

Article

Integrated sCO₂–SOEC Process for Carbon Monoxide Production from Natural Gas

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Abstract

Traditional fossil-fuel-based industries, including electric power generation and petrochemical production, are plagued by their inherent carbon dioxide emissions. Current efforts to minimize CO₂ discharge tend to focus on costly capture and sequestration rather than utilizing the process energy and products to produce profitable chemicals. This work describes a concept and design strategy for a comprehensive facility to convert natural gas and air to carbon monoxide, which could be further used to synthesize desirable compounds and fuels. The basic components of the process are an improved supercritical carbon dioxide (sCO₂) electric power plant for driving solid-oxide electrolysis cell (SOEC) reduction in CO₂ to CO. This study examines opportunities for integrating a sCO₂ process with an associated air separation unit (ASU) and an SOEC, including material recycling and heat integration. A Life Cycle Assessment (LCA) is expected to show more positive results for separate processes. Preliminary economic evaluations provide a profitable process and product route for the combined process, suggesting that carbon emission reduction could be good business.

Keywords: sCO₂ power cycle; carbon capture; carbon monoxide; solid-oxide electrolysis cell

1. Introduction

While there are many factors contributing to climate change, the principal contribution is carbon dioxide (CO₂) emissions from a variety of sources. The question posed by Cordova et al. [1], “What should we do with CO₂ from biogas upgrading?”, also applies to processes based on fossil fuels. A recent National Academies Report [2] describes in detail the “market opportunities for carbon dioxide and coal-waste utilization, the status of utilization technologies and their research, development, and demonstration needs, and the needs for and impacts of infrastructure for CO₂ utilization”. Farooqi et al. [3], Zito et al. [4], and Jin et al. [5] have performed extensive literature reviews of electrochemical processes for converting CO₂ into valuable chemicals. The International Union of Pure and Applied Chemistry (IUPAC) has identified Electrochemical Carbon Capture and Conversion as one of the Top Ten Emerging Technologies in Chemistry for 2025 [6] with “the greatest capacity to open new opportunities in chemistry and beyond.”

As with all the apparent opportunities, selecting implementations with the greatest impact requires investigation. For the processes of interest herein, technoeconomic factors

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are foremost, beginning with the formulation and computational simulation of integrated process systems to uncover the most effective and economic configurations. Thermal, electrical, and material integration should significantly reduce capital and operating costs compared to traditional standalone processes [7,8].

Molière et al. [9] present an overview of sCO₂ power cycles, describing their advantages, limitations, and challenges to industrialization of their technology.

Grim et al. [10] present a thorough technoeconomic analysis of potential products from CO₂ conversion. They conclude that based on manufacturing costs and prices for the period 2014–2018 [11], no product would be economically viable without significant advances in technology and lower input costs. Since that time, prices have increased considerably [12], along with recent advances in power generation efficiency and electrolytic chemical synthesis, indicating that the situation could now be more favorable. In particular, supercritical carbon dioxide power cycles (sCO₂) have achieved efficiencies exceeding 55% (LHV) at scales of 200–300 MW, as demonstrated in the Allam cycle [13–15], while solid-oxide electrolysis cells (SOECs) now operate at current densities of 1–2 A/cm² with cell efficiencies of 80–90% [16–18]. While theoretical and experimental research is still needed to improve SOEC catalytic chemistry [19], it is appropriate to reconsider the technoeconomics of producing chemicals and fuels from fossil fuels without carbon emissions or sequestration.

Stevenson [20] has discussed in detail the issues associated with the utilization of CO₂, which they define as, “the conversion of CO₂ into fuels or usable chemicals that results in a net decrease in CO₂ emissions”. Four generally true assumptions about utilization have been examined along with thermodynamic arguments and stoichiometric constraints. The conclusion was that proposals for CO₂ utilization often do not reduce emissions and renewable energy resources will probably be needed to significantly reduce emissions. It is anticipated that the proposed route herein obviates these concerns and is both environmentally beneficial and economically feasible. This issue is expanded in Section 4.

Concept: This work explores electrolysis of CO₂ integrated with power generation to add value by synthesizing chemical products. This was prompted by results for a 300 MW supercritical CO₂ (sCO₂) power plant [15–17], whose combined CAPEX and OPEX exceeded expected revenue from power sales [17]. The configuration uses oxygen from an air separation unit (ASU) to combust natural gas and generate energy for power and its CO₂ working fluid. The power, heat, and produced CO₂ would be reduced to CO in an SOEC process for manufacturing useful chemicals, such as alcohols; partially oxygenated solvents, such as acetates; and fuels from Fischer–Tropsch synthesis [2–5,7–11,21]. CO was selected as the initial product from a combined process based on thermodynamic and economic analyses. CO was also suggested as the product with the lowest gap between cost and selling price [7].

