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A Circular Approach to Upgrade Cement-based CO_2 and Renewable H_2: Techno-economic Analysis of SNG Production

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The present work introduces a feasible, carbon-free integrated process that is based on the catalytic hydrogenation of industrially captured CO_2 via green H_2 towards the production of synthetic natural gas (SNG). Overall, three systems are being assessed from an economic point of view (sizing and simulation through Aspen Plus and HOMER software): a) H_2 production through a combination of renewable energy sources (RES) and water electrolysis, b) cement-based CO_2 capture through amines in a continuous absorption/regeneration cycle and c) CO_2 catalytic hydrogenation. The basis for the process simulation and the subsequent economic analysis lies on the exploitation of 1,400 kt/y CO_2 that are emitted from a typical cement industrial plant. According to this capacity, the integrated system is assessed for a yearly SNG production rate of ~500 kt/y. As was derived from the screening of different market exploitation scenarios (e.g., funding options, increase of CO_2 tariffs), a sustainable and feasible solution can be achieved (based on current market values), as long as the RES-H_2 system is funded by up to ~35 % or CO_2 penalty costs reach levels of >140 €/t in the near future.

1. Introduction

Cement-based CO_2 emissions have been estimated at 1.5 Gt/y (~5 % of total fossil-fuel based emissions) and with a highly increasing trend worldwide (Andrew, 2018). To this end, the deployment of CO_2 capture and utilization (CCU) processes into added value fuels/chemicals by excess RES-powered electrolytic hydrogen can reach carbon neutrality in the coming future. Such a concept can be realized with the aid of a MEA-CO_2 capture unit that can provide a high - purity CO_2 stream to a catalytic methanation process (other C_1 products can equally be considered). Simultaneously, a hybrid photovoltaic/wind generator system can provide the necessary H_2 through water electrolysis and serve as an efficient approach to store intermittent renewable energy into chemicals/fuels. The H_2/CO_2 process stream (at the desired ratio ~4) can then enter the proposed catalytic system for SNG production, followed by a purification system according to the desired end uses specifications. While the aforementioned individual systems have been studied from the perspective of process simulation and techno-economic assessment, only a few studies - to the best of the authors' knowledge - have provided a point-by-point preliminary cost analysis for the interconnected operation. The latter can be further enhanced by considering possible co/by-product valorisation and a feasible heat/energy management scheme. Indicatively, in a recently published study (Becker et al., 2019), a power-to-gas concept that is based on the partial utilization of the total CO_2 emitted from a 500 MW coal-fired power plant was assessed. While CO_2 capture and RES-H_2 systems were not thoroughly analysed, the options of excess heat exploitation and the need to provide an SNG drop-in stream to the existing gas pipeline network were clearly revealed. The nominal production cost for SNG was estimated at >100 €/MWh (~80 t/d) with the cost of RES-H_2 dominating process economics. In a similar approach, (Buchholz et al., 2014) a methanation reactor and its integration with a 80 MW_{el} alkaline water

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electrolysis and a CO_2 capture system was evaluated in a fossil fuel-based power plant. The plant economics indicated an SNG production cost that is several times higher than current natural gas market prices (700–1,000 €/MWh for a ~60 t/d production). In a similar concept, the research group of (Giglio et al., 2015) reported a cost of less than 100 €/MWh that is strongly dependent on both the CO_2 penalty cost and electricity prices. In another study (Davis and Martín, 2014), the authors compared the use of solar/wind power for SNG production and they concluded that the use of various RES sources, construction area and CO_2 penalty costs are strongly related within a techno-economic assessment. Recently, (Chauvy et al., 2020, 2021) a detailed feasibility assessment of an SNG production plant was conducted. Their operating scheme utilizes 10 % of a cement plant's flue gases (~10 t/h of CO_2) and wind-based hydrogen. Among several novel issues, the authors proposed an optimized heat recovery system towards the production of surplus electricity. A break-even price of 870 €/MWh was estimated for a continuous 3.6 t/h SNG production.
In light of the above issues, the present work assesses the feasibility of an SNG production plant utilizing a typical CO_2-containing flue gas stream from a cement plant and RES-based H_2. Within the assessment, issues such as equipment sizing, operating limitations, break-even prices and technology barriers are fully addressed. The aim of the study is to provide a viable and realistic solution (both from an economic and operating point of view) that can be flexibly incorporated "on-site" by similar cement production plants. The basis for this study was built upon recently reported techno-economic assessments from our research group regarding H_2 production (Ipsakis et al., 2018) and SNG production (Ipsakis et al., 2021.

