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ORIGINAL ARTICLE

Analysis of production routes for silicon carbide using air as carbon source empowering negative emissions

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Abstract

A rapid defossilisation of the industry sector is required to stop further greenhouse gas emissions and to curb global warming. Additionally, to avoid irreversible consequences caused by climate change, the deployment of negative emission technologies is required to reduce the carbon dioxide (CO_2) concentration in Earth's atmosphere to a sustainable level. A novel approach to store gaseous CO_2 from direct air capture facilities in solid silicon carbide (SiC) is presented. A chain of established processes to produce SiC from renewable electricity and air is evaluated in terms of energy and mass balances. Furthermore, possible fields of SiC utilisation are considered. Electricity-based SiC (e-SiC) can serve the growing global market for technical ceramics and can possibly be used to tackle increasing construction sand shortages in the construction industry by partially substituting sand. Calculations of the levelised cost of carbon dioxide removal show that storing ambient CO_2 in solid SiC that can be subsequently sold on the world market can eventually create profit. In 2050, a net benefit of 259 €/tCO_2 or 631 €/tSiC can be realised if the SiC product is sold at the world market with additional carbon compensation. Therefore, the proposed SiC production chain might be able to challenge conventionally produced SiC, while empowering negative emissions. In 2050, the net CO_2 emission potential is limited to about 290 MtCO_2/a for technical ceramics, but may reach up to 13.6 GtCO_2/a for construction sand. Results show that e-SiC production is economically feasible for technical ceramics but not for construction sand without further process cost decrease. Alternative processes to produce e-SiC are described and evaluated. Future research opportunities are discussed.

Keywords Negative emissions · Silicon carbide · Carbon dioxide removal · Defossilisation · Industry · Power-to-X

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1 Introduction

Mitigating the effects of global warming requires urgent action to limit the atmospheric concentration of carbon dioxide (CO_2) to not exceed a global average temperature rise of 1.5°C as agreed upon in the Paris Agreement of 2015 (UNFCC 2015). To limit global warming to a level at which humanity will be able to adapt, a radical transformation to 100% renewable energy systems is needed and proven to be economically feasible (Bogdanov et al. 2021; Breyer et al. 2022b). To keep global warming at 1.5 °C or even below, future energy systems will require negative emission technologies (NETs) (Fuss et al. 2018) in addition to a rapid energy transition (Breyer et al. 2020). Proposed NETs such as direct air capture (DAC) with carbon capture and storage (DACCS) (Breyer et al. 2019; Chen and Tavoni 2013; Realmonte et al. 2019) or bioenergy with carbon capture and storage (BECCS) (Kemper 2015) capture gaseous CO_2 to store it on the long term. Carbon capture and utilisation (CCU) (Mertens et al., 2023) aims to utilise captured CO_2 as a renewable carbon source for fuels and chemicals (Bui et al. 2018; Galán-Martín et al. 2021; Galimova et al. 2022). While CCU can be realised with net-zero emissions (Bogdanov et al. 2021; Breyer et al. 2019), the re-emission of CO_2 at the end of the products lifetime hinders the approach to be net-negative (Hepburn et al. 2019). In contrast, chemically inert and long-term carbon capture and storage (CCS) can, in combination with a defossilised energy system, enable negative emissions needed for mitigating global warming (Gabrielli et al. 2020).

Currently, one possible method to store CO_2 is to sequester it in its gaseous phase in sub-surficial aquifers or other geological formations. However, this method bears the risk of potential leakage over the long storage duration that is required (Aminu et al. 2017). A major leakage event could bring significant risks for both humanity and ecosystems (Vinca et al. 2018). To avoid such risks, a method was investigated to store gaseous CO_2 in a solid-state product that shows a high combustion point as well as chemical inertness in order to provide options for safe and long-term storage of ambient CO_2 and to empower effective negative emissions. Silicon carbide (SiC) was identified to fulfil these criteria. The material can be used for power electronic applications or as a technical ceramic (Mukasyan 2017). SiC shows a high chemical inertness that makes it attractive for utilisation in chemical industry. Also, it has a decomposition temperature of around 2830°C (Guichelaar 1996). Today, SiC is mainly produced via the Acheson process (Guichelaar 1996; Mukasyan et al. 2013), where carbon black and silicon dioxide (SiO_2) are processed at elevated temperature above 1557°C to SiC (Guichelaar 1996). Whereas today, the utilised carbon black comes from fossil sources such as petroleum (Fyven 2022), or fossil methane (Boretti 2021), process routes that can substitute fossil carbon sources with ambient CO_2 can be identified. Other approaches avoiding fossil resources mainly focus on biomass as carbon source for the SiC production (Chiew and Cheong 2011; Thomas et al. 2021).

This study aims to present a novel production route using atmospheric CO_2 as the carbon source, thus enabling negative emissions, for which the mass and energy balances, as well as cost assumptions are provided. The results are suitable for including the novel power-to-SiC (PtSiC) option in future energy system modelling and assessment of the new NET option regarding its mitigation potential within the conventional NET portfolio.

2 Methods and data

Value chains of chemical processes to produce SiC from atmospheric CO_2 are identified to create an effective carbon sink in a material that shows outstanding resistance to wear off, as well as to chemical and thermal stress. Subsection 2.1 deals with the conventional SiC production. Subsection 2.2 introduces the integrated pyrolysis and Acheson (IPA) process to produce electricity-based silicon carbide (e-SiC). Subsection 2.3 describes an evaluation of IPA economics to produce e-SiC. In subsection 2.4, alternative processes are introduced for a possible substitution of sub-processes within the IPA value chain if the technology readiness level (TRL) increases. All energy and mass balances were either derived from theoretical chemical reaction equations or calculated from numbers given in literature. The data are normalised to 1 t of output products. In this study, electric heating for high-temperature levels and heat supply via heat pump for temperatures not higher than 100°C are assumed. Therefore, all heat required for the processes will be satisfied with electricity in the latter process chain models. The resulting energy balances for all the processes used in this study are shown in the Appendix (Table 3).

2.1 Conventional silicon carbide production

Currently, the majority of SiC is produced via the Acheson process (Fyven 2022; Guichelaar 1996; Mukasyan 2017). In fact, this process was first proposed by Edward Acheson as early as 1893 (Acheson 1895) and was initially meant to produce a crystal from the materials carbon and alumina (Guichelaar 1996). However, this process is still the main way to produce SiC (Fyven 2022). SiO_2 is reduced with carbon to synthesise the crude SiC, with carbon monoxide (CO) as a by-product as shown in Eq. (1) (Chiew and Cheong 2011). The input carbon black for the Acheson process is usually derived from fossil petroleum (Fyven 2022) or from fossil methane (Boretti 2021). The process is conducted at elevated temperatures around 1700–2500°C and is endothermic. The high-temperature level in the Acheson furnace is reached with electric heaters. The coke is placed together with SiO_2 in the Acheson furnace unit with the heating rod placed in the centre and a plastic cover to capture produced CO (Guichelaar 1996).

$$SiO_2(s) + 3\ C(s) + 618.5\ kJmol^{-1} \rightarrow SiC(s) + 2\ CO(g) \tag{1}$$

The theoretical energy requirement per t of SiC produced is 5.74 MWh (Guichelaar 1996). A realistic assumption is that the production of 1 t of SiC requires 6.5 MWh of energy (Guichelaar 1996). This energy is needed in the form of electricity to heat the arc furnace with an electric heating system (Guichelaar 1996). Usually, the feedstock utilised shows a SiO_2/C mass ratio of 1.7 (Guichelaar 1996). This is in line with the stoichiometric mass balance calculated from Eq. (1). About 22.5 wt% of carbon and SiO_2 input mass are reacted to SiC in a single Acheson furnace run (Guichelaar 1996).

2.2 Electricity-based silicon carbide production

Gaseous CO_2 can be captured from the atmosphere via low-temperature DAC (Fasihi et al. 2019). The required heat at a temperature level of 100°C for the regeneration of the solid sorbent can be supplied by a heat pump.

Electricity-based methane (e-methane), the feedstock for methane pyrolysis (Parkinson et al. 2021) to produce solid carbon, can be produced in a methanation process (Thema et al. 2019). Methanation is seen as an integral process of various power-to-gas (PtG) approaches (Götz et al. 2016; Peters et al. 2019; Sterner and Specht 2021). Therefore, its techno-economic specifications can be derived thoroughly. The theoretical reaction from gaseous CO_2 to methane is given in Eq. (2).

$$CO_2 + 4\,H_2 \rightarrow CH_4 + 2\,H_2O + 165\,kJmol^{-1} \tag{2}$$

The catalytic methanation reaction typically takes place in adiabatic fixed bed reactors at a pressure level of 1–100 bar and at temperatures ranging from 200 to 550°C (Götz et al. 2016). The process is exothermic. However, the practical reaction equation differs from theory. Since the conversion process is not ideal, some amount of CO_2 and hydrogen will remain in the product (Götz et al. 2016). Overall, the process requires an energy input of about 423 kWh_{el} of electricity for the methanation of CO_2 to 1 t of e-methane, according to DVGW (2013). This e-methane consists of 96 v% methane, 2 v% hydrogen and 2 v% CO_2. For simplification, it is assumed that for 1 t of methane, 2.86 t of CO_2 is reacted with 0.51 t of hydrogen. Therefore, the carbon conversion efficiency, i.e. the share of carbon converted, is assumed to be 100% for the methanation process. The required hydrogen for the methanation unit can be partly fed from methane pyrolysis (Boretti 2021; Parkinson et al. 2017), which is described in detail below. The remaining amount of hydrogen is produced via water electrolysis. Water electrolysis is a mature and commercialised process. There are various designs proposed that require different catalysts, electrolytes and temperature levels. In the present study, an alkaline water electrolyser is used (Fasihi and Breyer 2020).

