



Combined heat and power generation with lime production for direct air capture



Dawid P. Hanak*, Vasilije Manovic

Power Engineering Centre, Cranfield University, Bedford, Bedfordshire MK43 0AL, UK

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ABSTRACT

Carbon capture and storage (CCS) has been shown to be the least cost-intensive option for decarbonisation of the power, heat, and industrial sectors. Importantly, negative-emission technologies, including direct air capture (DAC), may still be required after near-complete decarbonisation of the stationary emission sources. This study evaluates the feasibility of a novel polygeneration process for combined heat and power using a solid-oxide fuel cell, and lime production for DAC (CHP-DAC) that could contribute towards decarbonisation of the power, heat, and industrial sectors. Evaluation of the thermodynamic performance indicated that such process can achieve the total efficiency and effective electric efficiency of 65%_{LHV} and 60%_{LHV}, respectively, while removing CO₂ from the air at a rate of 88.6 gCO₂/kW_{ch}h. With the total expenditure spread over a number of revenue streams, the product prices required for the CHP-DAC process to break even were found to be competitive compared to figures for the existing standalone technologies, even if there was no revenue from CO₂ capture from the air. Moreover, the considered process was shown to be economically feasible, even under uncertainty. Hence, it can be considered as the carbon-neutral polygeneration process for sustainable and affordable production of heat, power, and lime that is negative-emission ready.

1. Introduction

Decarbonisation of the energy and industrial sectors is key to meeting the Paris Agreement that recommended keeping the global mean temperature well below 2 °C and undertaking efforts to limit it to 1.5 °C above pre-industrial levels to significantly reduce the risks and impacts of climate change [1]. The power sector can be primarily decarbonised via deployment of carbon capture and storage (CCS), switching from fossil fuels to biomass, and wide deployment of renewable energy sources [2]. It has been reported, however, that decarbonisation of the power sector without CCS will be significantly more expensive and the additional investment associated with higher share of renewables may reach at least £3.5 billion by 2050 [3]. Similarly, a reduction of CO₂ emissions in the heat sector is heavily dependent on the large-scale deployment of CCS. This is because the electrification of heating in buildings will be achieved primarily by wide deployment of heat pumps or direct electric heaters, and large-scale implementation of low-carbon district heating systems, which are expected to be key measures to meet the emission reduction target by 2050 [4]. Importantly, a combination of CCS with hydrogen production [5] and combined heat and power (CHP) generation [6,7] is predicted to play a pivotal role in decarbonisation of process heating in the industrial

sector. The industrial processes are also highly energy intensive, and their decarbonisation is even more challenging than decarbonisation of the power and heat sectors. This is because CO₂ emissions not only stem from fossil fuel combustion, but in many cases also from the chemical process itself. Decarbonisation of the lime industry is of particular interest, as lime has multiple applications in other industries and sectors, such as in environmental protection (flue gas treatment, water purification), agriculture (fertiliser production, soil and wastewater treatment), chemical production (calcium carbide production) and manufacturing (high-performance materials production). Therefore, to achieve near-complete decarbonisation of this industry, CCS is required to avoid the CO₂ emissions from both the fuel combustion and the process itself [8]. Importantly, use of lime as a sorbent for CO₂ capture from fossil fuel power plants [9,10] and industrial processes [11,12] has been recently regarded as a feasible option to reduce the energy and economic penalties associated with mature CCS technologies, such as chemical solvent scrubbing or oxy-fuel combustion. Therefore, the potential exists for synergy between the power and heat sectors, and the lime industry that would lead to reduced environmental burden associated with these processes, while ensuring that the product costs are affordable.

The near-complete decarbonisation of the power, heat and

* Corresponding author.

E-mail address: d.p.hanak@cranfield.ac.uk (D.P. Hanak).

Nomenclature

AC_k	cross-section area of heat exchanger k or solid-oxide fuel cell, m^2
C_j	capital cost of equipment j , £
CT	corporate tax, £
D	depreciation rate, £
$e_{CO_2,air}$	specific negative CO_2 emission, gCO_2/kW_{chh}
$e_{CO_2,seq}$	specific CO_2 sequestered, gCO_2/kW_{chh}
E	total expenditure, £
IRR	Internal rate of return
L_I	loan interest, £
L_P	loan principal, £
LHV	lower heating value of fuel, kJ/kg
m_{calc}	calcined material production rate, kg/s
$m_{CO_2,air}$	rate of CO_2 removal from air, kg/s
$m_{CO_2,seq}$	rate of CO_2 sequestered, kg/s
m_{fuel}	fuel consumption rate, kg/s
m_{O_2}	O_2 production rate in the air separation unit, kg/s
CF	net cash flow, £
NPV	net present value, £
PI	Profitability index
q	corporate tax rate, –
R	total revenue, £
SV	salvage value, £

T_{SOFC}	solid-oxide fuel cell operating temperature, °C
TCI	total capital investment, £
TCR	total capital requirement, £
Q_{DH}	heat output to district heating network, kW_{th}
$W_{j,BRK}$	brake power output/requirement of equipment j , kW_{el}
W_{net}	net power output of the entire system, kW_{el}
$W_{SOFC,DC}$	solid-oxide fuel cell DC power output, kW_{el}
η_b	thermal efficiency of conventional natural gas-fired boiler, –
η_{el}	effective electrical efficiency, –
η_{tot}	total system efficiency, –

Abbreviations

ASU	Air separation unit
CHP	Combined heat and power
CHP-DAC	Combined heat and power, and lime production for direct air capture
CCS	Carbon capture and storage
CCU	CO_2 compression unit
DAC	Direct air capture
NGCC	Natural gas combined cycle power plant
NPV	Net present value
SOFC	Solid-oxide fuel cell

industrial sectors may not be sufficient to meet the CO_2 emission reduction targets, and negative-emission technologies that remove CO_2 directly from the atmosphere may be required [13,14]. Direct air capture (DAC) can address emissions from both point and distributed sources, including emissions from agriculture, buildings and transportation sectors that account roughly for half the annual anthropogenic CO_2 emissions [15,16]. However, due to extremely low concentrations of CO_2 in the ambient air, the cost of CO_2 capture from the air has been estimated to fall between 400 £/t CO_2 and 800 £/t CO_2 [15], which is an order of magnitude higher than those reported for CO_2 capture from combustion processes [17]. Use of lime as a sorbent in DAC concepts has been found to be effective in removing CO_2 from the air. Lackner et al. [18] was the first to propose the concept of using calcium hydroxide for DAC. Although such concept was found to capture CO_2 from

air efficiently, it was deemed unfeasible due to high regeneration energy of the calcium hydroxide. Zeman and Lackner [19] proposed an alternative DAC concept based on the Kraft process, in which sodium hydroxide is used instead of calcium hydroxide for CO_2 capture, while the latter is used for regeneration of the sodium hydroxide from sodium carbonate. Yet, such concept was also shown to require a similar amount of energy for regeneration of sorbent. Finally, Nikulshina et al. [20] proposed using lime as a solid sorbent for DAC in a fluidised bed. The heat required for sorbent regeneration was provided by solar energy. Such DAC concept was shown to achieve a higher CO_2 capture level compared to the one using Na-based sorbents, but required the air to enter the reactor at an elevated temperature (375 °C). As a result, the heat requirement for such concept was shown to be higher than that with alkali metal hydroxide solutions. Interestingly, the concepts