2. Process System Description

Figure 1 illustrates the proposed integrated concept of SNG production through the direct use of cement-based CO_2 and RES-based H_2. Briefly, CO_2 capture unit is designed and simulated (via Aspen Plus) with an aim to process the total flue gas (FG) exit stream of a typical cement industry (~1,200 t/h, 97 °C, 1 bar) via an aqueous solution of MEA:H_2O at 66:34 (w/w %). A ~30 % w.t. water content has been suggested in (Chauvy et al., 2020) .RES-based H_2 production is designed and simulated (via HOMER software) with an aim to provide a fixed H_2/CO_2 ratio of 4 (exit of the electrolyser at 70 °C, 10 bar). Furthermore, the catalytic methanation unit is designed and simulated (via Aspen Plus) based on the catalytic performance of the most promising materials that have been recently reported one of our group studies(Varvoutis et al., 2021)). The SNG stream is comprised of CH_4 at a purity > 90 % and with a mass production rate of ~65 t/h.

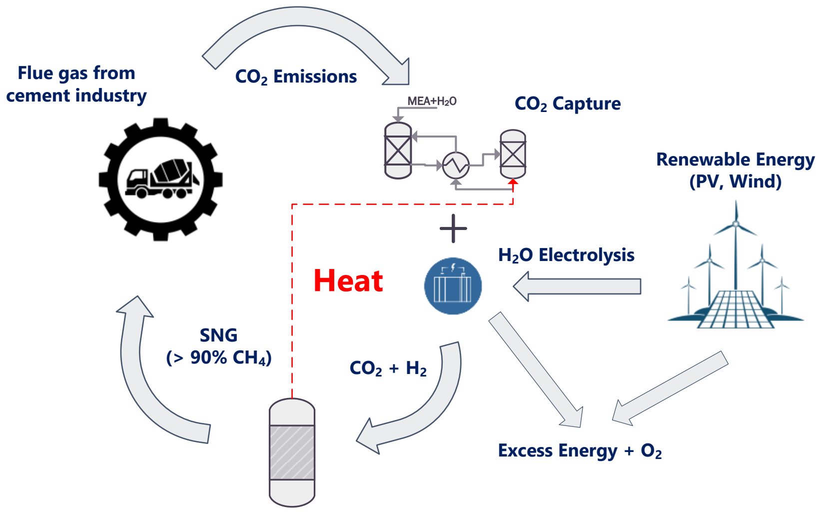

Figure 1: A scaled-up concept for SNG production through catalytic CO_2 hydrogenation (aided by RES)

A feasible heat management scheme was realized by a) considering a cooling tower (capacity at 650 MW_{th}) that meets the total cooling requirements of the integrated process, b) the heat coupling of the exothermic methanation unit with the endothermic MEA-CO_2 reboiler (heat recovery ~85 %) and c) an external steam provision (from C-free sources) that meets the remaining 15 % of the MEA-CO_2 heat requirements (~40 MW_{th}) and a series of low overall heat demands (~4 MW_{th}). Regarding electricity needs (e.g., compressors, blowers and pumps) of the integrated system, the excess energy from the RES-H_2 system fully meets the respective requirements (~20 MW_{el}). The SNG final stream can be recycled back to the cement industry and used internally to meet individual heat/energy needs and increase economic factors. Table 1 summarizes the necessary process operating conditions for inlet and outlet streams and more information can be found in (Ipsakis et al., 2021).