This paper is in the spirit of the pioneering 1988 textbook, *Conceptual Design of Chemical Processes*, Section 1.2, by J.M. Douglas [22] that introduced a Hierarchical Approach to Conceptual Design. He called for “first developing simple solutions and adding successive layers of detail.” Figure 1 provides a simple “superstructure” for an integrated natural-gas-to-chemical electrochemical manufacturing facility by conceptually illustrating possibilities for material recycling and heat energy integration to maximize process productivity and efficiency. This paper also adds layers that combine sCO₂ power generation with SOEC processes to produce CO. Details, such as experimental verification and economics, can be addressed in the future.

Figure 1 includes an ASU, a sCO₂ power plant, a SOEC for the reduction in CO₂, and the chemical synthesis using CO. In addition to oxygen (O₂) for the power cycle, the ASU produces highly pure nitrogen (N₂), which could be used to purge the SOEC; to feed the synthesis of chemicals, such as nitriles and cyanides; or be sold directly. The SOEC

produces oxygen, which can be fed back along with the N_2 purge to the sCO_2 process, limiting the ASU burden. The power cycle generates electricity and CO_2 for reaction in the SOEC. It also emits heat energy that preheats the SOEC feed and produces water for chemical synthesis.

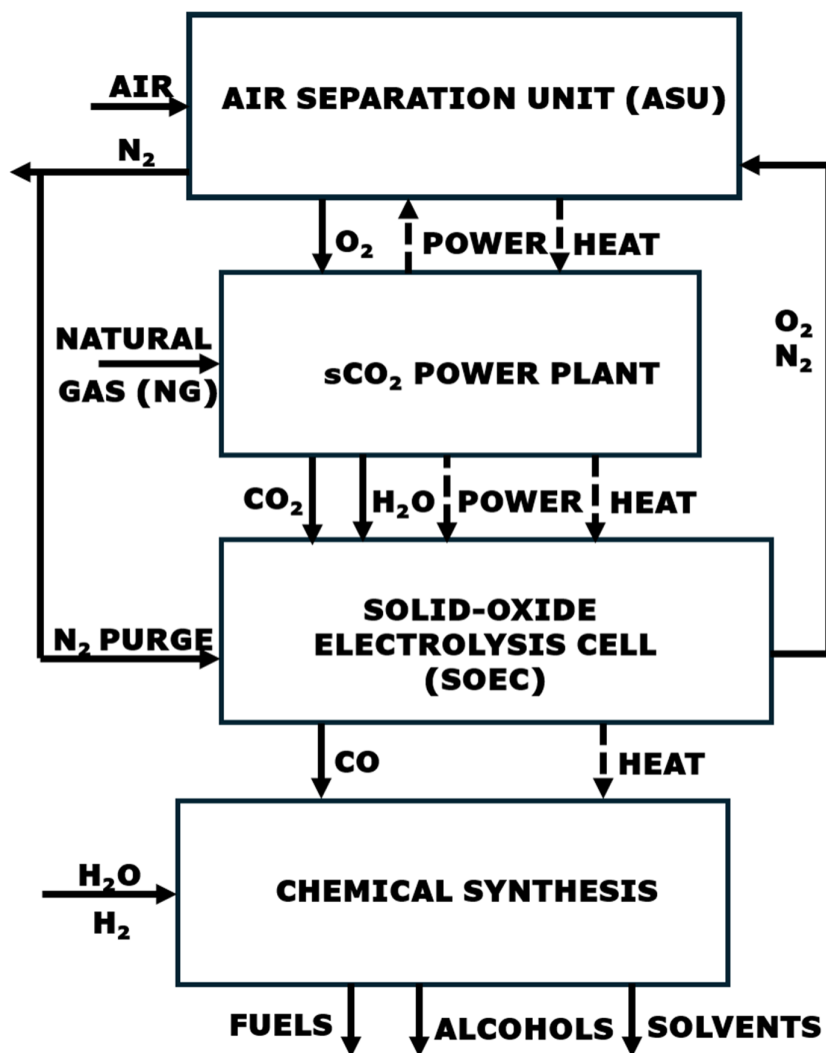


Figure 1. Concept diagram of an integrated natural gas-to-chemical electrochemical manufacturing process.

While the Allam-type sCO_2 power cycle [13–15] and solid-oxide electrolysis cells [16–18,23–26] have been extensively studied as standalone technologies, an integrated configuration for CO production has not been systematically evaluated. Previous power-to-X studies have typically examined CO_2 electrolysis as an independent process, based on assumed external renewable electricity and CO_2 feed from carbon capture processes [7–10]. Likewise, technoeconomic analyses of sCO_2 cycles have focused on power generation without considerations of using highly pure CO_2 output as feedstock for chemical synthesis [14,15]. The specific knowledge gap addressed herein is whether the integration of these two technologies, including material and energy exchange between SOEC and sCO_2 , as well as its associated ASU processes, would have favorable economics compared to the standalone alternatives.