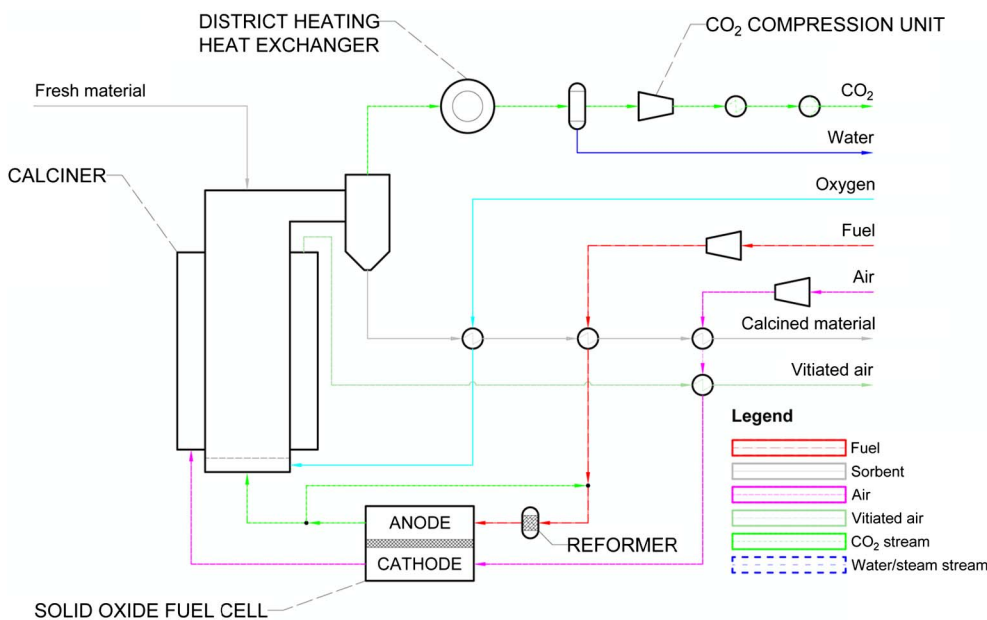


Fig. 1. Process flow diagram of system for combined heat and power generation, and lime production for direct CO_2 removal from the air.

presented in the current literature did not consider linking the DAC process with energy recovery systems for heat and/or power production. As a result, these have been deemed as energy intensive, due to high regeneration temperatures (800–950 °C) and oxy-fuel combustion in the calciner to maintain high purity of the produced CO₂ stream [15]. Recently, Hanak et al. [21] proposed a process for simultaneous power generation and CO₂ removal from the air using solid sorbents. That process utilised high-grade heat from a solid-oxide fuel cell (SOFC) to drive lime production. Although such process generated electricity at a high efficiency (47.7%_{LHV}) and was shown to have the potential to remove CO₂ from the air at a rate of 463.5 gCO₂/kW_{el,h}, its poly-generation capabilities, and thus additional revenue streams, were not fully exploited. Namely, in addition to generating power and producing lime only for DAC, such system could also produce heat for district heating. This allows a more efficient utilisation of the low-grade heat available in the process. Also, part of the produced lime can be sold for other uses, instead of being wholly utilised for DAC.

Deployment of such polygeneration systems for combined heat and power, and lime production for DAC (CHP-DAC) could contribute towards decarbonisation of the power, heat, and industrial sectors (especially the lime industry), without compromising their competitiveness and affecting product affordability. This study, therefore, evaluated the techno-economic performance of a novel polygeneration CHP-DAC process to demonstrate its feasibility, with particular attention paid to the economic performance assessment aimed at identifying and evaluating the main sources of revenue. Also, a sensitivity analysis was conducted to assess the effect of the assumptions in both the process and economic models on a number of key performance indicators including efficiency, specific negative emissions, break-even carbon tax, profitability index, payback time and internal rate of return. Finally, the stochastic approach was employed to assess the effect of uncertainty in the input parameters on the profitability index.

2. Process description

The CHP-DAC process proposed in this study (Fig. 1) includes a SOFC, fresh material calciner, CO₂ compression unit (CCU), heat exchanger network, and a district heating heat exchanger. The heart of the process is the flash calciner, where the fresh material is decomposed upon heating. The temperature at which the calcination takes place depends on the CO₂ partial pressure and type of fresh material fed to the calciner. Under pure CO₂ conditions, this can vary between 550 °C for magnesite and dolomite (partial calcination), to 900 °C for dolomite (complete calcination) and limestone [22,23]. In this study, limestone is considered as the fresh material due to its low cost and wide availability, and thus the calciner is being operated at 900 °C. Moreover, this sorbent has been shown to provide the optimum balance between thermodynamic and economic performance compared to dolomite and magnesite [21]. The amount of fresh material fed to the calciner and the size of this reactor are determined by the availability of high-grade heat in the high-temperature gas streams from the 25 MW_{el,DC} SOFC at 950 °C that is used to drive the calcination process. Importantly, the CO₂/H₂O stream leaving the anode contains some fuel, which was not completely utilised in the SOFC [24,25]. Therefore, a high-purity O₂ stream (~95%_{vol} O₂), which is produced in the air separation unit (ASU), is fed directly to the calciner to burn the unutilised fuel. As a result, heat is generated directly in the calciner to sustain the calcination process. To ensure that the CO₂ stream purity is suitable for geological sequestration (> 90%_{vol} CO₂ [26]), the vitiated air leaving the cathode indirectly provides heat to the calciner via a heat transfer jacket surrounding the reactor.

Importantly, the streams leaving the calciner, including the calcined material, CO₂ and vitiated air streams, carry a significant amount of high-grade heat that can be utilised within the system. As presented in Fig. 1, this heat is recovered for O₂, fuel, air, and fresh material pre-heating. Moreover, the high-grade heat from the concentrated CO₂ stream leaving the calciner is utilised in a district heating network. The calcined material, can then be distributed in the open environment for

Table 1
Design conditions and thermodynamic model assumptions.

Parameter	Value	
Solid-oxide fuel cell	Gibbs reactors for pre-reformer and anode. Component splitter for cathode. Linked with electrochemical calculator in MS Excel. SOFC model developed based on Zhang et al. [25]. Natural gas composition adapted from the revised NETL report [27]	
	Temperature (°C)	950
	Pressure (bar)	1.08
	Fuel utilisation (%)	85
	Reference conditions: Fuel composition (67% _{vol} H ₂ , 22% _{vol} CO, 11% _{vol} H ₂ O), fuel utilisation (U _f = 85%), air utilisation (U _a = 25%), operating temperature (T = 1000 °C), Operating pressure (P = 1 bar), H ₂ -to-H ₂ O partial pressure ratio (p _{H₂,ref} /p _{H₂O,ref} = 0.15), O ₂ partial pressure at cathode (p _{O₂,ref} = 0.164)	
Calciner	Gibbs reactor. Gibbs free energy minimisation model	
	Temperature (°C)	550–900
	Pressure drop (mbar)	150
	Excess O ₂ (%)	2
	Power requirement (kW _{el,h} /tO ₂)	200
Heat loss (%)	4	
Heat exchanger network	Minimum approach temperature in the O ₂ preheater (°C)	10
	Fuel outlet temperature from the fuel preheater (°C)	200
	Calcined material outlet temperature from first air preheater (APX1) (°C)	50
	Minimum approach temperature in the second air preheater (APX2) (°C)	10
	Desired temperature increase in the district heating network (°C)	30
District heating water return temperature (°C)	30	
CO ₂ compression unit	Intercooling temperature (°C)	40
	CO ₂ delivery pressure (bar)	110
	Polytropic efficiency of CO ₂ compressors (%)	77–80
	Isentropic efficiency of CO ₂ pump (%)	85
Mechanical efficiency of compressors and pump (%)	99.6	
Fresh material	Limestone (95% _{wt} CaCO ₃ , 3.5% _{wt} MgCO ₃ , 0.6% _{wt} SiO ₂ , 0.4% _{wt} Fe ₂ O ₃ , 0.5% _{wt} Al ₂ O ₃)	
Fuel	Natural gas (93.1% _{vol} CH ₄ , 3.2% _{vol} C ₂ H ₆ , 0.7% _{vol} C ₃ H ₈ , 0.4% _{vol} C ₄ H ₁₀ , 1.0% _{vol} CO ₂ , 1.6% _{vol} N ₂)	

Table 2
Capital cost estimation and economic model assumptions.