Around 95% of global carbon black is produced from non-catalytic methane pyrolysis (Parkinson et al. 2019). The solid carbon powder can be used for several applications, such as rubber tire production, utilisation as catalyst or as structural material (Pérez et al. 2021). To achieve negative emissions and to produce carbon black for the SiC production, no fossil methane must be used. Methane pyrolysis attracts attention mainly because of the possibility to produce fossil methane-based hydrogen (H_2). However, some studies also acknowledge the idea of selling the side product carbon to reduce overall hydrogen production costs (Parkinson et al. 2019). The endothermic pyrolysis reaction to produce solid carbon and gaseous hydrogen from gaseous methane is given in Eq. (3) (Boretti 2021; Parkinson et al. 2018).

$$CH_4 + 74\,kJmol^{-1} \rightarrow 2\,H_2 + C \tag{3}$$

The pyrolysis can employ various catalysts such as carbon black itself (Boretti 2021). The pyrolysis reaction, according to the reaction equation, theoretically requires around 1285 kWh of heat at a temperature of 1000°C and at a pressure of 35 bar for the splitting of 1 t of methane (Parkinson et al. 2017). One t of carbon black and 0.3 t of hydrogen are pyrolysed from 1.3 t of methane (Boretti 2021). As described by Sánchez-Bastardo et al. (2020), unreacted methane can be looped back to the input methane. Therefore, no carbon losses and subsequently a carbon conversion efficiency of 100% are assumed for methane pyrolysis.

The process utilised to produce SiC from carbon black remains the Acheson process, since this is the only process identified at a high TRL and wide commercialisation. The conversion efficiency within an Acheson furnace mentioned in subsection 2.1 does not affect the modelling of the production of e-SiC. Since all unreacted input material is recycled and used for another run of the furnace (Guichelaar 1996), the mass balance for modelling the production of e-SiC does not have to be adjusted regarding the conversion efficiency. However, unreacted by-products increase the throughput and therefore energy demand of the intermediate processes methanation and methane pyrolysis (cf. Fig. 1). As shown by Sun et al. (2019), the carbon purity for SiC production is of lesser relevance, as SiC can be synthesised from low-grade educts.

The Acheson process produces CO as a by-product, which is a synthesis gas of the methanation and can be fed back to the respective process step. However, since the molar masses of CO_2 and CO differ, the CO fed back to the methanation reduced the net CO_2 demand and the methanation process is modelled in a simplified way, the CO must be converted to a CO_2 equivalent by applying Eq. (4).

$$m_{\mathrm{CO,CO2eq}} = m_{\mathrm{CO}} \cdot \frac{M_{\mathrm{CO}}}{M_{\mathrm{CO2}}} \tag{4}$$

wherein $m_{\mathrm{CO,CO2eq}}$ represents the mass of CO when accounted for as CO_2, m_{CO} is the mass of CO, M_{CO} is the molar mass of CO of 28 g/mol, and M_{CO2} is the molar mass of CO_2 of 44 g/mol.

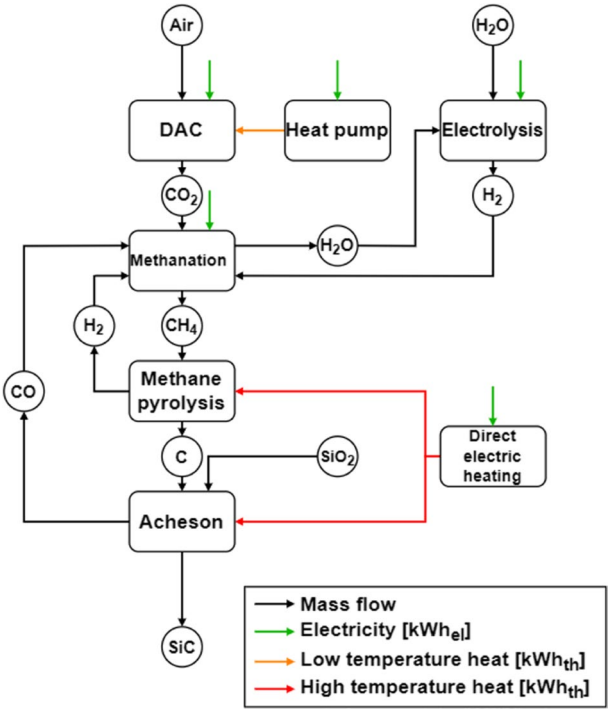

Fig. 1 Simplified schematic visualisation of the integrated e-SiC production route

2.3 Economics of electricity-based silicon carbide

There were no sufficiently reliable numbers available in the literature considering cost of processes for combustion synthesis from elements or by SiO_2 reduction (Mukasyan 2017) as well as for CO_2 electrolysis in molten lithium carbonate (Laasonen et al. 2022). Therefore, these processes were not evaluated in terms of costs. Only e-SiC production via methane pyrolysis could be fully evaluated regarding economic aspects: The schematic model of the e-SiC production chain based on air, water and electricity is shown in Fig. 1.

The levelised cost of carbon dioxide removal (*LCOCDR*) of this process chain is calculated according to Eq. (5):

$$\mathrm{LCOCDR} = \sum_{p}^{proc} \left(\mathrm{LCOP_p} \cdot m_\mathrm{p}\right) + H_\mathrm{LT} \cdot \mathrm{LCOH_{LT}} + \left(E_\mathrm{el} + H_\mathrm{HT}\right) \cdot \mathrm{cost_{el}} \tag{5}$$

wherein $LCOP_p$ is the levelised cost of each process p, m_p is the mass output produced from each process required to store 1 t of CO_2 in solid SiC, H_LT is the heat demand on a low-temperature level (max. 100°C), $LCOH_{LT}$ is the levelised cost of low-temperature heat, E_el is the electricity demand, H_HT is the high-temperature heat demand which is covered via direct electric heating, and finally, $cost_{el}$ describes the cost of electricity.

The *LCOP* of specific processes is calculated applying Eq. (6):

$$\mathrm{LCOP} = \frac{\left(\mathrm{capex} \cdot \left(\mathrm{crf} + \mathrm{opex_{fix}}\right)\right) \cdot \mathrm{capacity}}{\mathrm{out_p}} + \mathrm{opex_{var}} \tag{6}$$

wherein *capex* are the capital expenditures, $opex_{fix}$ are the fixed operational expenditures, $opex_{var}$ are the variable operational expenditures, and *crf* is the capital recovery factor. Process output out_p is defined in Eq. (7) including the annual capacity and availability factor τ that is set to 95% in this work.

$$\mathrm{out_p} = \mathrm{capacity} \cdot \tau \tag{7}$$

The *crf* is defined as in Eq. (8).

$$crf = \frac{WACC \cdot (1 + WACC)^N}{(1 + WACC)^N - 1} \tag{8}$$

The weighted average cost of capital *WACC* is assumed to 7% as a global average for all years.

LCOP for transformers with a given capex based on installed capacity or energy unit output are calculated with Eq. (9).

$$LCOP = \frac{capex \cdot \left(crf + opex_{fix}\right)}{FLH \cdot \tau} + opex_{var} \tag{9}$$

The $LCOH_{LT}$ is calculated using Eq. (10).

$$LCOH_{LT} = \frac{capex \cdot \left(crf + opex_{fix}\right)}{FLH \cdot \tau} + opex_{var} + \frac{cost_{el}}{COP} \tag{10}$$

The *COP* describes the coefficient of performance for the heat pump.

Table 1 Available economic input data for all processes used for modelling costs of carbon removed and SiC produced for 2030, 2040 and 2050

Process	Parameter	Unit	2030	Value 2040	2050	Reference
Acheson[1]	Production cost	€/tSiC	195	195	195	(Guichelaar 1996)
Methane pyrolysis	Capex	€/(tCB•a)	921.8	921.8	921.8	(Parkinson et al. 2018)
	$Opex_{fix}$	% of capex	5	5	5	
	Lifetime	Years	30	30	30	
	Capacity	tCB/a	272,000	272,000	272,000	
Methanation (incl. CO_2 compressor)	Oapex	€/$kW_{CH4,HHV}$	143	96	96	based on (Fasihi et al. 2017)
	$Opex_{fix}$	% of capex	4	4	4	
	$Opex_{var}$	€/$kWh_{CH4,HHV}$	0.0023	0.0023	0.0023	
	Lifetime	Years	30	30	30	
	Efficiency	%	78	78	78	
Alkaline electrolyser	Capex	€/$kW_{H2,HHV}$	417	241	181	(Fasihi and Breyer 2020)
	Capex	€/kW_{el}	547	304	221	
	$Opex_{fix}$	% of capex	3.5	3.5	3.5	
	$Opex_{var}$	€/$kWh_{H2,HHV}$	0.0012	0.0012	0.0012	
	Lifetime	years	30	30	30	
	Efficiency, HHV	%	76.2	79.2	82.1	
	Storage cost	€/$MW_{H2,LHV}$	5	5	5	
DAC	Capex	€/(tCO_2•a)	338	237	199	(Fasihi et al. 2019)
	$Opex_{fix}$	% of capex	4	4	4	
	Lifetime	Years	25	30	30	
	Capacity	tCO_2/a	360,000	360,000	360,000	
Heat pump	Capex	€/kW_{th}	590	554	530	(Fasihi et al. 2019)
	$Opex_{fix}$	% of capex	2	2	2	
	$Opex_{var}$	€/kWh_{th}	0.00170	0.00163	0.00160	
	Lifetime	Years	25	25	25	
	COP	-	3.26	3.41	3.51	

Table 1 (continued)

Process	Parameter	Unit	Value 2030	2040	2050	Reference
Electricity	Cost	€/MWh_{el}	20	15	10	(Fasihi et al. 2021)
SiC sales	Income	€/tSiC	833.3	833.3	833.3	(Businesswire 2022)
Construction sand sales[2]	Income	€/tSiC	15.8	32.6	67.2	(Statista 2022)
CO_2 pricing	Compensation	€/tCO_2	135	220	220	based on (IEA 2021)

[1] Adapted from Guichelaar with an assumed electricity price of 20 €/MWh (Guichelaar 1996)

[2] Price was estimated using the forecasted growth rate (Wrede 2019)

The *LCOP* was calculated for every sub-process except the Acheson process. Since no detailed economic input data for the Acheson process could be obtained from literature, a constant cost for the last step of e-SiC production was calculated from data provided in Guichelaar (1996). Input data of the economic assessment for the process chain are listed in Table 1.