Accomplishing the present goal requires comprehensive mass and energy balances for the quantification of the combined sCO₂–SOEC process, the net power requirements and CO production capacity for a specified sCO₂ plant, and a preliminary economic assessment specific to this integrated configuration, accounting for both capital and operating costs. The economic costing methodology is from established sources [27], the sCO₂ cycle simulations build upon prior work [15], and the estimates of SOEC properties are taken from the literature, whereas the integrated analysis and resulting process economics are new.

Next, a summary is given of the strategy introduced for designing this process, wherein electrical power is generated while decarbonizing natural gas to produce useful chemicals and liquid fuels. First, the combined sCO₂ power generation and SOEC reactor process is described. This is followed by thermodynamic and economic treatments for feasibility and profitability. Finally, life cycle analysis and future work are outlined.

2. Product Selection

The first decision in any design process is selecting the target product. The thermodynamics of possible reactions and their elementary economics, including mass and energy balances for electric power and CO production, are used. Following calculations of the minimum work energy based on the Gibbs energy of reaction, the economics of the process steps, including their efficiencies, are obtained using the literature's information. Initially, a simple return on investment (ROI) is estimated, with a range of ROI estimated from 10% uncertainty in the basic variable variations given. Appendix A provides details of all calculations.

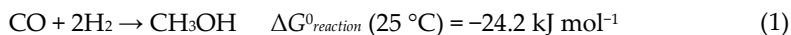
Thermodynamics: After screening various candidates, and by using the information provided by Küngas [24], carbon monoxide (CO) was selected as the target SOEC conversion product for the following reasons. Thermodynamic property values have been taken from [28].

Ease of Production: CO₂ can be converted to CO via catalytic dissociation. A sCO₂ cycle would provide the work energy. Here, a positive $\Delta G_{\text{reaction}}$ indicates the work energy required for the reaction:

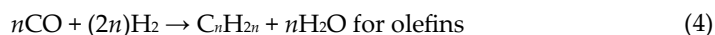
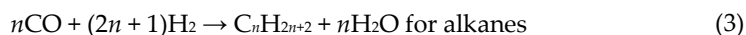


Note that $\Delta G^0_{\text{reaction}} (25\text{ }^\circ\text{C}) = 257.2\text{ kJ mol}^{-1}$, while $\Delta G_{\text{reaction}} (800\text{ }^\circ\text{C}) = 190\text{ kJ mol}^{-1}$

Chemical Product Value: In chemical synthesis, CO can be a precursor for many oxygenates via spontaneous reaction with H₂; for example,



Fuel Synthesis Value: Through the well-established Fischer–Tropsch synthesis [7,22], CO and H₂ can be catalytically converted into a spectrum of hydrocarbon fuels. This creates a direct bridge between clean power and liquid fuel sectors.



When $n = 1$ for Reactions (3) and (4), then $\Delta G^0_{\text{reaction}} = -142\text{ kJ mol}^{-1}$ for (3) and $\Delta G^0_{\text{reaction}} = +255.4\text{ kJ mol}^{-1}$ for (4), indicating olefins would be more easily obtained [7].

Power Consumption: Of importance is the balance of sCO₂ power and CO production. That is, can the power cycle generate enough, or more, electricity to convert all the CO₂ it produces, yielding excess power, or will there be excess CO₂? As shown below and in Appendix A, the estimates indicate that all the CO₂ can be converted with about one half of the generated electricity, so the rest can be used downstream or marketed.

This is an outcome of systems analysis that would not be apparent using separate processes.

The analysis draws on a detailed ASPEN PLUS process simulation of a slightly profitable sCO₂ power cycle [15], which provides inputs, outputs, equipment, and costs for power generation. The properties were calculated using the CPA model of Kontogeorgis et al. [29], which treats complex non-ideal mixtures. The simulations do not include oxygen recycle and nitrogen purge, so related improvements in overall *ROI* are not included.

Economics: The economic viability of integrating CO₂ electrolysis with a sCO₂ cycle can be evaluated against a baseline scenario using return on investment (*ROI*):

$$ROI_{cc} = (S_{co} - OPEX_{cc})/CAPEX_{cc} \quad (5)$$

where *ROI_{cc}* is the return on investment for the combined sCO₂–SOEC plant; *S_{co}* is the annual CO sales revenue; *OPEX_{cc}* is the annual cost of operating the SOEC plant, including maintenance; and *CAPEX_{cc}* is the total (installed) capital investment for the combined SOEC, sCO₂, and ASU plants. *OPEX_{cc}* has been set to equal the revenue from marketing the sCO₂ power plus a commonly used fraction of 4% of *CAPEX_{cc}* for maintenance and operations [27] (see Appendix A). We note that only the sCO₂ Allam power cycle and associated air separation unit (ASU) have been simulated using a detailed ASPEN PLUS simulation with cost estimates, yielding a slightly profitable plant. Next, the *ROI_{cc}* is estimated for the power cycle process integrated with the SOEC plant, using the approximate literature data and conceptual design estimates. When promising *ROI_{cc}* estimates are obtained, experimental and detailed theoretical calculations can be justified in the next conceptual design stage.