Equipment [Scaling parameter]	Correlation
Solid-oxide fuel cell stack [Active area, AC_{SOFC} (m ²); Operating temperature, T_{SOFC} (K) [31]]	$C_{SOFC} = AC_{SOFC} (2.96T_{SOFC} - 1907)$
DC-to-AC inverter [Rated power output, $W_{SOFC,DC}$ (kW) [31]]	$C_{SOFC,DC/AC} = 1e5 \left(\frac{W_{SOFC,DC}}{500} \right)^{0.7}$
Solid-oxide fuel cell auxiliaries [Stack cost, C_{SOFC} (USD) [31]]	$C_{SOFC,aux} = 0.1C_{SOFC}$
Fuel compressor [Brake power requirement, $W_{FC,BRK}$ (kW) [31,32]]	$C_{FC} = 91,562 \left(\frac{W_{FC,BRK}}{445} \right)^{0.67}$
Air compressor [Brake power requirement, $W_{AC,BRK}$ (kW) [31,32]]	$C_{AC} = 91,562 \left(\frac{W_{AC,BRK}}{445} \right)^{0.67}$
Fuel preheater [Heat exchange area, AC_{FPH} (m ²) [32]]	$C_{FPH} = 130 \left(\frac{AC_{FPH}}{0.093} \right)$
Oxygen preheater [Heat exchange area, AC_{OPH} (m ²) [32]]	$C_{OPH} = 130 \left(\frac{AC_{OPH}}{0.093} \right)$
Air preheater 1 [Heat exchange area, AC_{APH1} (m ²) [32]]	$C_{APH1} = 130 \left(\frac{AC_{APH1}}{0.093} \right)$
Air preheater 2 [Heat exchange area, AC_{APH2} (m ²) [31]]	$C_{APH2} = 2290(AC_{APH2})^{0.6}$
District heating heat exchanger [Heat exchange area, AC_{DHX} (m ²) [31]]	$C_{APH2} = 2290(AC_{DHX})^{0.6}$
Air separation unit [O ₂ production rate, m_{O_2} (kg/s) [33]]	$C_{ASU} = 2.926e5 \left(\frac{m_{O_2}}{28.9} \right)^{0.7}$
CO ₂ compression unit [Brake power requirement, W_{CCU} (kW) [34]]	$C_{CCU} = 1.22914e7 \left(\frac{W_{CCU,BRK}}{13000} \right)^{0.67}$
Calciner [Material production rate, m_{calc} (kg/s) [35]]	$C_{calc} = 1.30523e8 \left(\frac{m_{calc}}{344.24} \right)^{0.7}$

CO₂ capture over an elongated period of time, or sold as a feedstock to other industries. On the other hand, the concentrated CO₂ stream, after dehydration, is compressed to 110 bar and sent for storage. The main design conditions and thermodynamic model assumptions are reported in Table 1.

3. Techno-economic feasibility assessment

3.1. Thermodynamic performance indicators

The thermodynamic performance of the CHP-DAC was characterised using the key performance indicators that have been commonly used to assess the performance of conventional CHP systems. Primarily, the net power output (W_{net}), which accounted for the power output from the SOFC less any parasitic load, heat output to the district heating system (Q_{DH}), the total system efficiency (η_{tot}), and effective electric efficiency (η_{el}) are used. The total efficiency is defined in Eq. (1) as the ratio of the sum of the net power output and the heat output, and the chemical energy input to the system, which was calculated as the product of the fuel consumption rate (m_{fuel}) and its lower heating value (LHV). The effective electric efficiency is defined in Eq. (2) as the fraction of the net power output and the chemical energy input to the system less the chemical energy input associated with the heat output. This also considered the efficiency of the conventional natural gas-fired boiler (η_b) of 80%.

$$\eta_{tot} = \frac{W_{net} + Q_{DH}}{m_{fuel}LHV} \quad (1)$$

$$\eta_{el} = \frac{W_{net}}{m_{fuel}LHV - Q_{DH}/\eta_b} \quad (2)$$

The CO₂ capture performance and the environmental performance were quantified in terms of the specific CO₂ sequestered ($e_{CO_2,seq}$) and specific negative CO₂ emissions ($e_{CO_2,air}$) defined in Eq. (3) as the ratio of the CO₂ rate ($m_{CO_2,i}$), where the subscript i indicates whether sequestered (seq) or removed CO₂ (air) is concerned, and the chemical energy input to the system.

$$e_{CO_2,i} = \frac{m_{CO_2,i}}{m_{fuel}LHV} \quad (3)$$

Such a definition of the environmental performance relates the CO₂ emissions to the chemical energy input that is opposed to the general convention that relates the specific CO₂ emissions to the net power output of the system. However, as there are a number of products streams produced in the CHP-DAC process (heat, power, lime, and, potentially, concentrated CO₂), relating the specific emissions to one of them would not provide a representative figure. To provide a benchmark value for this environmental indicator, it can be calculated for the conventional natural gas combined cycle power plant (NGCC) that was used in the study by Biliyok et al. [28]. This power plant was characterised with the net power output of 440.6 MW_{el} and net efficiency of 59.6%_{LHV}. The specific CO₂ emissions would be 354.5 gCO₂/kW_{el}h and 211.4 gCO₂/kW_{ch}h, respectively, if net power and chemical energy input are considered as the basis for the environmental performance assessment.

3.2. Economic performance indicators

The economic performance of the CHP-DAC process was assessed using the net present value (NPV) approach that is commonly applied in assessment of engineering systems [29,30]. The NPV is defined in Eq. (4) as the difference between the total capital investment (TCI) and the sum of discounted cash flows throughout the project lifetime:

$$NPV = \sum_{t=1}^n \frac{CF_t}{(1+r)^t} - TCI \quad (4)$$

The total capital investment is calculated as the difference between the total capital requirement (TCR) and the amount of loan ($L_{p,t}$), which was assumed to be amortised according to the fixed principal loan schedule. The total capital requirement was determined from the capital cost correlations for each unit in the CHP-DAC process (Table 2). These correlations have been selected from literature studies that analysed systems of comparable scale (2–60 MW_{el,DC}) and were based on the exponential method function to consider the effect of system scale on the capital cost. Furthermore, the net cash flow (CF_t) in each year t was calculated using Eq. (5). This primarily considers:

- the total revenue (R_t) from electricity, heat, lime, and negative CO₂ emissions;
- total operating expenditure (E_t), which accounts for CO₂ transport and storage, fuel, sorbent, as well as operating and maintenance expenditure;
- corporate tax (CT_t);
- loan principal ($L_{p,t}$) and interest ($L_{i,t}$) payment; and
- salvage value (SV_t).

$$CF_t = R_t - E_t - CT_t - L_{p,t} - L_{i,t} + SV_t \quad (5)$$

Importantly, fixed and variable operating and maintenance costs are calculated as a fraction of total capital cost, while the revenue associated with electricity, heat, and lime production and negative CO₂ emissions, and operating costs associated with fuel and sorbent consumption, and CO₂ transport and storage, were determined based on process simulation outputs using economic data from Table 3. It is also assumed that the SOFC electric output degrades at a rate of 5 mV per 1000 h [36], which affects the revenue from electricity sales.

In the first part of the economic analysis, the cost associated with the negative CO₂ emissions was estimated with the assumption that it would be the minimum break-even cost at which NPV becomes zero after the lifetime of the project and was referred to as the break-even carbon tax. Furthermore, it needs to be noted that both the loan interest payment and the depreciation cost (D_t) were deducted as a business expense, as shown in Eq. (6), acting as a tax shield [37].

Table 3
Assumptions for the economic model.