CB carbon black

To provide another view on the results, a levelised cost of SiC production (*LCOSiC*) was calculated to evaluate the cost for each t of SiC produced from atmospheric CO_2. The *LCOSiC* was calculated as described by Eq. (11):

$$LCOSiC = LCOCDR \cdot \frac{m_{CO2,stored}}{m_{SiC,produced}} \tag{11}$$

wherein $m_{CO2,stored}$ is the amount of ambient CO_2 stored in SiC. The mass of produced SiC from gaseous CO_2 is $m_{SiC,produced}$.

Additionally, the potential for CO_2 storage in 1 t of SiC $CDR_{pot,SiC}$ is calculated using Eq. (12).

$$CDR_{pot,SiC} = \frac{1}{m_{SiC,produced,1tCO2}} \tag{12}$$

The economic evaluation of the proposed process chain is made for the years 2030, 2040 and 2050.

Furthermore, there is a globally rising challenge of increasing scarcity of construction sand. There are several technical norms stating a grain size distribution for construction sand. Generally, construction sand should consist of 49 wt% fine sand, 35 wt% medium sand and 14 wt% coarse sand as well as 2 wt% fine gravel (Elsner 2019). The grain size ranges from 0.063 to 2 mm (Elsner 2019). In terms of size, SiC can fulfil all the required grain sizes due to the large crude SiC particles produced in the Acheson process (Guichelaar 1996). Usually, theses large crude particles are crushed and milled to fulfil special properties needed for technical applications (Guichelaar 1996). This and the general angularity of SiC particles imply the general suitability of SiC as a construction sand substitute. Although this approach has yet to be validated in real projects, in the context of this research, the long-term CO_2 sequestration potential assessment is made based on the assumption of e-SiC substitution rates for sand of 3%, 15% and 50% in 2030, 2040 and 2050, respectively, reflecting high demand for construction sand and ambitious climate targets. Concrete demand until 2050 was calculated using the expected cement production in 2030, 2040 and 2050 (Farfan et al. 2019) as well as the typical cement concentration in concrete of around 10 wt% (PCA 2019). Concrete also consists of ca. 70 wt% aggregates, coarse and fine (PCA 2019). Considering the substitution of construction sand with e-SiC, it is assumed that 3 wt% in 2030, 15 wt% in 2040 and 50 wt% in 2050 of the aggregates in concrete can technically be substituted with e-SiC. The price for construction sand (CS) and gravel in 2021 was about 8.25 €/tCS (Statista 2022). It is expected to increase with a growth rate of 7.5% p.a. (Wrede 2019). Therefore, a construction sand price of 16 €/tCS in 2030, 33 €/tCS in 2040 and 67 €/tCS in 2050 is assumed.

2.4 Alternative processes

In general, pure carbon can be produced in various processes. For example, carbon black based on biochar can be produced via a pyrolysis process from different biomass feedstocks (Shalini et al. 2021). All information obtained about alternative processes is summarised in

Table 4 in the Appendix. Due to lack of detailed data for techno-economic modelling, the alternative processes presented in this subsection are not part of the assessment made in this study. However, in subsection 3.3, several of the abovementioned processes are discussed with regard to possible integration in the proposed process chain and the challenges and opportunities thereof.

There is growing interest in methods to produce solid carbon directly from CO_2 in an electrolysis process (Ren et al. 2017). However, most publications focus on gaseous or liquid products such as CO or ethene (C_2H_4) (Jouny et al. 2018). There are several approaches to split gaseous CO_2 into solid carbon and gaseous oxygen (O_2) in an electrocatalytic process. Most of the investigated reactors use molten lithium carbonate ($LiCO_3$) as an electrolyte (Laasonen et al. 2022; Licht et al. 2019). The reaction occurs at a temperature level of 750°C and is endothermic (Laasonen et al. 2022). In the follow-up reaction, the lithium oxide is synthesised with gaseous CO_2 to form lithium carbonate. The lithium carbonate synthesis also runs at a temperature level of 750°C but is exothermic, in contrast to the split reaction (Laasonen et al. 2022). The reaction from CO_2 to solid carbon and gaseous O_2 is endothermic and requires an overall temperature level of 750°C. An energy demand of 2494 kWh per t of CO_2 split can be estimated for the theoretic reaction. This aligns with 2 MWh per t of CO_2 if energy recovered from hot oxygen is accounted (Licht et al. 2019). This approach receives much attention in current research and is described as an economically feasible method (Licht et al. 2019). However, no information was found on conversion efficiencies of CO_2 electrolysis in molten lithium carbonate in literature.

There are further proposed approaches to produce solid carbon directly from gaseous CO_2. Esrafilzadeh et al. (2019) successfully showed the production of carbonaceous materials from CO_2 in an electrocatalytic reduction reaction. Liquid galinstan, an alloy of gallium (Ga), indium (In) and tin (Sn), was doped with elementary cerium (Ce), which increased the reactivity of the nanostructured catalyst. Carbon sheets showing nanostructures were produced in this experiment (Esrafilzadeh et al. 2019). Based on that work, there are several interesting follow-up works that focus on direct production of solid carbon from gaseous CO_2 (Zuraiqi et al. 2022). Also, Ye et al. (2023) studied the combined capture and storage of ambient CO_2 in solid carbon at near room temperature and atmospheric pressure using a liquid magnesium (Mg) and gallium (Ga) alloy. The use of relatively low-cost Mg can further facilitate the research in the field of direct CO_2 reduction (Ye et al. 2023).

Mukasyan et al. (2013) explain the self-propagating high-temperature synthesis (SHS), also referred to as combustion synthesis, as an alternative route to produce SiC (Mukasyan et al. 2013). This method is supposed to be more energy efficient than the Acheson process and can be conducted in two ways. The SiC can be produced from the elementary powders of silicon (Si) and carbon in a gasless combustion (Mukasyan 2017). The combustion synthesis reaction from elementary powders requires heat at a temperature level of 1600°C and is exothermic (Mukasyan 2017). However, to get the reaction started, an activation energy of around 5714 kWh_{th} of heat for the reaction of 1 t of carbon to SiC is required (Narayan et al. 1994). Also, because of the exothermic process characteristic, 4024 kWh_{th} of heat at a temperature of 1600°C must be removed from the reactor and can possibly be used as waste heat for other processes.

The combustion synthesis mentioned by Mukasyan et al. (2013) can also be conducted using the same educts as in the Acheson process. Additional magnesium (Mg) is added to the educts and magnesium oxide (MgO) is produced (Mukasyan 2017). Despite being an exothermic reaction, the specific activation energy for the combustion synthesis process must be provided (Narayan et al. 1994). The reaction occurs at a temperature level of 1726°C (Mukasyan 2017). From molecular weights, the difference in enthalpy and the

reaction equation, energy and mass balances for the combustion synthesis process with additional Mg can be estimated to 5714 kWh_{th} of heat for the reaction of 1 t of carbon. The combustion synthesis with added magnesium is exothermic, and 2890 kWh of heat at a temperature of 1726°C can be retrieved from the reactor for each t of carbon reacted.

Literature suggests several other ways to produce SiC such as carbothermal reduction, sol-gel methods or gas-phase reactions (Yang et al. 2009). Also, there are some approaches to produce SiC from biomass (Chiew and Cheong 2011), for instance experiments showed the successful sequestration of CO_2 prior stored in tobacco plants in plant-based SiC (Thomas et al. 2021). A review of SiC production from biomass mentions SiC from biomass waste (Chiew and Cheong 2011). It was decided to exclude these biomass-based routes to SiC in this study due to the relatively low TRL of these alternative approaches.

3 Results and discussion

The main production route evaluated in this research consists of a series of well established, readily available processes, i.e. the DAC, electrolysis, methane pyrolysis, Acheson, and methanation, which is proven to be able to work on a large scale (Thema et al. 2019). This process chain can also be referred to as IPA. This wording is adapted from a project aiming to produce carbon black with the abovementioned process (BMWK 2022). The overall mass and energy balances will be presented in the following subsection. In addition, the process costs will be evaluated normalised to 1 t of CO_2 stored in e-SiC as well as to 1 t of SiC produced from atmospheric CO_2. Furthermore, alternative processes such as CO_2 electrolysis and combustion synthesis are discussed in terms of their possible advantages.

3.1 Energy and mass balances

The generic model of the IPA for 2030 is visualised in Fig. 2, applying the specific energy and mass balance.

For each t of CO_2 removed from the atmosphere, 0.41 t of e-SiC can be produced. The overall process chain requires a total of 10.8 MWh_{el} to store a t of atmospheric CO_2 in solid SiC in 2030. The overall electricity demand decreases to 10.2 MWh_{el} and 9.9 MWh_{el} in 2040 and 2050, respectively. Details on the energy demand of each process step are listed in Table 2. The heat pump supplies 1500 kWh_{th} at 100°C for the DAC unit in 2030. The heat demand for DAC decreases to 1286 kWh_{th} in 2040 and 1102 kWh_{th} in 2050. The DAC plant requires 225 kWh_{el} to capture 1 t of CO_2 from the atmosphere in 2030. This electricity requirement decreases to 203 kWh_{el} and 182 kWh_{el} in 2040 and 2050, respectively. The methanation unit produces 0.49 t of methane and 1.11 t of water from 1.37 t of CO_2 as well as 0.25 t of hydrogen at an electricity input of 208 kWh_{el} for all years. Produced methane is then split in the methane pyrolysis reactor to 0.12 t of hydrogen, which is fed back to the methanation unit, and 0.37 t of carbon black. The energy demand of methane pyrolysis is 631 kWh_{th} in 2030, 2040 and 2050, which is fully covered by electricity via direct electric heating. Subsequently, the carbon black is reacted together with 0.62 t of SiO_2 to produce 0.41 t of e-SiC and 0.57 t of CO that is fed back to the methanation unit. The CO is used to substitute CO_2 and accounted for as 0.37 tCO_2e according to Eq. 4. The energy-intensive Acheson process requires 2667 kWh_{th}/tCO_2 in 2030, 2040 and 2050, which is also covered by electricity via direct electric heating.