At this point, as we describe calculations using Equation (5); the reader can find these details in Appendix A. First, the current production capacity and market conditions establish the potential annual sales revenue from carbon monoxide (CO) production. The best of the advanced sCO₂ power cycles simulated by ASPEN PLUS [15] produced power costing the national average of 142 USD/MWh and emitting 33 kg/s of CO₂. These are based on the analysis by Chen et al. [14], with values probably reliable to within ± 10%.

Therein, 0.75 kmol/s CO is produced from a minimum of 142.5 MW of electricity at the SOEC conditions of 800 °C. Assuming 90% transmission and Faradic efficiencies, a 300 MW power plant and SOEC is anticipated to produce 0.483 MMtonne/yr of CO and 124 MW power. The current market price for CO depends on the country and ranges from 321 USD to 813 USD/tonne [12]; a price of 600 USD/tonne has been assumed herein. This price is considerably higher than that given by Grim et al. [10], which was taken from the period 2014–2018. The projected annual sales revenue, *S_{co}*, is therefore 358 MMUSD/yr. The annual electricity cost would be 197 MM USD/yr. The SOEC electrolyzer *CAPEX_{cc}* is estimated to be 2000 USD/kW [16], which is equal to 1162 MMUSD annually. The *CAPEX_{cc}* for the best sCO₂ and ASU facility is 1500 MMUSD, giving a total of 2662 MMUSD. Excess power can be sold at 142 USD/MWh, yielding a revenue of 139 MM USD/yr.

These values lead to a *ROI* of 9.0%, which can be commercially attractive. This can be compared to the small *ROI* of 0.41% for power from the best standalone sCO₂ power plant [15]. A 45Q tax credit under the Inflation Reduction Act 2022, currently 0.085 USD/kg CO₂, was taken with the power plant, so it cannot be taken here. As detailed in Appendix A, the worst case scenario, assuming that these values are in error by 10%, reduces *ROI* to −0.9%, while the best case would yield *ROI* of 21.9%.

3. Process System Elements

Having introduced our conceptual design strategy, the process systems elements are considered next.

sCO₂ Power Generation: Conventional power generation cycles use air combustion of fuel, usually of fossil origin, to produce a high-temperature, high-pressure flue gas and steam that are expanded through power-producing turbines. The steam is condensed to water before being boiled again, while all gas emissions have diluted CO₂, which must be captured. When the fuel is natural gas (NG), such cycles are identified as NGCC-CC. The capture of CO₂ is energy inefficient and requires large facilities, expensive chemicals, and extremely great water usage. The ratio of power to fuel energy, or the efficiency, of these cycles ranges from 20 to 40% [21], as confirmed by simulations [15]. Cycles based on sCO₂ working fluid, such as the Allam cycle [13–15], have significantly higher efficiency and produce highly pure CO₂. Figure 2 shows the basic configuration adapted from Allam and coworkers [14]. Natural gas is combusted at high pressure with high-purity oxygen from an ASU to give a working fluid of highly pure CO₂ and some H₂O. This is expanded through the turbines, cooled in a recuperative heat-exchanger network, and followed by the emission of some high-purity CO₂ and water separately. The balance of the CO₂ is cooled to a dense phase and pumped for cycle reuse. Power generation efficiencies from these cycles have been shown to be as high as 55% [15], meaning less fuel is consumed. They also consume much less water than in NGCC-CC processes.

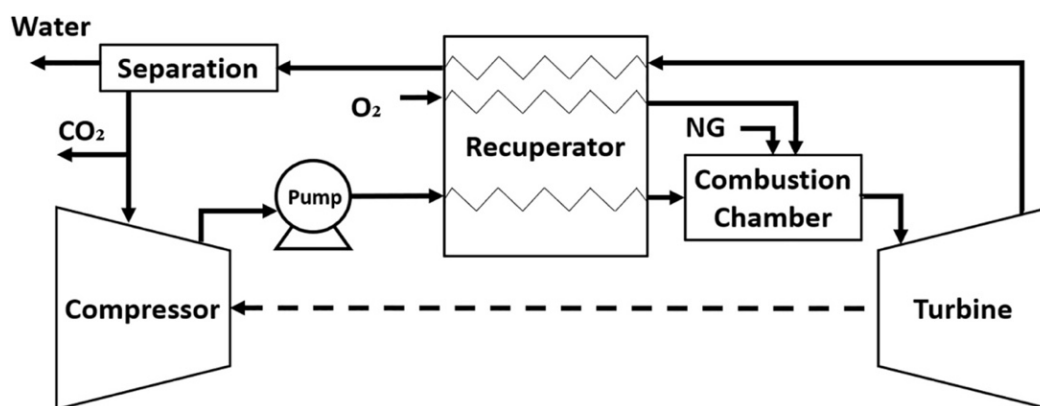


Figure 2. Block diagram for the original Allam sCO₂ cycle [14].