Parameter	Distribution	Nominal value	Variation ^a
<i>Project characteristics</i>			
Expected lifetime (years) [38]	Not considered	25	0
Capacity factor (%) [38]	Uniform	80.0	50–100
Project interest rate (%) [38]	Normal	8.78	10
Inflation rate (%)	Normal	1.0	10
Depreciation rate (%)	Uniform	1.0	0–10
<i>Technical assumptions</i>			
Fuel cell degradation (mV/1000 h)	Uniform	5	0–10
Sorbent conversion (%)	Uniform	80	20–80
Amount of calcined product to sales (%)	Uniform	50.0	25–75
<i>Total capital requirement distribution</i>			
Owner's equity share (%)	Uniform	50.0	30–100
Loan share (%)	Uniform	50.0	0–70
<i>Project costs</i>			
Variable operating cost as a fraction of total capital cost (%) [38]	Normal	2.0	10
Fixed operating cost as a fraction of total capital cost (%) [38]	Normal	1.0	10
Limestone cost (£/t) [38]	Normal	6.0	20
CO ₂ transport and storage cost (£/tCO ₂) [38]	Uniform	7.0	–15–40
Natural gas price (£/GJ) [39]	Normal	3.0	20
Loan interest rate (%)	Uniform	5.0	1–10
Corporate tax rate (%)	Normal	20.0	10
<i>Product prices</i>			
Electricity (£/MW _{el} h)	Normal	40.0	20
Heat (£/MW _{th} h)	Normal	30.0	20
Calcined product (£/t)	Normal	40.0	20

^a Coefficient of variation for normal distribution and a range for uniform distribution.

$$CT_t = q(R_t - E_t - L_{t,t} - D_t) \quad (6)$$

In the second part of the economic analysis, the influence of the carbon tax on the profitability index (*PI*), which is defined in Eq. (7) as a function of NPV and the total capital investment [40], the discounted payback time, and internal rate of return (IRR) are estimated.

$$PI = 1 + \frac{NPV}{TCI} \quad (7)$$

In addition, the effect of uncertainty in the assumptions for the economic model on prediction of the profitability index is evaluated using the Monte Carlo simulation. This is achieved by estimating the profitability index using the input dataset that contains ten thousand entries that have been randomly generated according to the assumed distributions of the input variables in the economic model (Table 3).

4. Results and discussion

4.1. Thermodynamic performance

The thermodynamic assessment of the CHP-DAC process feasibility (Table 4) revealed that under the initial design assumptions, the process was characterised with net heat and power outputs of 8.9 MW_{th} and 20.1 MW_{el}, respectively. Such outputs resulted in the total efficiency of the entire process of 65%_{LHV} and the effective electric efficiency of 60%_{LHV}, which are comparable to the performance of other fuel-cell-based CHP systems reported in the literature [41]. Notably, in addition to producing heat and electricity with reasonably high total efficiency, the considered process was shown to have the capacity to produce the calcined material (lime) at a rate of 307.4 t/d. Having assumed that half of this amount is utilised for direct CO₂ capture from the air and that lime can achieve 80% conversion in the long term when distributed in the open environment, the amount of CO₂ removed from the air was

estimated to be 88.6 gCO₂/kW_{ch}h. Such environmental performance would alleviate around 42% of the CO₂ emissions from the conventional NGCC without a CO₂ capture system [28] that emits 211.4 gCO₂/kW_{ch}h (354.5 gCO₂/kW_{ch}h). In addition, the CHP-DAC was shown to produce a concentrated CO₂ stream (> 98%_{v,oi}), which combines CO₂ emissions from the SOFC and the calciner, at a rate of 428.6 gCO₂/kW_{ch}h. This provides another potential revenue stream for the CHP-DAC process, if CO₂ was utilised, for example, for enhanced oil recovery.

Importantly, the performance of the considered CHP-DAC process was directly dependent upon the performance of the SOFC, which was the primary generator of heat and power in the overall system. Having analysed the effect of the fuel utilisation and current density in the SOFC (Fig. 2), a trade-off between the thermodynamic and environmental performance of the CHP-DAC process has been observed. First, an increase in the fuel utilisation in the SOFC from 85% to 90% caused a 1.7%-point increase in the total efficiency (Fig. 2a). This can be primarily associated with more electricity produced in the SOFC and lower O₂ requirement in the calciner. Importantly, as more fuel was utilised in the SOFC, less was available for calcination of limestone in the calciner. As a result, a lower amount of lime was produced and thus available for CO₂ removal from the air. In turn, the specific negative emissions reduced by 8.7% on the fuel utilisation increase from 85% to 90%. Second, an increase in the current density from 250 mA cm⁻² to 300 mA cm⁻² was shown to result in a 5%-point drop in the total efficiency (Fig. 2b). This can be primarily associated with a drop in the effective electric efficiency as a result of increased voltage losses [21,42]. Nevertheless, operation under such conditions increased the amount of heat available for the calcination of limestone, which, in turn, caused a 12.3% increase in the specific negative emissions. Therefore, the optimum fuel utilisation and current density must be a balance between the thermodynamic, environmental, and economic performance of the CHP-DAC process.

4.2. Economic performance

The economic assessment of the CHP-DAC process indicated that under the initial economic assumptions the specific capital requirement was 744.6 £/kW_{ch}. This figure was found to be higher than that of conventional NGCC without CO₂ capture, for which the specific capital requirement was estimated to be between 280 and 400 £/kW_{ch} (550–680 £/kW_{el}). If CO₂ capture was considered, the specific capital requirement increased to between 530 and 540 £/kW_{ch} (1080–1185 £/kW_{el}) [43,44]. The higher capital requirement of the CHP-DAC process was mainly a result of high capital cost of the SOFC, as it accounted for almost 70% of the total capital requirement (Fig. 3). Nevertheless, there is the potential for significant cost reduction of the SOFC as these become widely deployed [45]. Other important contributors to the capital requirement of the considered process were CCU and ASU, as these units accounted for 15.3% of the total capital requirement (Fig. 3). It can be expected that their contribution would be reduced under

Table 4
Key thermodynamic and environmental performance indicators.

Parameter	Value
Chemical energy input (MW _{ch})	44.7
District heating heat output (MW _{th})	8.9
Solid-oxide fuel cell gross power output (MW _{el})	23.0
System parasitic load (MW _{el})	2.9
System net power output (MW _{el})	20.1
Total efficiency (% _{LHV})	65.0
Effective electric efficiency (% _{LHV})	60.0
Calcined material production rate (t/d)	307.4
Specific negative CO ₂ emission (gCO ₂ /kW _{ch} h)	88.6
Specific CO ₂ sequestration rate (gCO ₂ /kW _{ch} h)	428.6

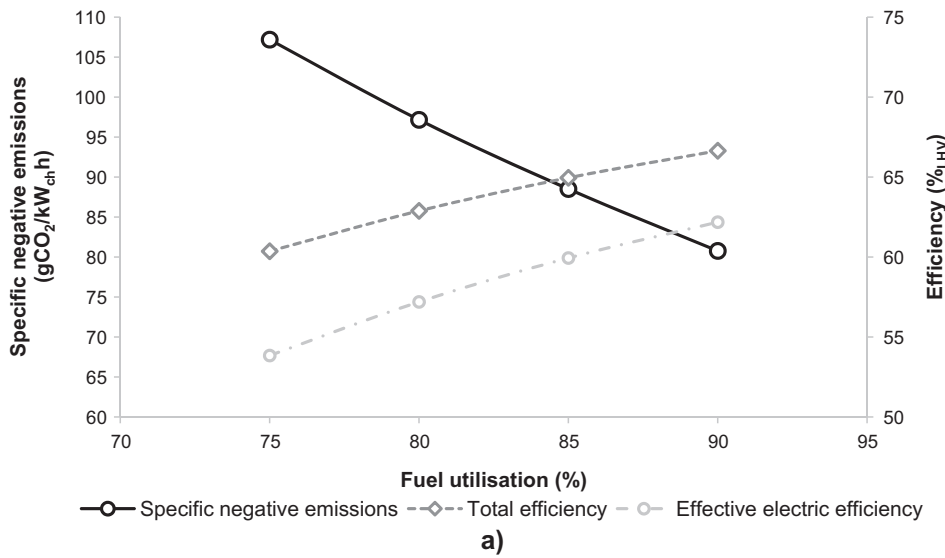


Fig. 2. Effect of (a) fuel utilisation of, and (b) current density in, the solid-oxide fuel cell on the thermodynamic and environmental performance of the CHP-DAC process.