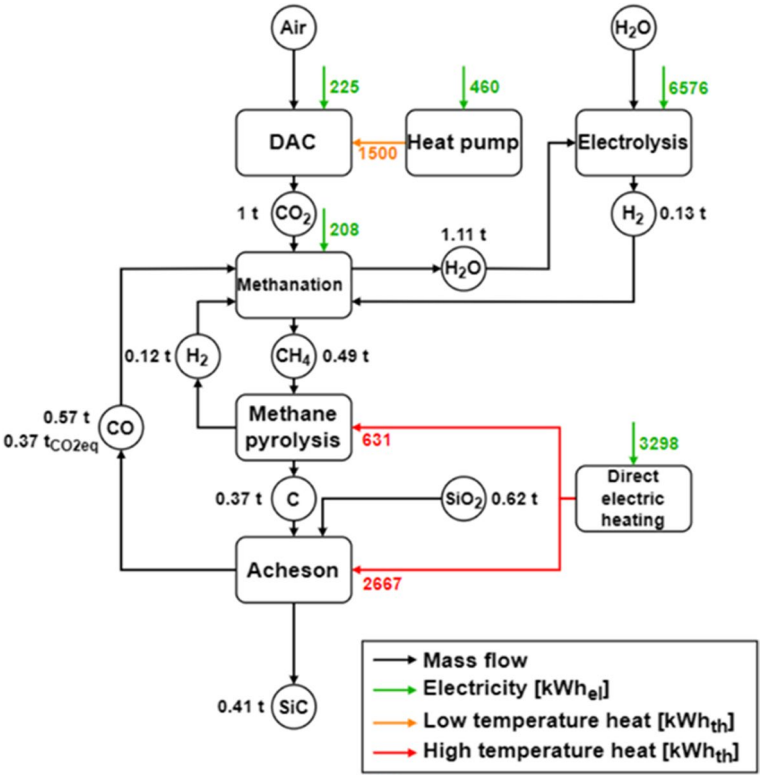

Fig. 2 Simplified process model of the IPA process with concrete energy and mass balances for the year 2030

Table 2 Resulting energy demand for storing atmospheric CO_2 in SiC. Low-temperature heat is provided via electric heat pumps and high-temperature heat with direct electric heating (cf. Table 1)

Process	Energy type	Unit	2030	2040	2050
Direct air capture	Electricity	kWh_{el}/tCO_2 stored	225	203	182
	Heat[a]	kWh_{th}/tCO_2 stored	1500	1286	1102
Electrolysis	Electricity	kWh_{el}/tCO_2 stored	6576	6327	6103
Methanation	Electricity	kWh_{el}/tCO_2 stored	208	208	208
Methane pyrolysis	Electricity	kWh_{el}/tCO_2 stored	631	631	631
Acheson	Heat[b]	kWh_{th}/tCO_2 stored	2667	2667	2667
Total	Electricity	kWh_{el}/tCO_2 stored	7640	7369	7124
	Heat[c]	kWh_{th}/tCO_2 stored	4167	3953	3769
Total electrified	Electricity	kWh_{el}/tCO_2 stored	10,766	10,209	9923

[a]Low temperature

[b]High temperature

[c]Low and high temperature combined

From another perspective, 2.44 t of ambient CO_2 are stored in 1 t of solid SiC. The overall energy demand to produce 1 t of e-SiC via IPA is 26.2 MWh_{el}/tSiC in 2030, 24.9 MWh_{el}/tSiC in 2040 and 24.2 MWh_{el}/tSiC in 2050. Low-temperature heat demand for the DAC unit per t of e-SiC decreases from 3.7 MWh_{th}/tSiC in 2030 to 3.1 MWh_{th}/tSiC in 2040 and 2.7 MWh_{th}/tSiC in 2050. Additionally, the DAC unit requires 548 kWh_{el}/tSiC in 2030, 495 kWh_{el}/tSiC in 2040 and 444 kWh_{el}/tSiC in 2050. For 1 t of e-SiC produced, the methanation unit must produce 1.2 t of methane, equivalent to 18.5 MWh_{HHV}, with 2.7 t of water as by-product from 3.3 t of CO_2 and 0.6 t of hydrogen. For the methanation, 506 kWh_{el}/tSiC are required in 2030, 2040 and 2050. From the methane, 0.9 t of carbon black as well as 0.3 t of hydrogen are produced in the methane pyrolysis unit. The hydrogen is fed back to the methanation unit. The high-temperature heat demand that is covered with direct electric heating is 1538 kWh_{el}/tSiC in 2030, 2040 and 2050. The solid carbon is reacted with 1.5 $tSiO_2$/tSiC to SiC, and 1.4 tCO/tSiC is fed back to the methanation unit to partially substitute CO_2. Applying Eq. 4, the CO is accounted for as 0.9 tCO_2e. The Acheson process requires a total of 6500 kWh_{th}/tSiC.

3.2 Cost structure for electricity-based silicon carbide production

To assess the economics of the proposed production route presented in this research, the *LCOCDR* was calculated using the obtained energy and mass balance as well as the numbers summarised in Table 1. In Fig. 3, the *LCOCDR* in- and excluding energy cost is presented.

The e-SiC cost in- and excluding energy cost decreases from 2030 to 2040 and 2050 by 20% and 34% including and 14% and 17% excluding energy cost, respectively. The development of *capex*, *opex*, lifetime and energy demand of specific processes favour this trend. The energy cost share is 50%, 45% and 36% in 2030, 2040 and 2050, respectively. This development can be explained by the smaller energy demand of the overall process chain and by the decline in electricity cost due to further improved economics of solar photovoltaics (Vartiainen et al. 2020; Victoria et al. 2021) and wind power in hybrid power plants (Fasihi and Breyer 2020). Therefore, the cost of energy decreases more significantly compared to the capital and operational expenditures of the production plants.

The Acheson process is the most cost intensive step in this production chain if energy cost is excluded. Interestingly, if energy cost is considered, water electrolysis is the most expensive process in 2030. This is due to the high energy demand of the electrolyser to produce the required amount of hydrogen. The energy cost share in 2030 for the electrolyser alone is 79%. Decreasing electricity cost and efficiency gain of the electrolyser accompanied by a significant reduction in *capex* will reduce the cost for electrolysis to a level below the Acheson process in 2040 and 2050. The energy cost share for the water electrolysis process alone is 81% in 2040 and 77% in 2050.

As shown in Fig. 3, the *LCOCDR* including energy in 2050 is 303 €/tCO_2 if no SiC sales are considered. The *LCOCDR* including energy cost and SiC sales at a global price of 883 €/tSiC, that was normalised to 1 t of CO_2 stored, is presented in Fig. 4. As it can be seen, if only the estimated SiC price is accounted for, e-SiC produced via the proposed process chain will be able to generate an economic benefit only in 2050. Storing 1 t of CO_2 in e-SiC to be sold on the world market at the proposed price creates a net profit of 39 €/tCO_2 (95 €/tSiC). The SiC price does not cover the cost for storing 1 t of CO_2 in e-SiC in 2030 and 2040. However, if a carbon compensation of 135 €/tCO_2, 220 €/tCO_2 and 220 €/tCO_2 for 2030, 2040 and 2050, respectively, is counted in, IPA e-SiC production creates

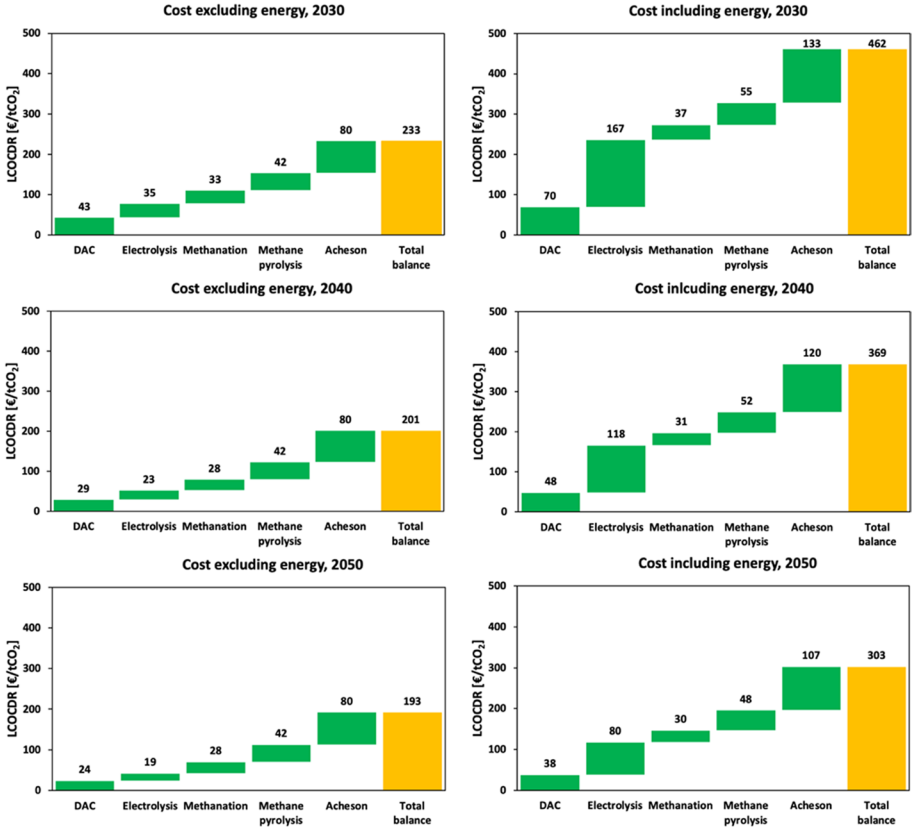

Fig. 3 LCOCDR of e-SiC production excluding (left) and including (right) cost for energy for the years 2030 (top), 2040 (centre) and 2050 (bottom)

economic benefits from 2030 onwards. A profit of 15 €/tCO_2 (37 €/tSiC) in 2030, 193 €/tCO_2 (471 €/tSiC) in 2040 and 259 €/tCO_2 (631 €/tSiC) in 2050 can be realised by storing ambient CO_2 in e-SiC if the final product is sold and a carbon compensation is claimed.