Key Advantages of sCO₂ Power Cycles:

- **Carbon Capture:** The cycle produces a high-purity, high-pressure CO₂ stream, which can be directly fed into chemical synthesis processes. There is no need for complex and energy-intensive post-combustion separation processes that are required in conventional plants.
- **High Efficiency:** Net power plant efficiencies can exceed 50% (LHV), which is significantly higher than NGCC-CC facilities [15].
- **Water Conservation:** The design consumes much less cooling water by avoiding steam condensation. Further, it produces a water stream for general use in water-scarce regions or for chemical synthesis.
- **Scalable CO₂ Output:** The scale of sCO₂ cycles (200–300 MW) aligns with the power requirements for industrial electrolytic processes.
- **An ASU is used to provide O₂ for NG combustion in sCO₂ systems.** It also produces N₂, which can be marketed, used as a purge for SOEC processes, or fed to synthesis plants to produce nitriles or cyanides.

Because CO₂ emissions remain from sCO₂ power cycles, it is important to consider how to deal with them, especially when they can generate revenue and mitigate emissions.

Status of sCO₂ Power Generation Technology: Presently, sCO₂ power generation is in the pilot phase of development, with a 10 megawatt supercritical carbon dioxide (sCO₂) test facility at Southwest Research Institute (SwRI) in San Antonio, Texas, USA [16]. It has completed its first phase of testing by achieving its full operational speed of 27,000 rpm and operating at a target turbine inlet temperature of 500 °C, and it is synchronized with the electrical grid.

Molière et al. [9] say the technology could have significantly greater potential through efficiency enhancements and reductions in component sizes. They list the most important technical challenges as: accuracy of working fluid properties, turbomachine rotor stability, materials for dealing with strong pressure and high thermal gradients, control of CO₂ leaks, and turbomachinery efficiency. Our analysis assumes that these challenges can be overcome and will not limit the possibilities of our conceptual design.

SOEC Electrolysis: Solid-oxide electrolysis cells (SOECs) consume CO₂ for the reduction to CO at high temperatures (800 °C) via Reaction (1) [17,18,24–26,30]. As summarized in Figure 3, at the cathode, some of the CO₂ decomposes into CO and oxide ions. The ions migrate through a solid-ceramic electrolyte to the anode, where they recombine to produce O₂. The unreacted CO₂ is recycled to the feed after separation with a PSA unit. Purge gas, such as N₂, sweeps the O₂ from the anode; conversely, the sweep gas preheats the purge. Additional heat exchange of the feed and recycle streams may be required to meet the necessary cell conditions. This technology has been extensively used by the Haldor-Topsoe company for their decarbonized CO production process (eCOs) [31]. PSA separation can give very high purity CO.

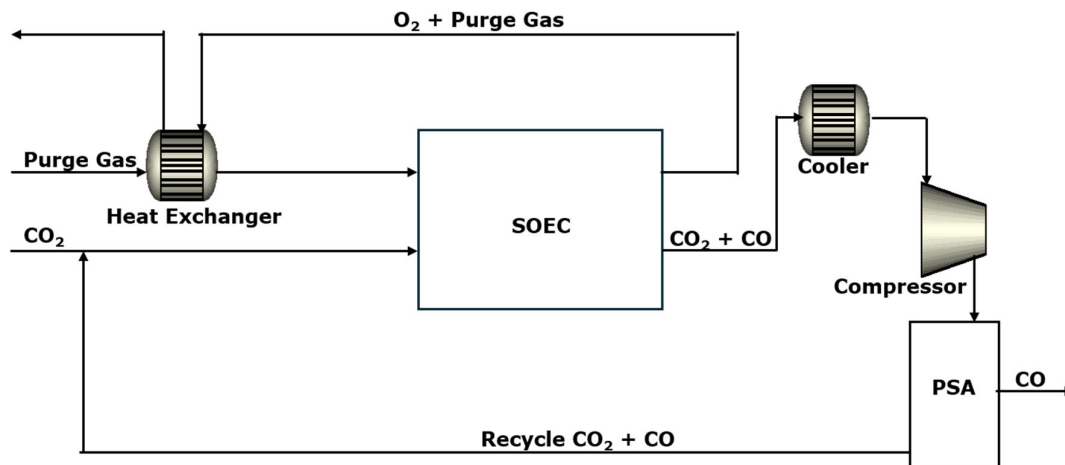


Figure 3. Block diagram for SOEC decomposition of CO₂; adapted following [30].