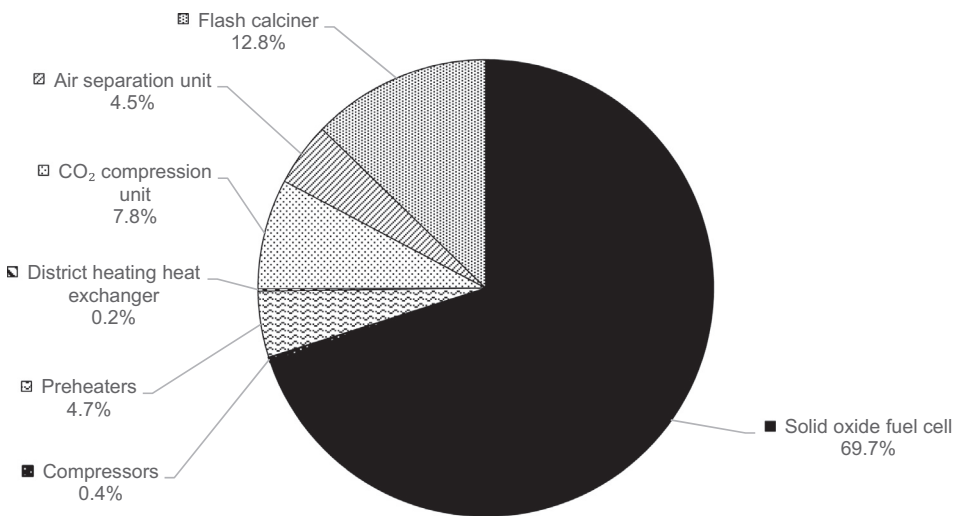
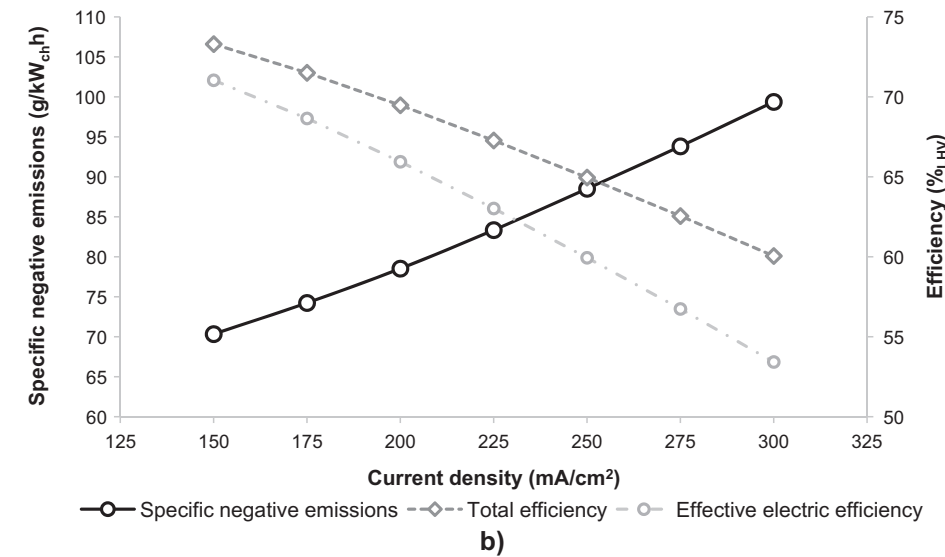


Fig. 3. Distribution of the total capital requirement.

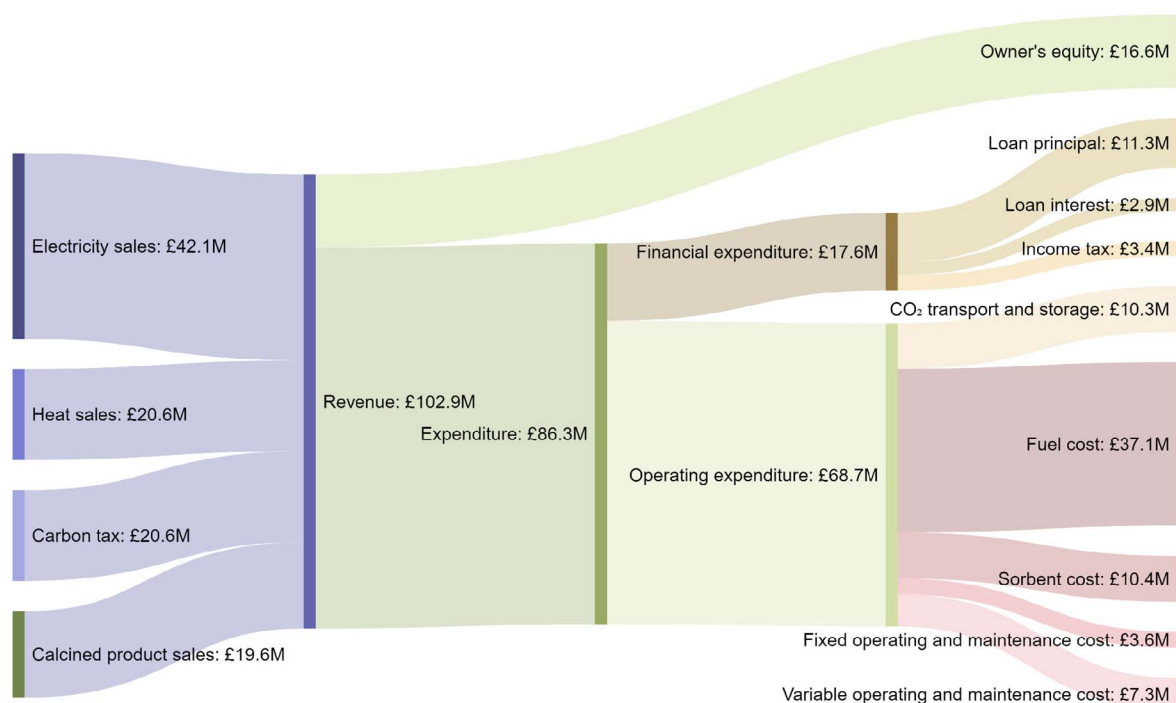


Fig. 4. Overview of the net present value analysis results.

operating conditions that result in high efficiency of the SOFC, for example, at low current density and high fuel utilisation, because such would reduce the amount of heat available for material calcination.

The NPV analysis (Fig. 4) indicated that for the CHP-DAC process to break even over the project lifetime (25 years), the carbon tax would need to be 68 £/tCO₂, assuming that the prices of electricity, heat and lime are 40 £/MW_{el}h, 30 £/MW_{th}h, and 40 £/t, respectively. This revealed the advantage of the proposed process over the conventional NGCC with CO₂ capture, as regardless of lower capital requirement, the cost of electricity and the cost of CO₂ avoided for that system were estimated to be 67 £/MW_{el}h and 75 £/tCO₂ [43], respectively. Importantly, the cost of electricity of the conventional NGCC without CO₂ capture was reported to be 46 £/MW_{el}h, which is higher than that used in the CHP-DAC process evaluation. This proved that the CHP-DAC process can be a competitive option to the existing power plant fleet. Moreover, the NPV analysis (Fig. 4) showed that the electricity sales were the most important revenue stream in the CHP-DAC process, nearly double that of the other streams. Importantly, these were balanced, bringing approximately the same revenue over the process lifetime. The analysis indicated also that the financial expenditure, which was associated with the loan interest, loan principal, and income tax, accounted for around 20% of the total expenditure over the process lifetime. This implied that the economic performance of the CHP-DAC process is much less dependent upon market conditions, compared to the thermodynamic performance. This also indicates that the economic performance of this process could be improved via optimisation of its thermodynamic performance to reduce the operating expenditure, primarily via reduction of the fuel consumption through improved heat integration. Moreover, it is expected that a significant cost reduction of the SOFC can be achieved once this technology becomes commercially deployed [5].