There is a significant uncertainty regarding future SiC demand and market prices. There are various variables, such as commercialisation of SiC in electronics, as well as CO_2 pricing of the fossil energy intensive carbon black production affecting these values. Taking profitability as the main condition to NET deployment, the produced e-SiC will be able to be sold if market prices for SiC will be at least as high as assumed in this study. For a compound annual growth rate of 16.8% p.a. for the global SiC market in the upcoming years, this would correlate with a flux carbon dioxide removal potential of 289.4 $MtCO_2$/a by 2050.

e-SiC might as well be utilised for substituting construction sand required for concrete. However, the *LCOCDR* for storing atmospheric CO_2 in solid SiC and selling the SiC as construction sand substitute is still not attractive in 2050 even if a carbon compensation of 220 €/tCO_2 is applied. However, the residual amount of 55 €/tCO_2 indicates that a CO_2 pricing of less than 300 €/tCO_2 may be sufficient to open a huge potential CO_2 storage for long-term and safe sequestration. Nevertheless, this CO_2 storage option

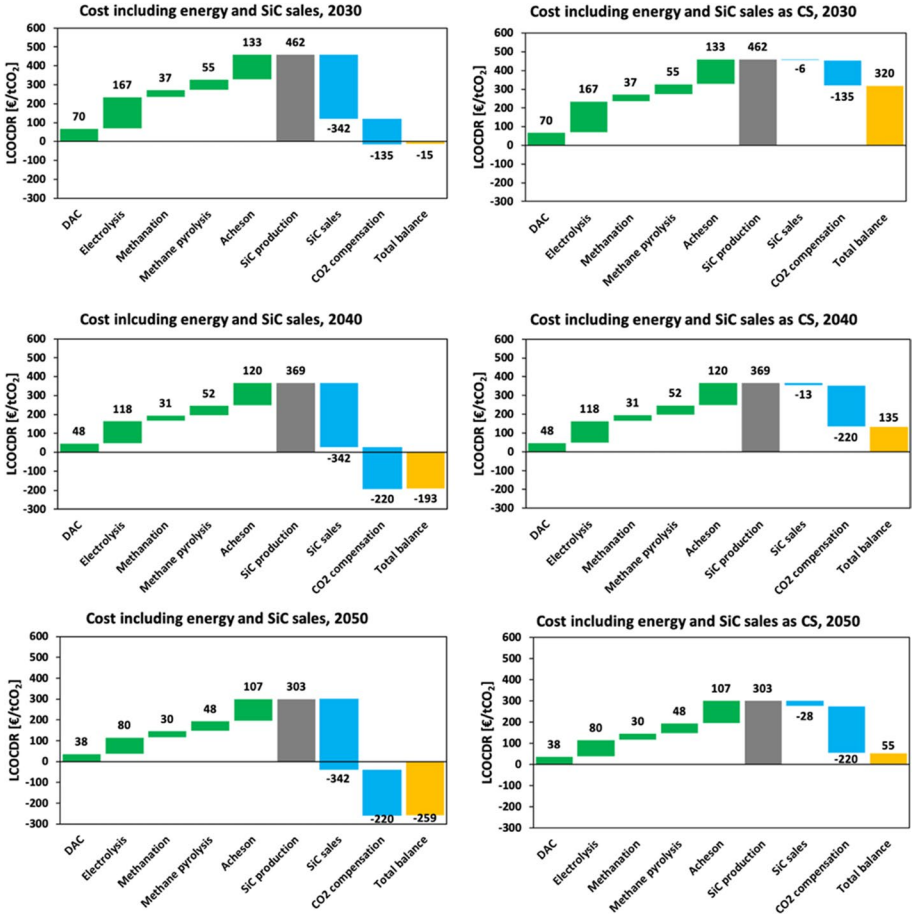

Fig. 4 LCOCDR of e-SiC production with energy cost including SiC sales (left) and to substitute construction sand (right) and CO_2 pricing for 2030, 2040 and 2050. Abbreviation: CS, construction sand

does not seem economically viable unless significant cost reductions in production processes or a significant increase in construction sand price occur in the future. A possible negative CO_2 emission potential in construction sand used for concrete production of 1.3 $GtCO_2$/a in 2030, 5.3 $GtCO_2$/a in 2040 and 13.6 $GtCO_2$/a in 2050 can be estimated at given market values and substitution rates. Further evaluation of these assumptions will be necessary to reduce the uncertainties related to the assumptions made.

Since the proposed production chain might be considered NET option from a climate change mitigation perspective and from a SiC material perspective, the results are also presented as *LCOSiC* considering SiC sales and CO_2 pricing, as shown in Fig. 5.

The CO_2 pricing assumed for 2030, 2040 and 2050 was normalised to 1 t of e-SiC produced and results in a deductible CO_2 pricing equivalent of 329 €/tSiC in 2030, 536 €/tSiC in 2040 and 536 €/tSiC in 2050. The possible economic benefit per t of e-SiC produced from atmospheric CO_2 if current SiC sales prices and future CO_2 pricing is applied is 37 €/tSiC in 2030, 471 €/tSiC in 2040 and 631 €/tSiC in 2050. If produced SiC is used to substitute construction sand and sold for the respective sales price and

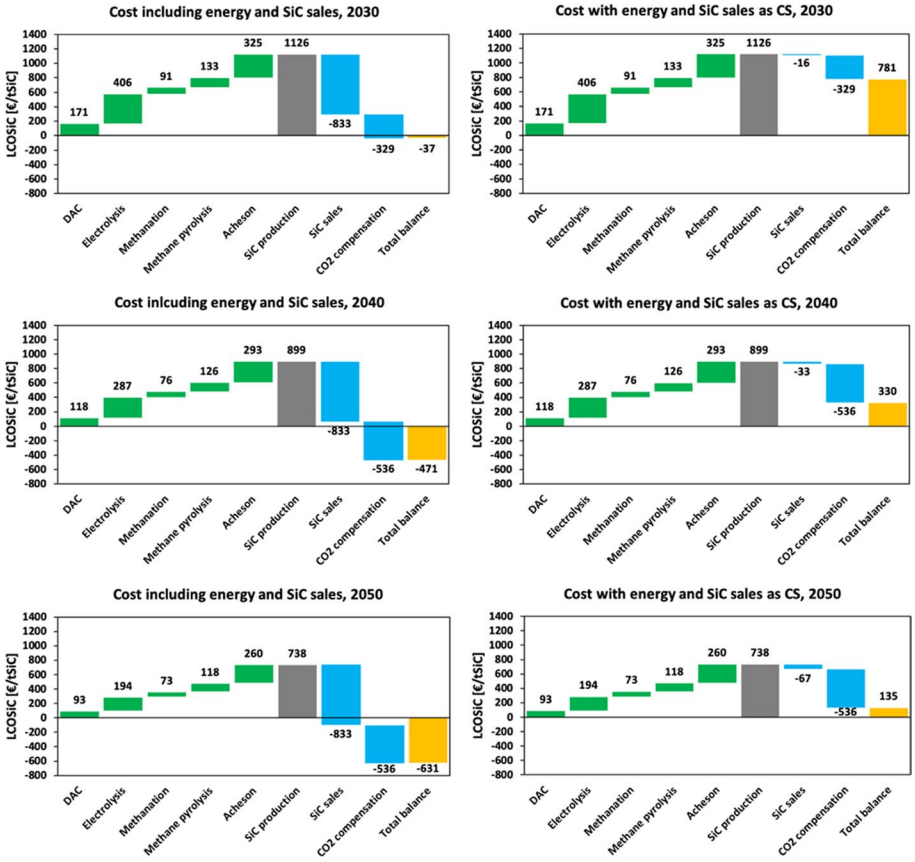

Fig. 5 LCOSiC for e-SiC production with energy cost including SiC sales (left) and to substitute construction sand (right) and CO_2 pricing for 2030, 2040 and 2050

future CO_2 pricing is considered, the *LCOSiC* is 781 €/tSiC in 2030, 330 €/tSiC in 2040 and 135 €/tSiC in 2050.

3.3 Discussion and research outlook

As described above, alternative processes, especially for the substitution of the costly Acheson process, increasingly attract attention. Additionally, process substitution would solve the problem of the low molar carbon efficiency of the Acheson process, since no CO is produced during combustion synthesis. Improved carbon efficiency would increase the net throughput of carbon from atmospheric CO_2 to e-SiC. However, the combustion synthesis would also bring some drawbacks, especially regarding input materials. The combustion synthesis using elementary carbon and silicon requires pure silicon that needs refining. The other possible alternative to the Acheson process is combustion synthesis from carbon, SiO_2 and pure magnesium (Aminu et al. 2017).

Although the future supply of sustainable biomass will be limited (Creutzig et al. 2015) and bio-based energy is desired by various sectors (Reid et al. 2020), biomethane

and biochar are potential alternative carbon sources to e-methane and carbon black from integrated pyrolysis, respectively. Biomethane can be obtained at cost as low as 464 €/t (33.7 €/$MWh_{th,LHV}$ for an energy density of e-methane of 13.75 $kWh_{th,LHV}$/kg) (Bose et al. 2022). The static techno-economic framework used in this work results in cost for e-methane production including renewable energy of 557.5 €/t (40.5 €/$MWh_{th,LHV}$), 401.5 €/t (29.2 €/$MWh_{th,LHV}$) and 300.9 €/t (21.9 €/$MWh_{th,LHV}$) in 2030, 2040 and 2050, respectively. Therefore, using biomethane could reduce the cost of CO_2 storage in e-SiC by about 9.9% in 2030 but would increase the cost of CO_2 storage by 8.3% and 26.4% in 2040 and 2050, respectively. Similarly, biochar can potentially substitute the carbon black produced in the integrated pyrolysis. The production cost of biochar is generally estimated to be on the order of 1000 €/t, and one study specifies the cost of biochar from date palm biomass as 883 €/t (assuming a long-term exchange rate of 1.2 USD/€) (Shahen et al. 2022). While the cost of biochar is subjected to uncertainty in, e.g. the cost of biomass feedstock, the cost are comparable to the cost of carbon black produced of about 892.0 €/t in 2030 as derived in this study. The potential reduction of the total cost of CO_2 storage is therefore negligible at about 0.7% in 2030, while the cost would increase by 20.7% and 42.6% in 2040 and 2050, respectively, if the cost of biochar is assumed to remain constant. There is significant uncertainty in the estimation of future cost of biomass-based products. Therefore, future developments in this domain should be monitored to assess the potential use of biomass-based products for e-SiC production.