Key Advantages for SOEC Electrolysis Cells:

- **Favorable Thermodynamics:** As the temperature is increased, the reversible work, given by the Gibbs energy of reaction ($\Delta G_{\text{reaction}}$), is increased, and the process irreversibilities related to entropy generation (S_{gen}) are reduced.
- **Fast Kinetics:** Elevated temperatures accelerate electrode reaction rates and reduce activation overpotentials.
- **Direct Production of CO:** The electrolysis product can be a pure CO stream without oxygenates, avoiding complex product purification of low-temperature processes.

- Coproduction of O₂: High-purity oxygen is produced at the anode, which can be directly recycled to the Allam cycle combustor, reducing the air separation unit (ASU) production.
- State-of-the-art SOEC cathodes typically employ Ni-YSZ cermets for their catalytic activity and stability [20], while perovskite-based anodes, such as lanthanum strontium cobalt ferrite (LSCF), are commonly used for oxygen evolution [31]. Mixed ionic–electronic conductors (MIECs), like BSCF, are also being explored to improve oxygen transport and reduce polarization losses [21].

SOEC—Electrolysis Cell Concerns: SOEC processes suffer parasitic side-reactions that could consume reactant or product species, reducing the CO yield. Careful control of the process conditions and innovative catalysts are needed for the most effective processing.

The Boudouard disproportionation reaction forms solid carbon via $2\text{CO} \rightleftharpoons \text{C} + \text{CO}_2$, with the solid carbon forming as coke whiskers [26,32]. This reaction is highly favored at temperatures typical of SOEC operation, especially in the presence of catalytic anode materials, e.g., nickel. Coke deposition not only directly reduces CO yield, but it can also deactivate catalysts, block porous electrode structures, and ultimately lead to mechanical degradation and cell failure.

Dai et al. [33] prevented this undesirable reaction with microwaves in the presence of a barium-carbonate promoter. The experimental results gave CO₂ conversion up to 99.9%, compared to 29% at 900 °C using conventional heating (CH) methods. Also, their kinetic studies found that microwave heating significantly reduced the activation energy for CO₂ reduction from 148.9 kJ/mol for conventional heating to 47.1 kJ/mol with microwaves.

Carbonate formation occurs from the direct reaction of electrochemically generated oxide ions (O^{2−}) at the cathode with incoming CO₂ to form carbonate ions (CO₃^{2−}) via $\text{CO}_2 + \text{O}^{2-} \rightarrow \text{CO}_3^{2-}$. This reaction sequesters carbon in a stable ionic form and prevents its reduction to CO. Carbonate ions can migrate through the electrolyte, contributing to unwanted ionic currents and causing long-term stability issues. This pathway occurs at the electrode surface with certain electrolyte materials and under local alkaline conditions. Although carbonate formation at high temperatures is difficult to avoid, potential prevention methods include modifying reaction environments by avoiding alkaline/neutral solutions [34] and constructing zero-gap membrane electrode assembly devices to reduce cell resistance in acidic conditions [35].

It is possible that these long-term effects could negatively affect economics, as the production rate would decrease while the costs of catalysts and installation would increase CAPEX_{cc}. We are unable to estimate these, but we expect that their impact would not decrease the ROI significantly. The analysis herein does not account for any product loss, as measured by a carbon balance from the input natural gas methane to the output CO.

4. Discussion

Luo et al. [36], Rijo et al. [37], and Artz et al. [38] have listed the following challenges to SOEC implementation: mechanisms of CO₂ reduction; strategies for electrode modification; improvement of electrode activity and durability; and economic and feasibility assessments of integrated processes. Herein, the last is addressed, but the others provide opportunities for theoretical and experimental research.

Returning to the Introduction, where we noted that Stevenson [21] had described fundamental challenges to CO₂ utilization, the proposed integrated process overcomes these by noting exceptions to his four assumptions.

Assumption 1 is that energy forms are interchangeable. Three exceptions can be identified.

1. Energy must be transported over large distances. The integrated process generates electricity from natural gas, with power and CO₂ used in nearby SOEC facilities, without any emissions.
2. Excess energy is available intermittently. Some of the power is used immediately and directly in the SOEC reaction, and the balance is continuously available.
3. Renewable energy is captured directly in a CO₂ conversion step. The energy here is not renewable, though the issue is covered under the second exception.

Assumption 2 is that process inefficiencies are similar for processes making the same product. One exception is:

4. The use of sCO₂ leads to higher production rates, better product selectivities, lower process heat losses, and otherwise improves overall process efficiency. The product is CO, so efficiencies for producing chemicals and fuels would be the same as in current design with this feed. Furthermore, SOEC electrolysis is more efficient than alternative processes for CO generation. Finally, because power generation is more efficient than traditional NGCC processes, this concern is also met.

Assumption 3 is that fossil fuels are the source of a significant amount of society's energy consumption. The energy source promoted herein is the fossil fuel natural gas, but no emissions occur.