Table 1: Operating conditions for the integrated process

	CCU Inlet (*MEA is recycled)	CCU Outlet (4,190 kmol/h)	RES-H_2 outlet (24,320 kmol/h)	SNG final stream (4,380 kmol/h)
O_2, v/v %	12.8	--	50	--
N_2, v/v %	64.6	--	--	--
CO_2, v/v %	13.7	96.8	--	1.9
H_2O, v/v %	8.9	3.2	--	--
CH_4, v/v %	--	--	--	90.6
H_2, v/v %	--	--	50	7.5
FG flowrate, kmol/h	37,700	--	--	--
MEA, flowrate, kmol/h	84,000	--	--	--

3. Methodology for the techno-economic assessment

Based on the aforementioned integrated process simulation and design, the techno-economic assessment is built herein upon a step-by-step strategy. As shown in Figure 2, the process operating data and the established literature correlations, regarding equipment cost and sizing, form the basis for the techno-economic analysis. The major equipment cost (MEC) is used to estimate direct (e.g., installation, piping, services), indirect (e.g., supervision, construction) and other (e.g., legal fees, contingencies) costs. Ultimately, these costs provide the total fixed capital investment (including the working capital). Simultaneously, raw utilities cost (electricity, amines, steam, catalyst etc.) and the income by the product revenue (SNG, electrolytic O_2 and excess energy from RES) that are based on current market values, aid towards the estimation of direct production, annual fixed and general costs. These three categories sum-up to the total annual production cost. It is highlighted that cost escalation was accounted for through Marshall and Swift (M&S) indices for years 2020-2021.

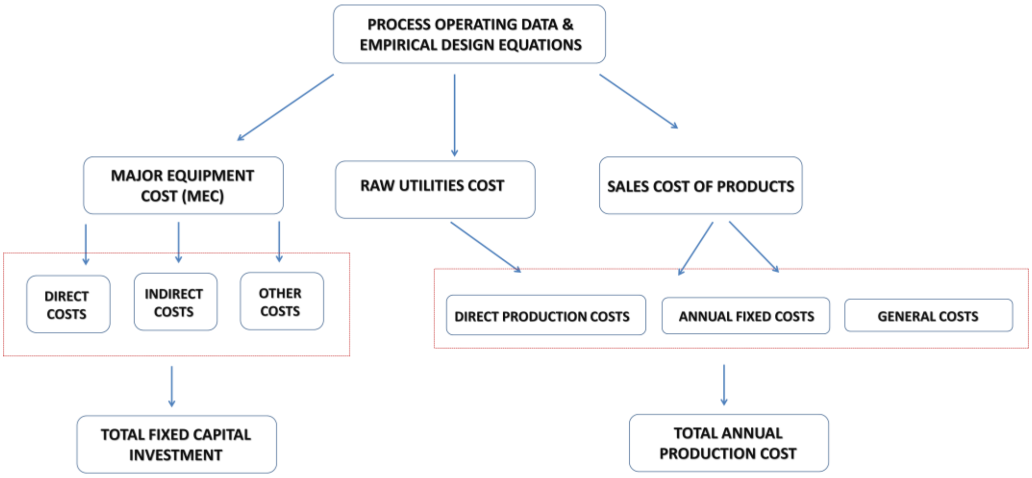

Figure 2: Step-by-step methodology for the techno-economic analysis

In order to properly assess the profitability of the proposed venture, a list of indices is also included in the presented analysis. These indices provide an overview of the yearly income and can lead to break-even prices: a) Gross Profit (R), b) Net Profit (P), c) Return on Investment (ROI) and d) Net Present Value (NPV). The useful life span of the project is assumed to be N = 25 y and with a plant operation factor of 85-90 %. As such, the depreciation and the capital recovery factors are assumed equal at 1/N or 5 %. The total income tax of 24 % on the net profit of the process plant is adapted throughout the yearly published economic performance calculations, while the risk factor and annual interest rate are equal to 8 and 4 %, respectively.