Furthermore, CO_2 electrolysis might be an interesting option for future carbon black production. The stoichiometric reaction equation implies an energy demand of 2494 kWh_{el}/tCO_2 split into solid carbon and gaseous CO_2. Licht et al. (2019) claim that an energy demand of 2 MWh is required to process one t of gaseous CO_2 into solid carbon (Licht et al. 2019). However, no specific cost numbers regarding CO_2 electrolysis to solid carbon in molten lithium salt are provided, which implies a relatively low TRL. However, the potential of this technology should be emphasised. For e-SiC production, CO_2 electrolysis could possibly substitute water electrolysis, methanation and methane pyrolysis. Therefore, it would concur with an electricity demand of 7414 kWh_{el} as for 2030. As mentioned above, CO_2 electrolysis might require about 2 MWh per t of CO_2 split. Also, CO_2 electrolysis cost would concur with 110 €/tCO_2 for production cost via water electrolysis, methanation and methane pyrolysis excluding energy cost. Therefore, if large-scale CO_2 electrolysis in molten lithium carbonate will be technically feasible in the future, the process will be very interesting to include in the process chain producing e-SiC from atmospheric CO_2. Also, water electrolysis will become increasingly more established as hydrogen will play a key role for hydrogen-to-X processes in the defossilisation of hard-to-abate energy sectors, as an integral part of the arising Power-to-X Economy (Breyer et al. 2022a). For an electricity-based production of hydrogen powered by 100% renewable electricity, only the electrolysis efficiency limits the overall process efficiency, as one advantage of renewable energy sources is a 100% conversion efficiency from primary energy to electricity (Kraan et al. 2019, Keiner et al. 2023). However, a massive rollout of solar PV and electrolysers might help to increase these efficiencies further by accelerated research and development activities. Also, novel approaches such as direct air electrolysis proposed and studied by Guo et al. (2022) discuss the decreasing cost and energy demand of hydrogen production with theoretic solar-to-hydrogen efficiencies of up to 32%, while highest realised efficiencies are around 20% (Wang 2021), compared to about 15% for the separated solar PV plus electrolyser route. Direct air electrolysis could also overcome the necessity of freshwater supply of conventional electrolysers by utilising the air's humidity and, therefore, enable

hydrogen production in arid and semi-arid locations with best solar PV potentials (Guo 2022).

Even though practical deployment must show feasibility, SiC production via IPA is a sequence of established standard processes. Therefore, storing atmospheric CO_2 in solid SiC can be a valuable option for safe and long-term storage. However, negative CO_2 emissions in Gt-scale remain unlikely to be realised in the form of e-SiC with the presented cost of the process chain hardly being profitable by 2050. Nevertheless, the amount of CO_2 that is required to fulfil the global demand for SiC via these production routes might be an interesting approach for the future. Therefore, a potential market of up to 3.3 MtSiC in 2027 (Businesswire 2022) could enable a flux carbon removal potential of up to 8.1 $MtCO_2$/a. With an expected compound annual growth rate of 16.8% p.a. (Businesswire 2022) for the years to come, a possible carbon removal potential of up to 289.4 $MtCO_2$/a could be enabled in 2050. However, considering the second option of construction sand substitution with 50% e-SiC, a negative emission potential of 13.6 $GtCO_2$/a can be enabled by 2050. While the CDR potential for SiC in its conventional application areas is lower than the CDR potential of afforestation and reforestation, BECCS, biochar, enhanced weathering, DACCS, ocean fertilisation and soil carbon sequestration at 0.5–5 $GtCO_2$/a (Fuss et al. 2018), widening the application area of e-SiC to the substitution of construction sand could cover about 65% of the total 21 $GtCO_2$/a of carbon removal requirement estimated for mid-century by Fuss et al. (2018). Since the conventional SiC production is energy and cost-intensive, the phase-in of the production routes presented in this study to replace fossil sources for carbon black production seems feasible. This can potentially lower production cost and therefore may result in faster growing SiC markets. e-SiC furthermore offers the combination of CCS and CCU, while both concepts should normally be strictly separated (Mertens et al. 2023, Bruhn et al. 2016). A similar concept to e-SiC is the production of electricity-based carbon fibres (e-CF), with an estimated CDR potential of 0.7 $GtCO_2$/a by 2050 (Keiner et al. 2024). Therefore, both approaches are situated within the CCUS nexus combining the storage and utilisation of captured atmospheric CO_2. While dedicated CCU will most probably be necessary to defossilise hard-to-abate energy sectors in a 100% renewable energy system, it offers no long-term storage of atmospheric CO_2 (Galimova et al. 2022, Mertens et al. 2023). In contrast to liquid or gaseous products of CCU approaches, SiC cannot be combusted and therefore can be seen as a permanent carbon sink without a carbon cycle as it is present in CCU applications. However, the proposed approach is strongly interlinked with other CCU approaches via DAC, water electrolysis, methanation and methane pyrolysis being also applied for producing different energy carriers such as e-fuels or e-hydrogen (Mertens et al. 2023, Boretti et al. 2021). These common processes can reduce cost of CCU, CCS and e-SiC production as well, by providing a common basis for technology learning.

Further research and development for processes such as SiC production via combustion synthesis and Acheson process are required to enable large-scale rollout. Also, technological specifications must be made to ensure the actual viabilities of the processes. As an example, the necessary carbon purity for SiC production in the proposed process chain must be determined, even though current work showed SiC synthesis from low-grade educts (Sun et al. 2019).

In addition to the global demand in SiC for various technical applications, in particular ceramics and semiconductors, other sectors could use e-SiC to replace crucial materials. In particular, the ever-increasing demand for construction sand in the civil engineering sector draws increasingly more attention. Sand from deserts is not suitable due to the round shape of the sand grain (WWF 2022). In contrast, river and coastal sand is typically very well

suited for concrete production. The global scarcity of construction sand brought up a phenomenon called sand robbery (WWF 2022). Because crude SiC from the Acheson process shows grain sizes large enough to cover the whole range needed (Guichelaar 1996) and the microstructure seems to be suitable and porous enough, the idea of storing e-SiC produced from atmospheric CO_2 in concrete is proposed. The cost calculated in this study shows that this approach is not economically viable from today's perspective, but close, as a CO_2 pricing of 300 €/tCO_2 would be sufficient in 2050. However, the implementation of alternative processes such as combustion synthesis to produce SiC or CO_2 electrolysis to split gaseous CO_2 and produce solid carbon might give this approach the required boost. This research aims to advance research and discussion on how new energy-industry-CDR systems and integrated modelling of the latter can promote climate change mitigation (Breyer et al. 2022b).

The large-scale deployment of NETs required for future CDR applications like DACCS or BECCS still brings drawbacks regarding the safe and long-term sequestration of captured CO_2. The production of e-SiC from air and renewable electricity might be one possible solution to this challenge.

4 Conclusion

Defossilisation of the industry sector plays a crucial role in mitigating global warming. In this study, a process chain to produce solid SiC from gaseous CO_2 captured from the atmosphere to empower negative CO_2 emissions with safe long-term storage was presented.

The integrated pyrolysis and Acheson process are a value chain consisting of established processes linked to store atmospheric CO_2 in solid SiC. A total of 10.8 MWh_{el} in 2030, 10.2 MWh_{el} in 2040 and 9.9 MWh_{el} in 2050 is required to store 1 t of atmospheric CO_2 in solid SiC. The *LCOCDR* of producing electricity-based SiC was estimated to be a net loss of 120 €/tCO_2 in 2030, 27 €/tCO_2 in 2040 and a net profit of 39 €/tCO_2 in 2050, if the produced SiC is sold on the SiC world market, without factoring in the value of permanently sequestered CO_2. Other applications for electricity-based SiC such as construction sand substitution were discussed. Since construction sand is becoming an increasingly scarce resource, the idea to utilise electricity-based SiC as a construction sand substitute was elaborated. However, the calculated net cost including the value of the output material shows this approach is not economically viable. The *LCOCDR* of the integrated pyrolysis and Acheson process with subsequent SiC utilisation for construction sand is 455 €/tCO_2 in 2030, 355 €/tCO_2 in 2040 and 275 €/tCO_2 in 2050, without considering income from permanent CO_2 sequestering.

The production of 1 t of electricity-based SiC requires a total of 26.2 MWh_{el} in 2030, 24.9 MWh_{el} in 2040 and 24.2 MWh_{el} in 2050. The *LCOSiC* if produced SiC that is sold at the world market is profitable with of 37 €/tSiC in 2030, 471 €/tSiC in 2040 and 631 €/tCO_2 in 2050 if income form CO_2 pricing is accounted for.

Future research opportunities were identified. Alternative processes that can possibly lower the energy demand as well as the overall production cost were presented and described. Also, additional future applications of electricity-based SiC were discussed. Electricity-based SiC can be considered an attractive production option that can enable safe and long-term negative CO_2 emissions. Electricity-based SiC contributes to defossilising the industry sector while simultaneously acting as a long-term and safe carbon sink.

Appendix

Table 3 Mass and energy balances used for IPA process modelling

Process	Reference material	Input material	Input value	Output material	Output value	Electricity demand	LT heat demand	HT heat demand	HT heat generation	Reference
Unit	-	-	t/t_{out}	-	t/t_{out}	kWh_{el}/t_{out}	kWh_{th}/t_{out}	kWh_{th}/t_{out}	kWh_{th}/t_{out}	
DAC	CO_2	Air	n/a	CO_2	1	225/203/182[1]	1500/1286/1102[1]			(Fasihi et al. 2019)
Methanation incl. CO_2 compressor	CH_4	CO_2	2.78	CH_4	1	422.7				(Fasihi and Breyer 2020)
		Hydrogen	0.51	H_2O	2.27					
Methane pyrolysis	CB	CH_4	1.33	CB	1			1713		(Parkinson et al. 2017)
				Hydrogen	0.33					
Acheson	SiC	CB	0.90	SiC	1			6500		(Guichelaar 1996)
		SiO_2	1.50	CO	1.40					

[1]As 2030/2040/2050 values

CB carbon black

Table 4 Alternative processes to the IPA process chain

Process	Reference material	Input material	Input value	Output material	Output value	Electricity demand	LT heat demand	HT heat demand	HT heat generation	Reference
Unit	-	-	t/t_{out}	-	t/t_{out}	kWh_{el}/t_{out}	kWh_{th}/t_{out}	kWh_{th}/t_{out}	kWh_{th}/t_{out}	
CO_2 electrolysis	Carbon	CO_2	3.66	Carbon	1	9134[1]				(Laasonen et al. 2022)
				Oxygen	2.66					
Combustion synthesis—elementary Si	SiC	Carbon	0.30	SiC	1			1710	1204	(Mukasyan et al. 2013; Narayan et al. 1994)
		Silicon	0.70							
Combustion synthesis—elementary Mg	SiC	Carbon	0.30	SiC	1			1710	865	(Mukasyan et al. 2013; Narayan et al. 1994)
		SiO_2	1.50	MgO	2.01					
		Magnesium	1.21							

[1] Partly needed for direct electric heating

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Author contributions AM: conceptualisation, methodology, investigation, resources, writing—original draft, writing—review and editing and visualisation; DK: conceptualisation, methodology, investigation, resources and writing—review and editing; TG: validation and writing—review and editing and CB: conceptualisation, methodology, validation, writing—review and editing and supervision

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Data availability Not applicable.