Assumption 4 is that the utilization of CO₂ does not affect the demand for individual chemical products or the relative production of individual energy sources. As shown in Appendix A, the production of CO uses only 174 MW of a 300 MW power plant and contributes only very small fractions to the current international CO market and power.

We consider these factors affecting emissions:

- a. The amount of CO₂ incorporated in the new product: The integrated processes utilize all carbon dioxide produced.
- b. The energy costs: The integrated process is profitable.
- c. The fate of the end product: The processes do not change from current practice.
- d. The carbon content of the replaced monomer relative to that of CO₂: All carbon is used.
- e. The efficiency of production of the monomer units that are replaced: The efficiency of the process is apparently greater than those currently used.

Stevenson [21] asks: "What about renewable energy?" Our answer: "It will be better to use available renewable electricity directly rather than using it less efficiently to upgrade CO₂." Stevenson concludes with a section, "What could work?", where we reiterate that technical and economic feasibility can be demonstrated in addition to meeting thermodynamic constraints, with the citation of guidelines and standards for their analyses [39]. Also, in Figure 5, Stevenson gives three processes for allocating renewable energy resources. Ours, shown in Figure 1, is another alternative, though not one built on renewable energy, and has the benefit of CO₂ emission only from disposal of the final product. Finally, Stevenson gives a list of approaches to reduce emissions without CO₂ utilization. We wholeheartedly support such initiatives while encouraging the adoption of integrated processes.

A technoeconomic analysis of a novel integration of high-temperature, solid-oxide electrolysis cells (SOECs) with sCO₂ power cycles to convert captured CO₂ into carbon monoxide (CO) has been presented. The primary goal was to evaluate the feasibility of transforming the power cycle from carbon sequestration into a carbon utilization platform. The conclusion is that such processes can be both technically and economically successful.

From a *technical perspective*, integration is fully feasible. A 300 MW sCO₂ cycle can drive the endothermic CO₂ electrolysis process for CO production, which can be used to synthesize important chemical intermediates, solvents, and fuels, as well as produce excess power.

Additionally, *economic analysis* has revealed profitability from producing CO. Reactions with this product to generate higher-value chemicals could yield greater profits or allow the synthesis of desirable, albeit less profitable, chemicals.

At this point, an ultimate chemical product has not been selected. Consequently, profitabilities for candidate chemicals have not been evaluated, mainly because integration of the selected process with CO generation processes could affect the technoeconomic outcome. Reductions in operating expenses (OPEX) are expected from material recycling and heat integration. The costs of equipment, materials, and fabrication will affect CAPEX. Our recommendations focus on the combined power and SOEC processes and indicate the need to provide a simplified Life Cycle Assessment (LCA), along with suggested research to close the knowledge gaps.

A Life Cycle Assessment (LCA) of the integrated process in Figure 1 would require selecting the final chemical synthesis product(s); the literature on this subject is available. Yue et al. [40] have produced LCAs for a variety of C1 and C2 chemicals, concluding that “C2-chemical production has more positive energy impacts, but more negative environmental impacts than C1-chemical production”. Artz et al. [34] discussed conversion of CO₂ to Synthesis Gas and oxygenated compounds, emphasizing their “relative improvement in carbon balance”. Jolaoso et al. [41] performed a LCA for SOEC production of H₂ from different energy sources. They concluded that solar photovoltaic (PV) energy is the best energy source. Herein, the power source is a sCO₂ process, for which a modified LCA has been performed [16], showing much less energy is used and the environmental impact is reduced than with conventional NGCC-CC processes. However, it is not likely to be as benign as PV energy. We note that energy integration and materials recycling can make significant improvements in energy and environmental impacts. As shown above, these can be maximized by their integration, as shown in Figure 1. The principal environmental effects of SOEC processes involve various materials for construction, like cell ceramics and metal catalysts and interconnects. The catalysts contain critical metals, so stack lifetime and catalyst recycling are important [41], but this is not treated herein.

Initial steps have already been taken by industry. Mitsubishi Heavy Industries (MHI) has completed a pilot demonstration of an integrated production route for Sustainable Aviation Fuel using a variation similar to this research by combining co-electrolysis of CO₂ and steam with downstream fuel synthesis [42]. Recent work on producing bulk chemicals and fuels other than methane has been attempted but has not generally been successful [43].

5. Conclusions and Future Research

A design strategy has been developed for a technically and economically sound and innovative closed-loop process for power generation from natural gas, with CO and H₂O as separated products. Integration of air separation, supercritical CO₂ power production, and SOEC electrolysis was shown to be technically and economically attractive. Widespread commercial deployment of the concept will depend on research to mitigate potential issues with SOECs, the recognition of candidate chemical products from CO, and investment in large, complex facilities.

Research is encouraged in:

- Developing sCO₂ power cycles in full.