4. Techno-economic analysis

Table 2 summarizes the overall results from the techno-economic analysis. As can be seen, the RES-H_2 system dominates system economics due to the currently high CAPEX of electrolysers (at a flowrate of 16,110 kmol/h as shown in Table 1) powered from the solar/wind hybrid energy system. As has been explained in (Ipsakis et al., 2021) and through the aid of HOMER optimal solution, the power capacity of RES amounts to ~6 GW_p whereas the required size of water electrolyser is 2.2 GW_p. On the other hand, the 2nd most expensive system is the utilities support system that refers to the cooling tower, whereas the other two process systems present a similar level for their equipment cost and total fixed capital investment. Regarding the total annual production cost, the MEA-CO_2 system is associated with a higher cost due to the costly amine solution and external heat needs. The explanation for the low total annual production cost on the RES-H_2 system, in contrast with its highest total fixed capital investment, lies on the fact that this system is characterized by a high level of autonomy for its operation. The total utilities (cooling water, MEA make-up rate, steam, catalyst, distilled water) cost amounts to ~30 M€/y, whereas the total process product revenues are ~445 M€/y. Especially, ~60-65 % of the total product income is attributed to SNG exploitation and the rest is attributed to pure oxygen from water electrolysis. All calculations were based on available market prices: O_2 from electrolysis at 80 €/t, SNG > 90 v/v % CH_4 at 500 €/t and excess electricity at 0.08 €/kWh.

Table 2: Major equipment cost, total fixed capital investment and annual production expenditures for the coupled process systems in €.

Cost Type	MEA-CO_2 system	RES-H_2 system	CO_2 hydrogenation system	Utilities support system
Major Equipment Cost	26,750,000	7,800,000,000	26,660,000	238,350,000
Total Fixed Capital Investment	159,300,000	9,060,667,000	158,800,000	328,923,000
Direct Production Costs	24,700,000	34,400,000	14,250,000	13,800,000
Annual Fixed Costs	23,350,000	5,250,000	23,350,000	1,750,000
General Costs	2,340,000	160,000	2,152,000	64,000
Total Annual Production Cost	50,390,000	39,810,000	39,752,000	15,614,000

Table 3 presents the Gross Profit (R), Net Profit (P), Return on Investment (ROI) and Net Present Value (NPV) for 3 different cases. The 1st case is the base (or nominal) case, where current market values for the SNG and O_2 are used. The next two cases report the break-even price for only one market product each time, in order to provide an NPV = 0 (at 25th year). As can be seen, the base case scenario leads to a non-viable solution at the 25th year (NPV < 0) and hence it is concluded that the proposed venture cannot compete with conventional SNG and oxygen market values. Moving to the next cases where individual break-even prices are calculated, a Net Profit of ~621 M€/y is obtained, which is ca. 2.5 times higher than the base case Net Profit. This value can ultimately lead to a complete depreciation of the coupled total fixed capital investment and annual production costs over the course of the projected lifespan. These calculations have been based on the recycle of SNG to the cement industry and on a 25 €/t CO_2 penalty cost. Overall, the following break-even prices for the necessary depreciation at 25 years are estimated:

i) 1,070 €/t for the SNG (~2 times higher than the currently available market price)
ii) 240 €/t for the O_2 (~3 times higher than the currently available market value)

Table 3: Economic indices and break-even prices for SNG and oxygen from electrolysis.

Cost Type	Nominal/Base Case (current market values)	Break-Even Price for SNG	Break-Even Price for O_2
R, M€/y	271	773	773
P, M€/y	240	621	621
ROI, %	2.47	6.40	6.40
NPV, M€	-5,950	0.00	0.00
SNG, €/t	500	1070	500
O_2, €/t	80	80	240

5. Screening of feasible economic scenarios

Based on the above-mentioned analysis, it was revealed that the proposed venture is economically infeasible, as long as a) currently available market values and b) CO_2 penalty at 25 €/t are to be considered (worst-case scenario). In order to improve economics, a screening of alternative market scenarios was considered as:

i) CO_2 penalty cost at the values of 25, 50 and 100 €/t
ii) Subsidizing the RES-based system by 0-50 % and with a low CO_2 penalty cost (25 €/t)