Declarations

Competing interests The authors declare no competing interests.

References

Acheson EG (1895) Production of artificial crystalline carbonaceous materials: US Patent Reissue (No. 11,473)

Aminu MD, Nabavi SA, Rochelle CA, Manovic V (2017) A review of developments in carbon dioxide storage. Appl Energy 208:1389–1419. https://doi.org/10.1016/j.apenergy.2017.09.015

Bogdanov D, Ram M, Aghahosseini A, Gulagi A, Oyewo AS, Child M, Caldera U, Sadovskaia K, Farfan J, Barbosa LD, Fasihi M (2021) Low-cost renewable electricity as the key driver of the global energy transition towards sustainability. Energy 227:120467. https://doi.org/10.1016/j.energy.2021.120467

Boretti A (2021) Concentrated solar energy-driven carbon black catalytic thermal decomposition of methane. Int J Energy Res 45:21497–21508. https://doi.org/10.1002/er.7142

Bose A, O'Shea R, Lin R, Long A, Rajendran K, Wall D, De S, Murphy JD (2022) The marginal abatement cost of co-producing biomethane, food and biofertiliser in a circular economy system. Renew Sustain Energy Rev 169:112946. https://doi.org/10.1016/j.rser.2022.112946

Breyer C, Bogdanov D, Khalili S, Keiner D (2020) Solar photovoltaics in 100% renewable energy systems. In: Meyers RA (ed) Encyclopedia of sustainability science and technology. Springer, New York, New York, NY, pp 1–30

Breyer C, Bogdanov D, Ram M, Khalili S, Vartiainen E, Moser D, Medina ER, Masson G, Aghahosseini A, Mensah TNO, Lopez G, Schmela M, Rossi R, Hemetsberger W, Jäger-Waldau A (2022a) Reflecting the energy transition from a European perspective and in the global context – relevance of solar photovoltaics benchmarking two ambitious scenarios. Prog Photovolt Res Appl 1-27. https://doi.org/10.1002/pip.3659

Breyer C, Fasihi M, Bajamundi C, Creutzig F (2019) Direct air capture of CO_2: a key technology for ambitious climate change mitigation. Joule 3:2053–2057. https://doi.org/10.1016/j.joule.2019.08.010

Breyer C, Khalili S, Bogdanov D, Ram M, Oyewo AS, Aghahosseini A, Gulagi A, Solomon AA, Keiner D, Lopez G, Ostergaard PA, Lund H, Mathiesen BV, Jacobson MZ, Victoria M, Teske S, Pregger T,

Fthenakis V, Raugei M et al (2022b) On the history and future of 100% renewable energy systems research. IEEE Access 10:78176–78218. https://doi.org/10.1109/ACCESS.2022.3193402

Bruhn T, Naims H, Olfe-Kräutlein B (2016) Separating the debate on CO_2 utilisation from carbon capture and storage. Environ Sci Policy 60:38–43. https://doi.org/10.1016/j.envsci.2016.03.001

Bui M, Adjiman CS, Bardow A, Anthony EJ, Boston A, Brown S, Fennell PS, Fuss S, Galindo A, Hackett LA, Hallett JP, Herzog HJ, Jackson G, Kemper J, Krevor S, Maitland GC, Matuszewski M, Metcalfe IS, Petit C et al (2018) Carbon capture and storage (CCS): the way forward. Energ Environ Sci 11:1062–1176. https://doi.org/10.1039/c7ee02342a

Bundesministerium für Wirtschaft und Klimaschutz (BMWK) (2022) Energie von morgen Wie Forschung und Förderung erfolgreich zur Energiewende beitragen, Berlin

Businesswire (2022) Global Silicon Carbide Market Size, Trends & Growth Opportunity Report 2021-2027. https://www.businesswire.com/news/home/20220207005566/en/Global-Silicon-Carbide-Market-Size-Trends-Growth-Opportunity-Report-2021-2027%2D%2D-ResearchAndMarkets.com. Accessed 27 April 2022

Chen C, Tavoni M (2013) Direct air capture of CO_2 and climate stabilization: a model based assessment. Clim Change 118:59–72. https://doi.org/10.1007/s10584-013-0714-7

Chiew YL, Cheong KY (2011) A review on the synthesis of SiC from plant-based biomasses. Mater Sci Eng: B 176:951–964. https://doi.org/10.1016/j.mseb.2011.05.037

Creutzig F, Ravindranath NH, Berndes G, Bolwig S, Bright R, Cherubini F, Chum H, Corbera E, Delucchi M, Faaij A, Fargione J, Haberl H, Heath G, Lucon O, Plevin R, Popp A, Robledo-Abad C, Rose S, Smith P et al (2015) Bioenergy and climate change mitigation: an assessment. GCB Bioenergy 7:916–944. https://doi.org/10.1111/gcbb.12205

Deutscher Verein des Gas- und Wasserfaches e.V. (DVGW) (2013) Entwicklung von modularen Konzepten zur Erzeugung, Speicherung und Einspeisung von Wasserstoff und Methan ins Erdgasnetz. Bonn.

Elsner H (2019) BGR untersucht Sand aus der Sahara und von der Arabischen Halbinsel: Zu fein – Wüstensand kein hochwertiger Baurohstoff. https://www.bgr.bund.de/DE/Gemeinsames/Oeffentlichkeitsarbeit/Pressemitteilungen/BGR/bgr-2019-08-06_wuestensand_%20baurohstoff.html?nn=1557798. Accessed 27 April 2022

Esrafilzadeh D, Zavabeti A, Jalili R, Atkin P, Choi J, Carey BJ, Brkljača R, O'Mullane AP, Dickey MD, Officer DL, MacFarlane DR, Daeneke T, Kalantar-Zadeh K (2019) Room temperature CO_2 reduction to solid carbon species on liquid metals featuring atomically thin ceria interfaces. Nat Commun 10:865. https://doi.org/10.1038/s41467-019-08824-8

Farfan J, Fasihi M, Breyer C (2019) Trends in the global cement industry and opportunities for long-term sustainable CCU potential for Power-to-X. J Clean Prod 217:821–835. https://doi.org/10.1016/j.jclepro.2019.01.226

Fasihi M, Bogdanov D, Breyer C (2017) Long-term hydrocarbon trade options for the Maghreb region and Europe—renewable energy based synthetic fuels for a net zero emissions world. Sustainability 9:306. https://doi.org/10.3390/su9020306

Fasihi M, Breyer C (2020) Baseload electricity and hydrogen supply based on hybrid PV-wind power plants. J Clean Prod 243:118466. https://doi.org/10.1016/j.jclepro.2019.118466

Fasihi M, Efimova O, Breyer C (2019) Techno-economic assessment of CO_2 direct air capture plants. J Clean Prod 224:957–980. https://doi.org/10.1016/j.jclepro.2019.03.086

Fasihi M, Weiss R, Savolainen J, Breyer C (2021) Global potential of green ammonia based on hybrid PV-wind power plants. Appl Energy 294:116170. https://doi.org/10.1016/j.apenergy.2020.116170

Fuss S, Lamb WF, Callaghan MW, Hilaire J, Creutzig F, Amann T, Beringer T, de Oliveira Garcia W, Hartmann J, Khanna T, Luderer G (2018) Negative emissions—part 2: costs, potentials and side effects. Environ Res Lett 13:63002. https://doi.org/10.1088/1748-9326/aabf9f

Fyven (2022) Silicon carbide production process. https://www.fiven.com/world-of-silicon-carbide/sic-production-process/. Accessed 27 April 2022

Gabrielli P, Gazzani M, Mazzotti M (2020) The role of carbon capture and utilization, carbon capture and storage, and biomass to enable a net-zero-CO_2 emissions chemical industry. Ind Eng Chem Res 59:7033–7045. https://doi.org/10.1021/acs.iecr.9b06579

Galán-Martín Á, Tulus V, Díaz I, Pozo C, Pérez-Ramírez J, Guillén-Gosálbez G (2021) Sustainability footprints of a renewable carbon transition for the petrochemical sector within planetary boundaries. One Earth 4:565–583. https://doi.org/10.1016/j.oneear.2021.04.001

Galimova T, Bogdanov D, Fasihi M, Gulagi A, Ram M, Karjunen H, Mensah TNO, Khalili S, Breyer C (2022) Global demand analysis for carbon dioxide as raw material from key industrial sources and direct air capture to produce renewable electricity-based fuels and chemicals. J Clean Prod 373:133920. https://doi.org/10.1016/j.jclepro.2022.133920

Götz M, Lefebvre J, Mörs F, McDaniel Koch A, Graf F, Bajohr S, Reimert R, Kolb T (2016) Renewable power-to-gas: a technological and economic review. Renew Energy 85:1371–1390. https://doi.org/10.1016/j.renene.2015.07.066

Guichelaar PJ (1996) Acheson Process. In: Weimer AW (ed) Carbide, nitride and boride materials synthesis and processing. Springer, Netherlands, Dordrecht, pp 115–129

Guo J, Zhang Y, Zavabeti A, Chen K, Guo Y, Hu G, Fan X, Li GK (2022) Hydrogen production from the air. Nat Commun 13:5046. https://doi.org/10.1038/s41467-022-32652-y