- Developing coke-resistant electrode materials for SOECs to improve conversion efficiency and lifespans, inhibit undesirable byproducts, and extend catalyst lives.
- Increasing process integration by greater examination of material recycling, as well as work and heat integration.
- Expanding product portfolio details by integrating a methanol synthesis reactor or a Fischer–Tropsch unit directly downstream of the SOEC.
- Examining the technoeconomics of other chemical products from CO.

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Data Availability Statement: Data supporting the findings of this study are available in Appendix A accompanying this paper. Additional data may be obtained from the corresponding author upon request.

Conflicts of Interest: The authors declare no conflict of interest.

Acronyms

ASU	Air Separation Unit
CAPEX	Capital Expenditure
CO	Carbon Monoxide
CO ₂	Carbon Dioxide
F-T	Fischer–Tropsch Synthesis
IUPAC	International Union of Pure and Applied Chemistry
LCA	Life Cycle Assessment
LHV	Lower Heating Value
NG	Natural Gas
NGCC	Natural Gas Combined Power Cycle
NGCC-CC	Natural Gas Combined Power Cycle with Carbon Capture
OPEX	Operating Expenses
ROI	Return on Investment
sCO ₂	Supercritical Carbon Dioxide
SOEC	Solid-Oxide Electrolysis Cell
Syngas	Synthesis Gas

Appendix A. Calculations of CO Production and Profitability

Appendix A details the economic calculations in Section 2. Thermodynamic properties were taken from [28].

Quantity	Results	Data
Reversible SEOC Work	$W_{rev} = \Delta G_{reaction} (25\text{ }^{\circ}\text{C})$ $= 257.2\text{ kJ/mol CO}$	$\Delta G_f^0(\text{CO}) = -137.2\text{ kJ mol}^{-1}$
$\text{CO}_2 \rightarrow \text{CO} + 1/2\text{O}_2$	$W_{rev} = \Delta G_{reaction} (800\text{ }^{\circ}\text{C})$ $= 190\text{ kJ/mol CO}$	$\Delta G_f^0(\text{CO}_2) = -394.4\text{ kJ mol}^{-1}$

ASPEN PLUS simulations in [16] provided the CO₂ flow rate from the best sCO₂ power cycle.

$$\begin{aligned}\text{Mass rate of CO from 300 MW} &= 33 \text{ kg/s CO}_2 = 0.75 \text{ kmol/s CO}_2 = 0.75 \text{ kmol/s CO} = 21 \text{ kg/s CO} \\ &= 0.0025 \text{ kmol CO/MJ} = 0.07 \text{ kg CO/MJ}\end{aligned}$$

For the minimum required power for 0.75 kmol/s CO at 800 °C:

$$= (190 \text{ MJ/kmol})(0.75 \text{ kmol/s}) = 142.5 \text{ MW}$$

Assumptions about SOEC process efficiency were from [9] and its operation.

For 90% reversible power, 90% transmission, and SOEC Faradic efficiency:

$$\text{Required Power} = 142.5/(0.9 \times 0.9) = 176 \text{ MW} < \mathbf{300 \text{ MW}}$$

Operation with 10% down time = 7884 hours/year

$$\text{Annual CO production} = 21 \text{ kg/s} = 0.596 \text{ MMtonne/yr} = 0.24\% \text{ global market}$$

Global Market = 250 MMtonne/yr [7] Economics were evaluated using relations in Seider et al. [27] and uninstalled costs in [17].

$$\text{Bare module cost} = 3.3(\text{uninstalled cost})(\$2000/\text{kW}) [19] = \$6.6 \text{ MM/MW}.$$

The following are inserted in Equation (5):

$$\text{Revenue from CO @\$600/tonne } S_{\text{CO}} = \$600(0.596) = \$358 \text{ MM/yr}$$

$$\text{CAPEX}_i = \text{CAPEX (SOEC)} + \text{CAPEX (power + ASU)}$$

$$= \$6.6(176) \text{ MM} + \$1500 \text{ MM} = \$2662 \text{ MM}$$

$$\text{OPEX}_{\text{CC}} = \text{Power} + \text{SOEC maintenance}$$

$$= \$142(176)(7884) + 0.04(1500) = \$257 \text{ MM/yr}$$

$$\text{Revenue from 124/MWh excess power @ \$142/MWh} = (124)(\$142)(7884) = \$139 \text{ MM/yr}$$

$$\text{ROI} = \frac{\$(358 + 139 - 257)}{\$2662 \text{ MM}} = 9.0\%$$

The sensitivity of ROI to the assumptions above can be considered by adjusting the costs of power and bare modules, the total power requirements, the CO production rate, and the CO selling price. If CAPEX, OPEX, and the power requirement are 10% higher while the production rate and power prices are 10% lower, and where CO sales price is the current lowest, this worst case scenario would yield an ROI of −0.9%. Conversely, with estimates that are 10% more favorable and the CO price is the highest currently, the best case scenario would yield 21.9% ROI.

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