £108/MW e (10.8 p/kW e), the SI will also reduce by 50%, to £5864/kW e; if the capital cost of the system increases 50%, the relevant BESP will increase to £315/MW e (31.5 p/kW e) and the SI will rise to £17 592/kW e.

The fuel cost has a little effect on BESP, as shown in Fig. 6, comparing to the effect of the capital costs. For the basic case of the system and the fuel (lignite) cost of £20/ton, with a discounted cash flowrate (DCF) of 10%, the BESP is £211/MW e (21.1 p/kW e). While the fuel cost reduces by 50%, to £10/ton, the BESP lower down to £209/MW e (20.9 p/kW e), only reduces 0.2 p/kW e, the change is 0.9%. When the fuel price increases by 50%, to £30/ton, the BESP increases to £213/kW e (21.3 p/kW e), the change is also 0.9%. The results of the effect of fuel cost on BESP for the DCF of 7.5% and 5% are also shown in Fig. 6, similar trends are found as that of DCF of 10%.
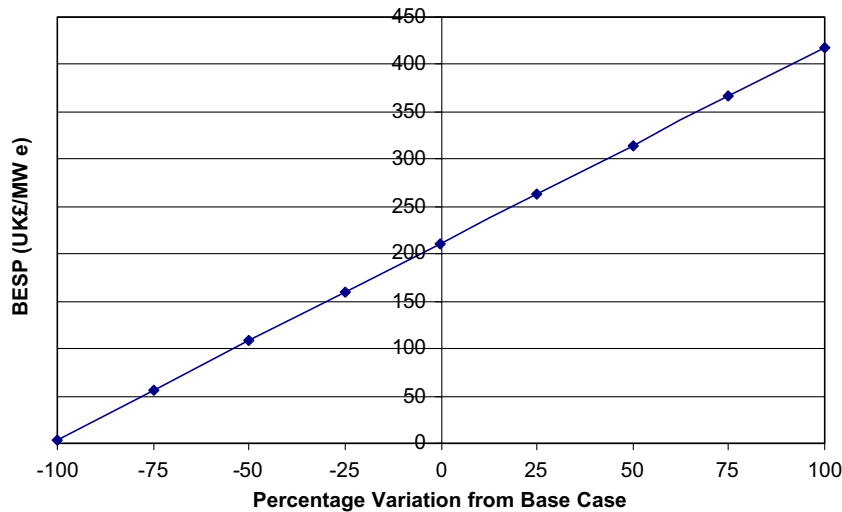

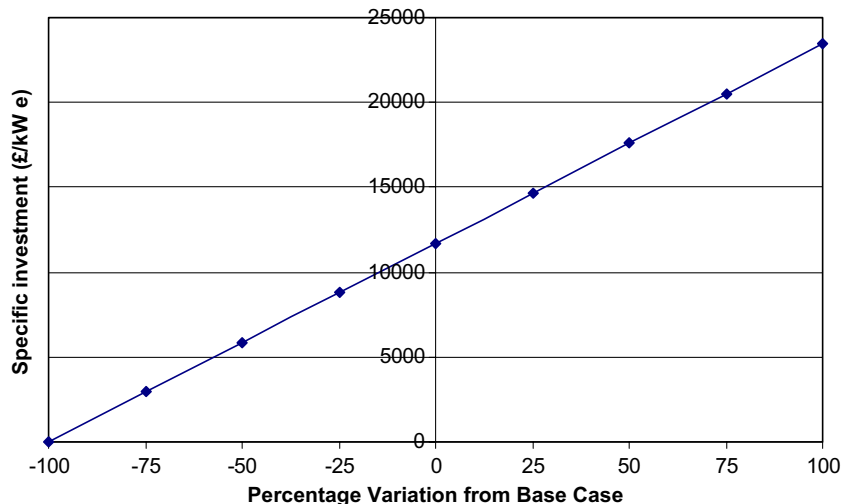Fig. 4. Sensitivity of electricity selling price versus (MCFC + AER) cost.

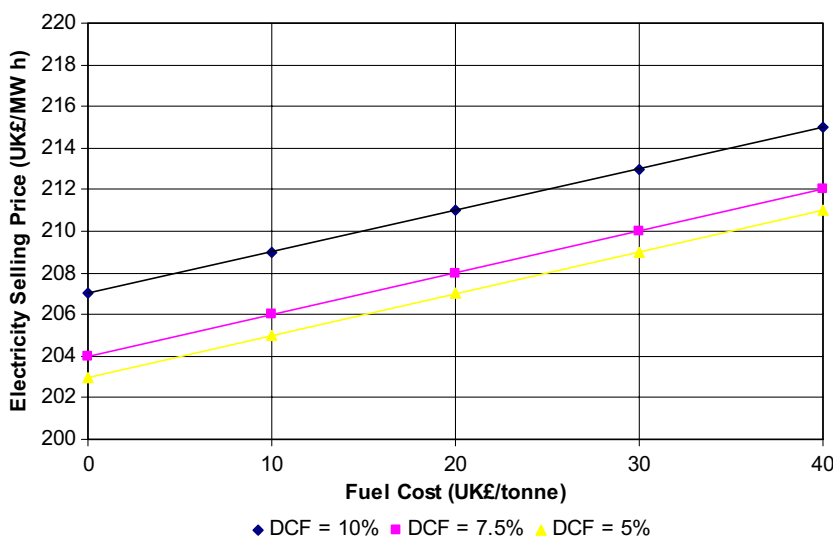Fig. 5. Sensitivity of specific investment versus (MCFC + AER) cost.

Fig. 6. Sensitivity of fuel cost.

5. Conclusions

The above assessment of a combination of absorption enhanced reforming (AER) of lignite with molten carbonate fuel cell (MCFC) was successfully completed using the ECLIPSE process simulator.

The results show that the AER + MCFC has an electricity output of 206 kW, with an electrical efficiency of 44.7% (LHV) and CO_2 emissions of 751 g/kW h, while fuelled with lignite.

The SI of AER + MCFC is £11 642; the BESP is 21 p/kW e. The results show that the SI and BESP are all high for the proposed system due to high cost of MCFC currently.

The results from the sensitivity analysis show that: (a) the capital costs have a significant effect on the BESP of AER + MCFC. When the capital cost of the system reduces 50%, the relevant BESP lowers down 50%, the SI will also reduce by 50%; (b) the fuel cost has a little effect on the BESP. When the fuel cost reduces by 50%, the BESP only reduces by 0.9%.

The results from techno-economic analysis show that the AER process to produce hydrogen from lignite to fuel MCFC has its environmental advantages over the conventional coal power plants. This would make it an attractive option for zero greenhouse gas emission power generations if CO_2 capture is applied. The technology may have its potential application when carbon tax comes into force in the future.

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