Hepburn C, Adlen E, Beddington J, Carter EA, Fuss S, Mac Dowell N, Minx JC, Smith P, Williams CK (2019) The technological and economic prospects for CO_2 utilization and removal. Nature 575:87–97. https://doi.org/10.1038/s41586-019-1681-6

International Energy Agency (IEA) (2021) World Energy Outlook 2021, Paris

Jouny M, Luc W, Jiao F (2018) General techno-economic analysis of CO_2 electrolysis systems. Ind Eng Chem Res 57:2165–2177. https://doi.org/10.1021/acs.iecr.7b03514

Keiner D, Gulagi A, Breyer C (2023) Energy demand estimation using a pre-processing macro-economic modelling tool for 21st century transition analyses. Energy 272:127199. https://doi.org/10.1016/j.energy.2023.127199

Keiner D, Mühlbauer A, Lopez G, Breyer C (2024) Techno-economic assessment of atmospheric CO_2-based carbon fibre production enabling negative emissions. Mitig Adapt Strateg Glob Chang. In press. https://doi.org/10.1007/s11027-023-10090-5

Kemper J (2015) Biomass and carbon dioxide capture and storage: a review. Int J Greenhouse Gas Control 40:401–430. https://doi.org/10.1016/j.ijggc.2015.06.012

Kraan O, Chappin E, Kramer GJ, Nikolic I (2019) The influence of the energy transition on the significance of key energy metrics. Renew Sustain Energy Rev 111:215–223. https://doi.org/10.1016/j.rser.2019.04.032

Laasonen E, Ruuskanen V, Niemelä M, Koiranen T, Ahola J (2022) Insights into carbon production by CO_2 reduction in molten salt electrolysis in coaxial-type reactor. J Environ Chem Eng 10:106933. https://doi.org/10.1016/j.jece.2021.106933

Licht S, Liu X, Licht G, Wang X, Swesi A, Chan Y (2019) Amplified CO_2 reduction of greenhouse gas emissions with C2CNT carbon nanotube composites. Mater Today Sustain 6:100023. https://doi.org/10.1016/j.mtsust.2019.100023

Mertens J, Breyer C, Aming K, Bardow A, Belmans R, Dibenedetto A, Erkman S, Griekoven J, Léonard G, Nizou S, Pant D, Reis-Machado AS, Styring P, Vente J, Webber M, Sapart CJ (2023) Carbon capture and utilization: more than hiding CO_2 for some time. Joule 7:442–449. https://doi.org/10.1016/j.joule.2023.01.005

Mukasyan AS (2017) Silicon carbide. In: Concise encyclopedia of self-propagating high-temperature synthesis. Elsevier, pp 336–338

Mukasyan AS, Lin Y-C, Rogachev AS, Moskovskikh DO (2013) Direct combustion synthesis of silicon carbide nanopowder from the elements. J Am Ceram Soc 96:111–117. https://doi.org/10.1111/jace.12107

Narayan J, Raghunathan R, Chowdhury R, Jagannadham K (1994) Mechanism of combustion synthesis of silicon carbide. J Appl Phys 75:7252–7257. https://doi.org/10.1063/1.356660

Parkinson B, Balcombe P, Speirs JF, Hawkes AD, Hellgardt K (2019) Levelized cost of CO_2 mitigation from hydrogen production routes. Energ Environ Sci 12:19–40. https://doi.org/10.1039/C8EE02079E

Parkinson B, Matthews JW, McConnaughy TB, Upham DC, McFarland EW (2017) Techno-economic analysis of methane pyrolysis in molten metals: decarbonizing natural gas. Chem Eng Technol 40:1022–1030. https://doi.org/10.1002/ceat.201600414

Parkinson B, Patzschke CF, Nikolis D, Raman S, Dankworth DC, Hellgardt K (2021) Methane pyrolysis in monovalent alkali halide salts: kinetics and pyrolytic carbon properties. Int J Hydrogen Energy 46:6225–6238. https://doi.org/10.1016/j.ijhydene.2020.11.150

Parkinson B, Tabatabaei M, Upham DC, Ballinger B, Greig C, Smart S, McFarland E (2018) Hydrogen production using methane: techno-economics of decarbonizing fuels and chemicals. Int J Hydrogen Energy 43:2540–2555. https://doi.org/10.1016/j.ijhydene.2017.12.081

Pérez L, José B, Jiménez MJA, Bhardwaj R, Goetheer E, van Sint AM, Gallucci F (2021) Methane pyrolysis in a molten gallium bubble column reactor for sustainable hydrogen production: proof of concept & techno-economic assessment. Int J Hydrogen Energy 46:4917–4935. https://doi.org/10.1016/j.ijhydene.2020.11.079

Peters R, Baltruweit M, Grube T, Samsun RC, Stolten D (2019) A techno economic analysis of the power to gas route. J CO2 Util 34:616–634. https://doi.org/10.1016/j.jcou.2019.07.009

Portland Cement Association (PCA) (2019) How concrete is made. https://www.cement.org/cement-concrete/how-concrete-is-made. Accessed 27 April 2022

Realmonte G, Drouet L, Gambhir A, Glynn J, Hawkes A, Köberle AC, Tavoni M (2019) An inter-model assessment of the role of direct air capture in deep mitigation pathways. Nat Commun 10:3277. https://doi.org/10.1038/s41467-019-10842-5

Reid WV, Ali MK, Field CB (2020) The future of bioenergy. Glob Chang Biol 26:274–286. https://doi.org/10.1111/gcb.14883

Ren J, Johnson M, Singhal R, Licht S (2017) Transformation of the greenhouse gas CO_2 by molten electrolysis into a wide controlled selection of carbon nanotubes. J CO2 Util 18:335–344. https://doi.org/10.1016/j.jcou.2017.02.005

Sánchez-Bastardo N, Schlögl R, Ruland H (2020) Methane pyrolysis for CO2-free H2 production: a green process to overcome renewable energies unsteadiness. Chem Ing Tech 92:1596–1609. https://doi.org/10.1002/cite.202000029

Shaheen J, Fseha YH, Sizirici B (2022) Performance, life cycle assessment, and economic comparison between date palm waste biochar and activated carbon derived from woody biomass. Heliyon 8:e12388. https://doi.org/10.1016/j.heliyon.2022.e12388

Shalini S, Palanivelu K, Ramachandran A, Raghavan V (2021) Biochar from biomass waste as a renewable carbon material for climate change mitigation in reducing greenhouse gas emissions—a review. Biomass Conv Bioref 11:2247–2267. https://doi.org/10.1007/s13399-020-00604-5

Statista (2022) Average price of construction sand and gravel in the U.S. from 2010 to 2021. https://www.statista.com/statistics/219381/sand-and-gravel-prices-in-theus/#:~:text=In%20the%20United%20States%2C%20the,per%20metric%20ton%20in%202021.. Accessed 27 April 2022

Sterner M, Specht M (2021) Power-to-gas and power-to-X—the history and results of developing a new storage concept. Energies 14:6594. https://doi.org/10.3390/en14206594

Sun K, Wang T, Chen Z, Lu W, He X, Gong W, Tang M, Liu F, Huang Z, Tang J, Chien T, Tan G, Fan M (2019) Clean and low-cost synthesis of high purity beta-silicon carbide with carbon fiber production residual and a sandstone. J Clean Prod 238:117875. https://doi.org/10.1016/j.jclepro.2019.117875

Thema M, Bauer F, Sterner M (2019) Power-to-gas: electrolysis and methanation status review. Renew Sustain Energy Rev 112:775–787. https://doi.org/10.1016/j.rser.2019.06.030

Thomas ST, Shin Y, La Clair JJ, Noel JP (2021) Plant-based CO_2 drawdown and storage as SiC. RSC Adv 11:15512–15518. https://doi.org/10.1039/D1RA00954K

United Nations Framework Convention on Climate Change (UNFCC) (2015) Report of the conference of the parties on its twenty-first session FCCC(CP/2015/Add.1.Pariss

Vartiainen E, Masson G, Breyer C, Moser D, Román Medina E (2020) Impact of weighted average cost of capital, capital expenditure, and other parameters on future utility-scale PV levelised cost of electricity. Prog Photovolt Res Appl 28:439–453. https://doi.org/10.1002/pip.3189

Victoria M, Haegel N, Peters IM, Sinton R, Jäger-Waldau A, Del Cañizo C, Breyer C, Stocks M, Blakers A, Kaizuka I, Komoto K, Smets A (2021) Solar photovoltaics is ready to power a sustainable future. Joule 5:1041–1056. https://doi.org/10.1016/j.joule.2021.03.005

Vinca A, Emmerling J, Tavoni M (2018) Bearing the cost of stored carbon leakage. Front Energy Res 6. https://doi.org/10.3389/fenrg.2018.00040

Wang Y, Sharma A, Duong T, Arandiyan H, Zhao T, Zhang D, Su Z, Garbrecht M, Beck FJ, Karuturi S, Zhao C, Cathpole K (2021) Direct solar hydrogen generation at 20% efficiency using low-cost materials. Adv Energy Mater 11:2101053. https://doi.org/10.1002/aenm.202101053

Wrede I (2019) Not enough sand for construction industry despite abundance? https://p.dw.com/p/3L2Lu. Accessed 27 April 2022

Yang Y, Lin Z-M, Li J-T (2009) Synthesis of SiC by silicon and carbon combustion in air. J Eur Ceram Soc 29:175–180. https://doi.org/10.1016/j.jeurceramsoc.2008.06.013

Ye L, Syed N, Wang D, Guo J, Yang J, Buston J, Singh R, Alivand MS, Li GK, Zavabeti A (2023) Low-temperature CO_2 reduction using Mg-Ga liquid metal interface. Adv Mater Interfaces 10:2201625. https://doi.org/10.1002/admi.202201625

Zuraiqi K, Zavabeti A, Clarke-Hannaford J, Murdoch BJ, Shah K, Spencer MJS, McConville CF, Daeneke T, Chiang K (2022) Direct conversion of CO_2 to solid carbon by Ga-based liquid metals. Energ Environ Sci 15:595–600. https://doi.org/10.1039/d1ee03283f