Improvement in the thermodynamic performance of the CHP-DAC process can be primarily associated with an increase in the efficiency of the SOFC. As indicated above in Fig. 2, this can be primarily realised by increasing the fuel utilisation and reducing the current density in the SOFC. However, the opposite trend was observed in the economic performance of the CHP-DAC process. Namely, an increase in the fuel utilisation from 85% to 90% resulted in a 10% surge in the break-even

carbon tax (Fig. 5a). This can be associated with a higher influence of a 3% increase in the specific capital requirement on the expenditure compared to a 2.6% increase in the total efficiency on the revenue over the process lifetime. Moreover, an 8.7% reduction in the specific negative emissions could be associated with lower revenues from direct air capture. Similarly, a reduction in the current density from 250 mA cm⁻² to 150 mA cm⁻² resulted in a 90% increase in the break-even carbon tax (Fig. 5b). Again, such degradation in the economic performance arises from a 28% increase in the specific capital requirement, mainly due to an increase in the SOFC area, and a 21% reduction in the specific negative emissions. Such results indicated the trade-off between the thermodynamic, environmental, and economic performance of the CHP-DAC process, the last of which would be maximised when the specific negative emissions were maximised.

The economic performance of the CHP-DAC process was also found to be sensitive to the economic assumptions related to utility and product prices, as well as the project characteristics. The results of the sensitivity analysis presented in Fig. 6a indicated that the break-even carbon tax was mostly sensitive to the fuel cost, because when it doubled from 3 £/GJ to 6 £/GJ, the break-even carbon tax increased by a factor of 2.2, from 68 £/tCO₂ to 149 £/tCO₂. Importantly, the break-even carbon tax was found to be nearly equally sensitive to variations in both the sorbent, and CO₂ transport and storage cost. Interestingly, the break-even carbon tax would become zero, which implies that there is no cost associated to the CO₂ capture from the air, if the concentrated CO₂ stream can be sold at 7 £/tCO₂. With the CO₂ price of 10–30 £/tCO₂ being considered feasible for enhanced oil recovery [17], the CHP-DAC process can become economically competitive to other technologies that produce concentrated CO₂ streams. Importantly, if linked with enhanced oil recovery, the prices of the other products (heat, electricity, lime) can be reduced, increasing the process competitiveness in these markets.

Furthermore, the results of the sensitivity analysis presented in Fig. 6b indicated that the economic performance of the CHP-DAC system was highly sensitive to changes in the product prices, with the highest sensitivity observed for variation in the electricity price. This analysis also revealed that there exists a price for each product that results in a break-even carbon tax of zero. First, the break-even carbon

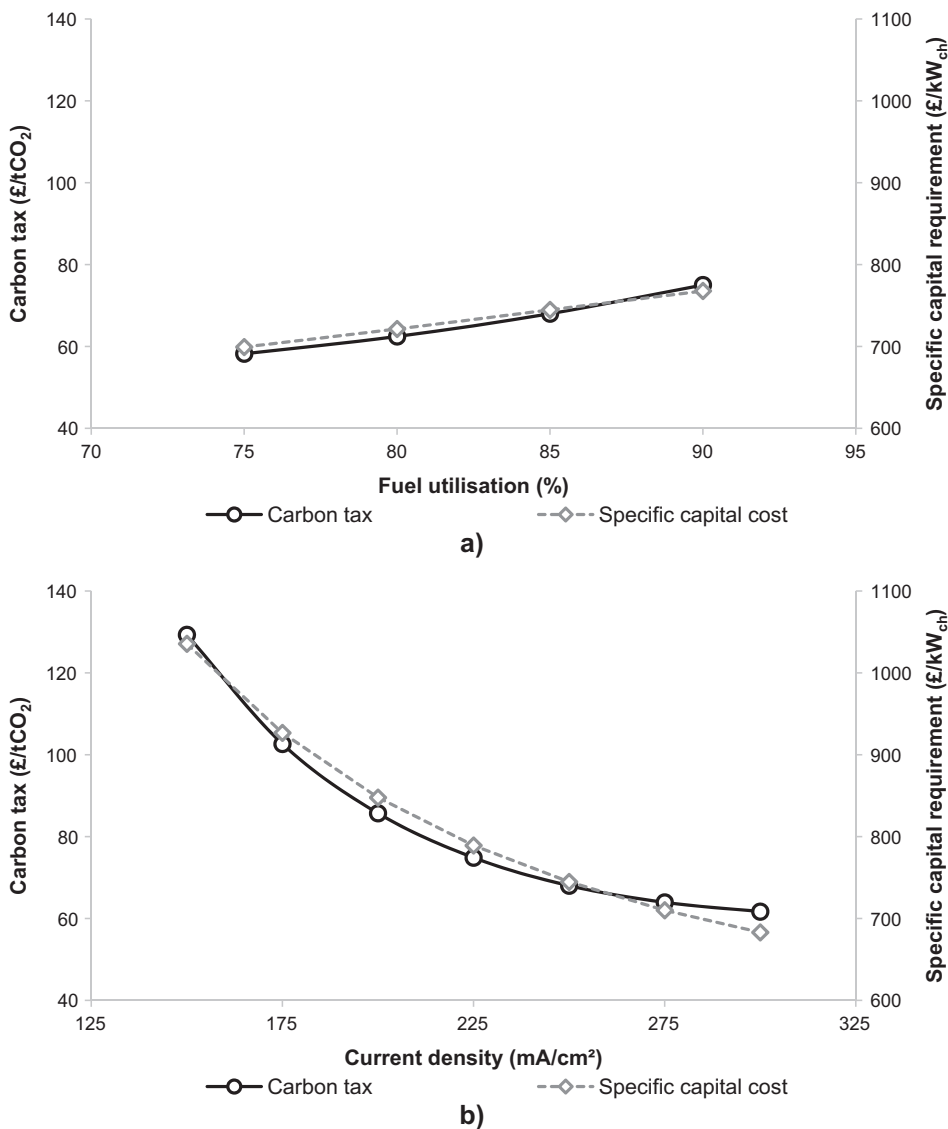


Fig. 5. Effect of (a) fuel utilisation of, and (b) current density in, the solid-oxide fuel cell on the economic performance of the CHP-DAC process.

tax became zero when the electricity price exceeded 60 £/tCO₂, which is comparable to the figures reported for fossil fuel power plants without (28–55 £/MW_{el}h) and with CO₂ capture (39–78 £/MW_{el}h) [21]. Such a figure for the price of electricity could be regarded as competitive with respect to other low-carbon power generation technologies, such as hydroelectric (48–63 £/MW_{el}h), biomass (65–93 £/MW_{el}h), nuclear (80–90 £/MW_{el}h), wind (35–170 £/MW_{el}h), and solar (53–290 £/MW_{el}h) power plants [46]. Second, similarly to the previous case, the break-even carbon tax became zero when the heat price exceeded 60 £/MW_{th}h. This figure is higher than the values considered for district heating of 34 £/MW_{th}h [47] to 49 £/MW_{th}h [48]. Nevertheless, with an increase in the electricity price from the initial figure of 40 £/MW_{el}h to 55 £/MW_{el}h, which is still competitive to other low-carbon power generation technologies, the break-even carbon tax became zero at a heat price of 37.5 £/MW_{th}h. Third, the break-even carbon tax became zero when the lime price exceeded 82 £/t, which is comparable to the market price of lime varying between 50–98 £/t [49,50]. Therefore, the CHP-DAC process can be considered as a competitive option that could replace the existing processes in a number of markets, as it has been shown to provide low-carbon electricity, heat, lime, and concentrated CO₂ stream at affordable prices.