5.1 Screening of CO_2 penalty cost and RES financing policy

As can be seen in Figure3a, the increase of CO_2 penalty at 50 and 100 €/t reduces SNG break-even price at the values of 950 €/t (11 % decrease) and 710 €/t (33.5 % decrease) respectively. For a penalty cost equal to 145 €/t (break-even cost for CO_2 tariff) the venture is completely comparable with current market SNG practices. Equivalently, O_2 break-even price is also closer with the currently postulated market value of 80 €/t, as CO_2 penalty fees are increasing. Regarding the effect of subsidizing part of the RES capital investment (see Table 2), Figure 3b highlights a significant advantage. Specifically, an increase of the financing strategy at 10 and 30 % (by considering a CO_2 penalty at 25 €/t), further reduces SNG break-even price by 14.5 % (915 €/t) and 44.5 % (595 €/t) and thus, closer to the current SNG market value. For a 36 % subsidizing policy, SNG and O_2 break-even prices are comparable to current market values (not shown in the bar-chart), whereas a 50 % financing policy can render these two commodities at their lowest possible values. It becomes obvious that if CO_2 penalty costs are higher than 25 €/t, then the financing policy can be significantly reduced.

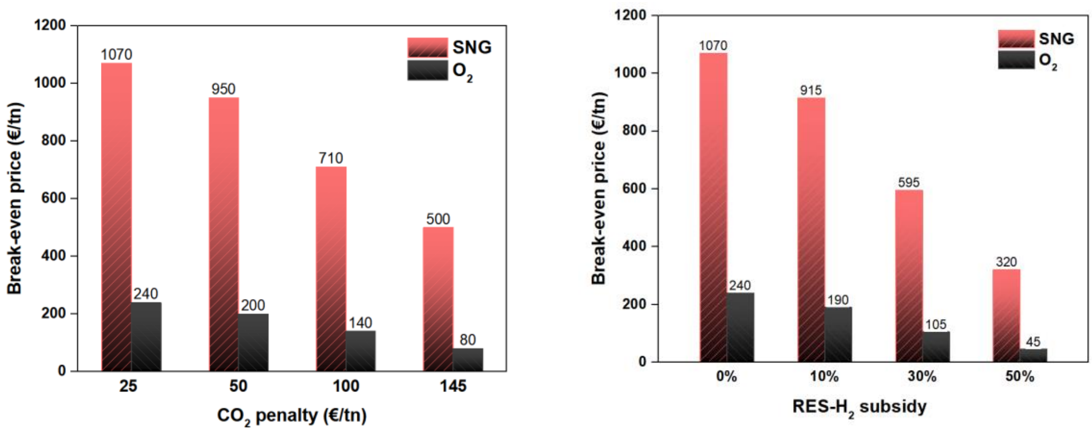

Figure 3: Break-even prices for SNG and O_2 as a function of CO_2 penalty costs (left) and RES subsidies (right).

5.2 Critical Analysis & Discussion

The screening of realistic scenarios clearly showed that the break-even price of SNG is not far from being competitive in the next few years. Moreover, the concept of CO_2-to-C_1 fuels, proposed herein, has a tremendous advantage of establishing a circular and C-free economy, in line with the current efforts for clean energy transition. Apart from exploiting industrially captured CO_2 emissions to generate SNG, the present scheme can also be regarded as an alternative approach to effectively store the excess RES power through electrolytic hydrogen. Hence, the large-scale integration of RES in the energy and transportation sectors can further be promoted. Even though the RES-hybrid system dominates overall economics, it is expected that their capital cost due to economies of scale will be substantially decreased with the progress toward complete decarbonization. If such a progress is achieved, then the break-even prices of SNG and co-products such as O_2 could be further decreased on the road to reach the future market values. Towards this scenario, the increase of CO_2 penalty costs and/or partial subsidy of the RES hybrid system also favor the proposed venture profit for the near future. Another advantage refers to the flexible modification of the catalytic methanation process system towards the production of other chemical commodities that will still be based on CO_2 hydrogenation: e.g., methanol, DME, olefins and Fischer-Tropsch liquid fuels. Hence, system economics can be further improved if the market trend switches to such value-added products.