A detailed sensitivity analysis was also performed to assess the influence of all economic assumptions on the break-even carbon tax

(Fig. 7). As has already been discussed above, this key economic performance indicator was mostly sensitive to variation in the total capital requirement, electricity price, and fuel cost. It needs to be noted that a $\pm 25\%$ change in the total capital requirement was shown to result in $\pm 54\%$ variation in the break-even carbon tax. This indicated that further reduction in the capital cost of the SOFC, which is the key contributor towards the total capital requirement, could bring nearly double the reduction in the break-even carbon tax. Moreover, the capacity factor and the sorbent conversion were shown to strongly affect the break-even carbon tax, as upon a 25% reduction in these parameters it increased by 54% and 33%, respectively. Therefore, operation of the CHP-DAC system at high capacity factors and sorbent conversions is required to achieve competitive economic performance. The detailed sensitivity analysis also indicated that degradation of the SOFC, which initially was considered to occur at a rate of 5 mV per 1000 h, would have an important effect on the break-even carbon tax. That economic performance parameter varied by -19% and $+16\%$ on a 25% reduction and increase in the SOFC degradation rate, respectively. Importantly, considering the project characteristics, only variation in the project interest rate and variable operating cost were shown to have a meaningful impact on the economic performance of the CHP-DAC process, while the remaining parameters resulted in break-even carbon tax variations of less than $\pm 5\%$. Importantly, a negligible sensitivity to

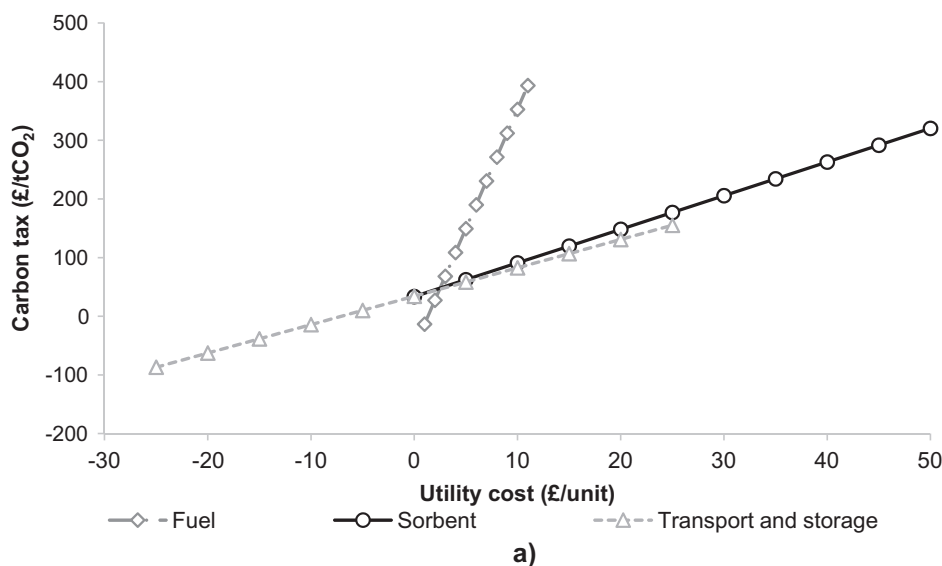
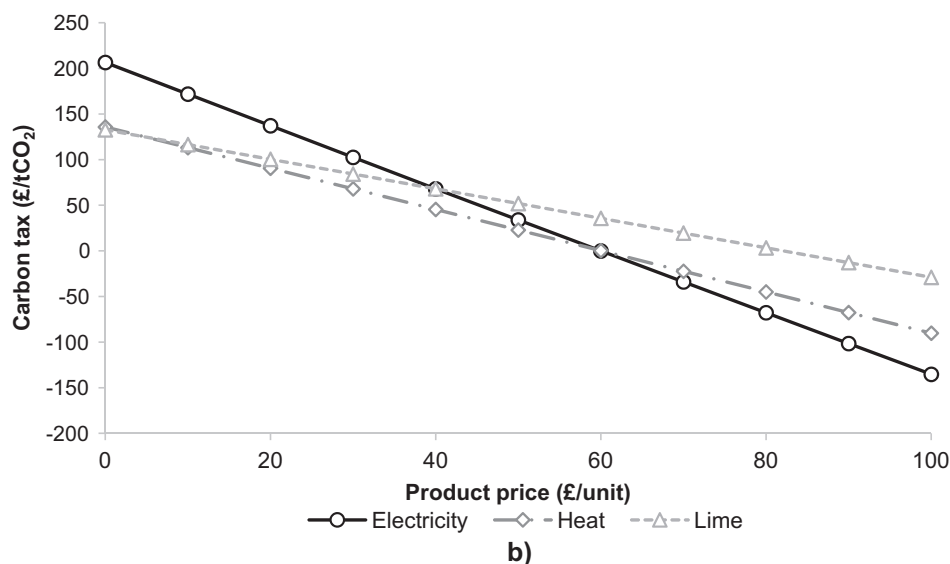


Fig. 6. Effect of utilities and product prices on the economic performance of the CHP-DAC process.



variation in the fraction of calcined product (Fig. 7), as well as comparable revenue from sales of 50% of the entire amount of the calcined product and using the remaining part for DAC indicate that the proposed process can break even if 100% of the calcined product is sold to other industries. Although in such case the process can be only considered as carbon neutral, it confirms that it can be a technically and economically feasible option in the current economic conditions. The proposed process can be described as negative-emission ready, because it will be capable of removing CO₂ directly from the air once the proper economic incentives are implemented.

In the economic analysis presented above, the minimum break-even cost associated with the negative CO₂ emissions that resulted in zero NPV after the lifetime of the project was estimated. It is pertinent, therefore, to evaluate other economic indicators that determine investment feasibility. The assessment of the CHP-DAC economic feasibility indicated that under the initial set of assumptions presented in Table 3, the profitability index is higher than 1 and the internal rate of return is higher than the assumed project interest rate for the carbon tax of 68 £/tCO₂ (Fig. 8). Yet, this figure corresponds to the break-even carbon tax estimated earlier and implies that the payback period for the project is 25 years. As a result the total revenue generated by the CHP-DAC process will only cover the total expenditure over its lifetime. Therefore, to generate profit, which is reflected in the profitability

index higher than 1 and the internal rate of return higher than the project interest rate, and to achieve the payback period of 6–10 years, which has been reported for other CHP systems [51,52], the carbon tax would need to fall within 110–150 £/tCO₂ (Fig. 8).

Nevertheless, the deterministic nature of the considered economic model may not provide the definitive actual performance of the CHP-DAC process, as the input parameters presented in Table 3 are associated with uncertainty. Therefore, to assess the effect of uncertainty in the input parameters on the economic feasibility of the considered process, the stochastic approach was implemented by considering the statistical distributions of the input parameters (Table 3). In addition, the carbon tax was assumed to have a uniform distribution with a range of 0–300 £/tCO₂. The results of the stochastic analysis (Fig. 9) indicated that the probability that the profitability index will be higher than 1 is 45%. Moreover, the figures for the 5th, 50th and 95th percentile were estimated to be -1.7, 0.8, and 3.7, respectively. This implies that there is high likelihood that the CHP-DAC process will be profitable if the carbon tax falls between 0 and 300 £/tCO₂. Therefore, the economic performance of this process can be judged to be superior to other DAC concepts, the break-even carbon tax for which was estimated to fall between 400 £/tCO₂ and 800 £/tCO₂ [15].