6. Conclusions

The present work delved into the techno-economic evaluation of a direct CO_2-to-SNG scaled-up integrated process. It was revealed that an annual production of ~500 kt/y of synthetic natural gas (SNG) can be achieved under the capture of ~1,400 kt/y CO_2 (derived from a cement industry) and through the aid of 32 t/y of RES-based H_2. The integrated process systems were designed with an aim to operate autonomously and with a minimum environmental impact, where it was shown that the catalytic methanation process can satisfy the 85 % of the total heat demands of the MEA-CO_2 capture system, whereas the excess energy from the RES-based system can meet the total electrical energy needs. A break-even price of 1,070 €/t for SNG was estimated, which was aided by the revenue of electrolytic O_2. Although this value is nearly 2 times higher than currently reported prices, it was shown that the decrease of RES-H_2 costs through a dedicated financing policy and the expected increase of CO_2 penalty costs will render the proposed coupled process more viable, in terms of plant economics and environmental benefits.

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References

Andrew R.M., 2018, Global CO_2 Emissions from Cement Production, Earth System Science Data, 10, 195-217.

Becker W.L., Penev M., Braun R.J., 2019, Production of Synthetic Natural Gas from Carbon Dioxide and Renewably Generated Hydrogen: A Techno-Economic Analysis of a Power-to-Gas Strategy, Journal of Energy Resources Technology, Transactions of the ASME, 141, 1-11.

Buchholz O. S., Van Der Ham A.G.J., Veneman R., Brilman D.W.F, Kersten S.R.A., 2014, Power-to-Gas: Storing Surplus Electrical Energy a Design Study,Energy Procedia, 63, 7993-8009.

Chauvy R., Dubois L., Lybaert P., Thomas D., De Weireld G., 2020, Production of Synthetic Natural Gas from Industrial Carbon Dioxide, Applied Energy, 260, 1-13.

Chauvy R., Verdonck D., Dubois L., Thomas D., De Weireld G., 2021, Techno-Economic Feasibility and Sustainability of an Integrated Carbon Capture and Conversion Process to Synthetic Natural Gas, Journal of CO2 Utilization, 47, 1-10

Davis W., Martín M.,. 2014, Optimal Year-Round Operation for Methane Production from CO_2 and Water Using Wind and/or Solar Energy, Journal of Cleaner Production, 80, 252-261.

Giglio E., Lanzini A., Santarelli M., Leone P., 2015, Synthetic Natural Gas via Integrated High-Temperature Electrolysis and Methanation: Part I-Energy Performance, Journal of Energy Storage, 1, 22-37.

Ipsakis D., Varvoutis G., Lambropoulos ., Papaefthimiou S., Marnellos G.E., Konsolakis M., 2021, Techno-Economic Assessment of Industrially-Captured CO_2 Upgrade to Synthetic Natural Gas by Means of Renewable Hydrogen, Renewable Energy (accepted)

Ipsakis D., Kraia T., Konsolakis M., Marnellos G., 2018, Remediation of Black Sea Ecosystem and Pure H2 Generation via H2S-H2O Co-Electrolysis in a Proton-Conducting Membrane Cell Stack Reactor: A Feasibility Study of the Integrated and Autonomous Approach, Renewable Energy, 125, 806-818.

Varvoutis G., Lykaki M., Stefa S., Binas V., Marnellos G.E., Konsolakis M., 2021, Deciphering the Role of Ni Particle Size and Nickel-Ceria Interfacial Perimeter in the Low-Temperature CO_2 Methanation Reaction over Remarkably Active Ni/CeO2 Nanorods, Applied Catalysis B: Environmental, 297, 1-14.