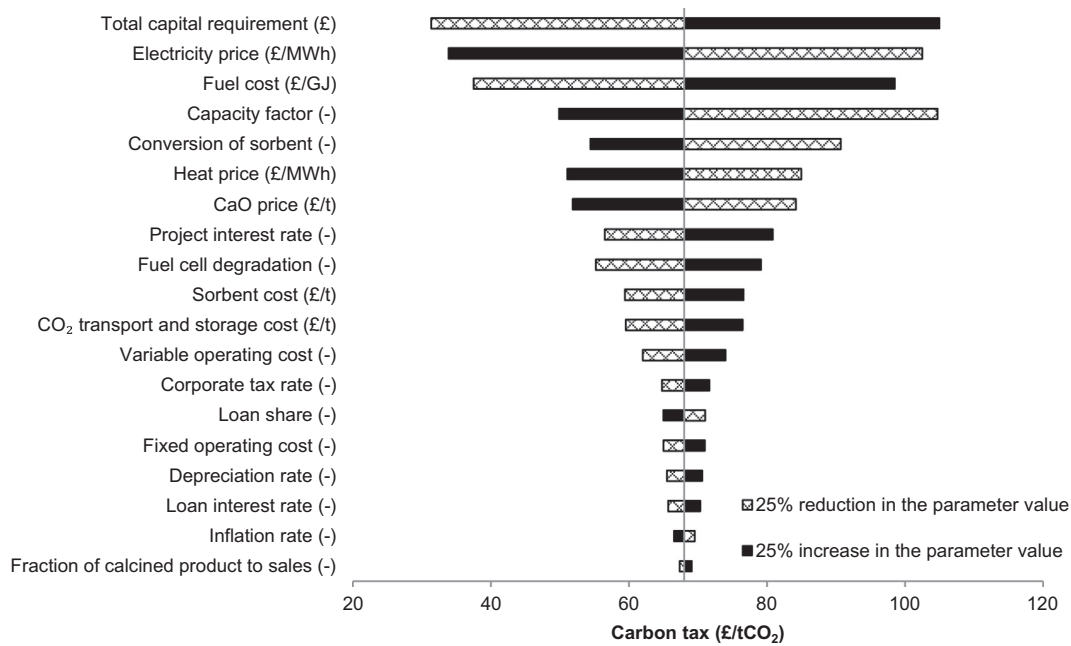


Fig. 7. Effect of variation of key economic parameters on the economic performance of the CHP-DAC process.

5. Conclusions

Decarbonisation of the energy and industrial sectors to meet the ambitious emission reduction targets set via the Paris Agreement requires a wide deployment of novel low-carbon technologies. These technologies should not only reduce the environmental burden of those sectors, but also ensure their competitiveness and affordability of the products. This study evaluated the feasibility of a novel polygeneration CHP-DAC process that combines power and heat generation with lime production for CO₂ removal from the air.

Analysis of the thermodynamic performance of the CHP-DAC process indicated that under initial design conditions it can achieve total efficiency and effective electric efficiency of 65%_{LHV} and 60%_{LHV}, respectively. Such performance was found to be comparable to other fuel-cell-based CHP systems. However, as opposed to these systems, the CHP-DAC process was shown to be capable of producing lime at a rate of 307.4 t/d. Having assumed that half of that figure was used for DAC (at 80% sorbent conversion in the long term), the amount of CO₂ removed from the air could reach 88.6 gCO₂/kW_{ch}. Such environmental performance would alleviate around 42% of the CO₂ emissions from the

conventional 440 MW_{el} NGCC without CO₂ capture system. Moreover, it could contribute to decarbonisation of the transportation industry.

Evaluation of the economic performance of the CHP-DAC process revealed that under the initial economic assumptions the specific capital requirement was 744.6 £/kW_{ch}. This was found to be higher than the corresponding figure for the NGCC with CO₂ capture (530–540 £/kW_{ch}), mostly due to the high capital requirement of the SOFC. Nevertheless, the CHP-DAC process exploits a number of revenue streams, including sales of power, heat, and lime, as well as potential revenue from the CO₂ removal from the air. Importantly, by spreading the total expenditure over a number of revenue streams, the product prices required for the CHP-DAC process to break even have been shown to be competitive compared to those reported for the existing standalone technologies, even if there was no revenue associated with CO₂ capture from the air (no carbon tax). Finally, the economic assessment revealed that the CHP-DAC process is an economically feasible technology, even under uncertainty in the market conditions.

This study has proved that novel polygeneration processes, such as the CHP-DAC, can bring a significant cost reduction in decarbonising the power and industrial sectors, achieving negative emissions, while

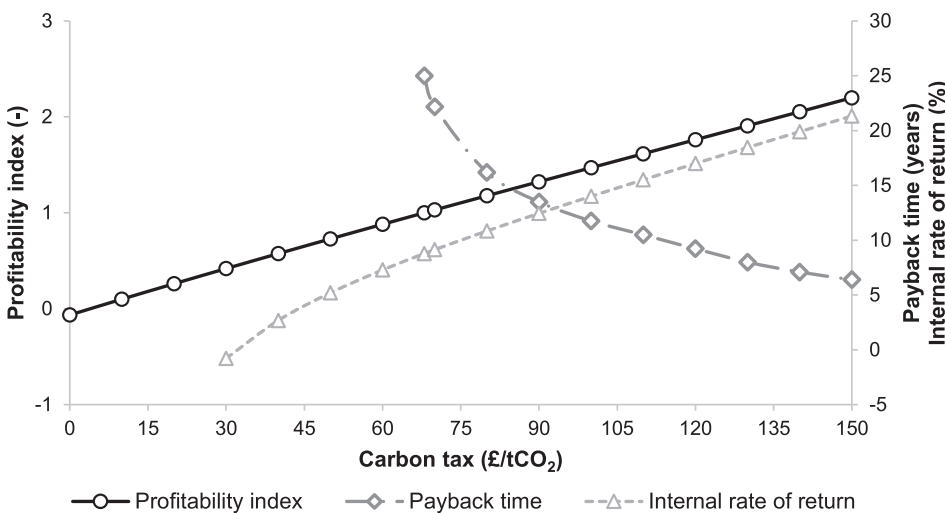


Fig. 8. Effect of carbon tax on profitability index, payback time, and internal rate of return.

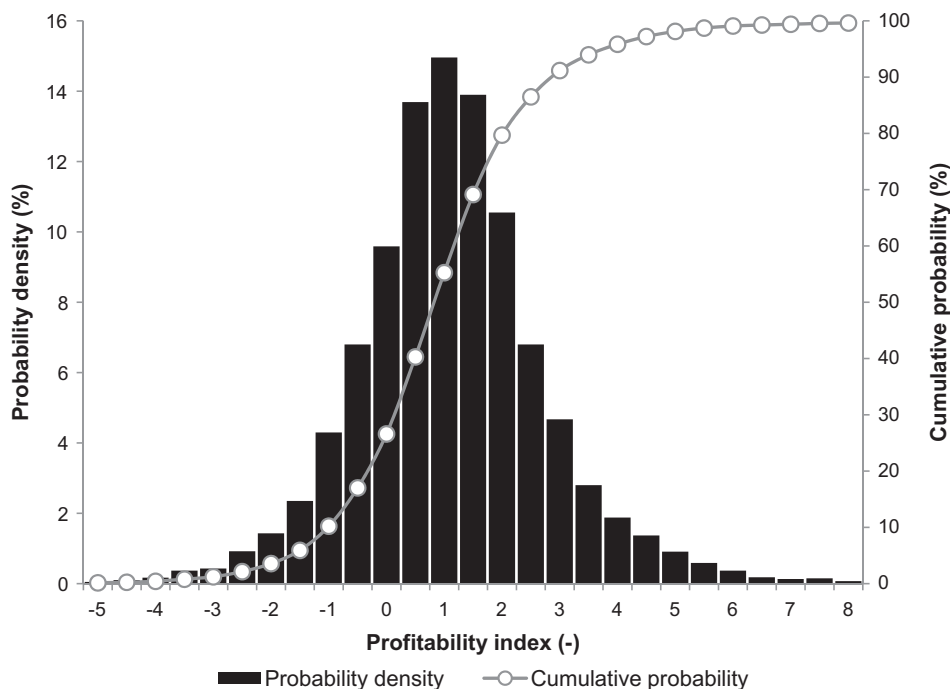


Fig. 9. Effect of uncertainty on profitability index.

maintaining their competitiveness and affordability. Importantly, even with no economic incentives for CO₂ removal from the air, the proposed process can be considered as a carbon-neutral polygeneration process for sustainable and affordable production of heat, power, and lime that is negative-emission ready. Also, exploration of potential integration links between the power, heat and industrial sectors will be essential in the future low-carbon energy scenarios and should account for connections with renewable energy sources and energy storage systems.

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Combined heat and power generation with lime production for direct air capture

Hanak, Dawid P.

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