Why Turquoise Hydrogen Will Be a Game Changer for The Energy Transition

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Abstract

With hydrogen being promoted as a promising energy vector for a decarbonized world, low-carbon hydrogen production methods are of interest to replace the current Steam-Methane-Reforming production of "grey" hydrogen. While existing studies focus on the life-cycle-assessment of green hydrogen produced by water electrolysis, an alternative, which has attracted growing interest due to a much lower energy intensity (10-30 kWh/kgH2 < 50-60 kWh/kgH2), is turquoise hydrogen produced by the pyrolysis of methane. Specifically, this study conducts a life-cycle-assessment on hydrogen produced by the pyrolysis of methane via thermal-plasma. A sensitivity analysis is also conducted on the environmental-metric time-horizon and on the methane emissions rates. Results show that the carbon-intensity of hydrogen produced using this novel method is 88.3-90.8% lower than that of grey hydrogen. Furthermore, using renewable-natural-gas with a feedstock percentage as low as 8-18% leads to a negative hydrogen carbon-intensity (reaching -4.09 to -10.40 kgCO2e/kgH2 at 100% renewable natural gas), the lowest compared to grey, blue, and green hydrogen, making turquoise hydrogen a game-changer for the energy transition.

Keywords: Hydrogen, Thermal Plasma, Methane Pyrolysis, Life Cycle Assessment, Natural Gas, Renewable Natural Gas

1. Introduction

With the latest release of the Intergovernmental Panel on Climate Change (IPCC)'s contributions for the sixth climate change assessment report, strong statements have been made on the role of human activity on global warming [1]. With that, the IPCC emphasizes in their report the urge of reaching at least net-zero CO2 emissions to limit global warming effect.
Several strategies have been assessed to reduce CO2 emissions, with hydrogen being promoted as a potential energy carrier for a carbon-neutral world. The Hydrogen Council projects a significant growth of hydrogen demand by 2050, with the role of hydrogen being distributed in large-scale power generation, transportation, industry energy use, and building heating [2]. With the advancements and projected growth of hydrogen in several sectors, it is imperative to discuss its production methods to evaluate carbon footprints.
According to the World Energy Council in 2019, 96% of hydrogen is produced from fossil fuels either through steam methane reforming (SMR) of natural gas (labelled "grey" hydrogen) or from coal gasification (labelled "brown" hydrogen) [3]. Consequently, hydrogen production is responsible for 830 million tons of carbon dioxide per year [4]. It is obvious that a transition towards hydrogen with the current production share is not an environmentally sustainable solution [5]. Thus, other production methods are gaining interest, mainly hydrogen produced from water electrolysis. If electricity is supplied by renewable energy, then this hydrogen is labelled as "green" as it has a low GHG emissions profile [6].
However, the problem lies in the cost of such electrolytic hydrogen. An intermediate lower cost solution to improve the carbon footprint of hydrogen production than electrolytic hydrogen, is to couple the SMR of natural gas or coal gasification with a CO2 capture and storage, or what is known as "blue" hydrogen [7]. The role of blue hydrogen in the energy transition is a controversial topic: proponents portray it as a cost-effective low-carbon hydrogen production [8] while opponents of blue hydrogen argue on its environmental incentive, claiming that if taking methane emissions into consideration, then blue hydrogen is merely "a distraction, something that may delay needed action to truly decarbonize the global energy economy" [9].
Another alternative that is most often disregarded in environmental studies is turquoise hydrogen, i.e. hydrogen made from the pyrolysis of methane at high temperature for the co-production of hydrogen and carbon black. Therefore, turquoise hydrogen is based on direct methane decomposition (DMD), a process that has been put forward in the 2000s by the father of DMD, Muradov [10-13]. The main advantage of turquoise hydrogen is that it is significantly less energy intensive compared to water electrolysis and SMR from a thermodynamic perspective [14], and benefits from the existing infrastructure of natural gas [15].
Several turquoise hydrogen production methods were investigated throughout the years, such as catalytic and non-catalytic thermal decomposition/pyrolysis, molten metal or salt bath, concentrated solar, and non-thermal plasmas, with each having

its own drawbacks preventing an industrial scale adoption of such technologies. For instance, combustion-based processes that are long used in industries to produce "thermal black" [16] consume part of the produced hydrogen to increase thermal blacks' surface area. As for catalytic thermal decompositions, problems were encountered on industrial scale production due to the burning of the carbon deposited on the catalyst during regeneration [17]. Regarding molten metal or salt bath, its relatively low maximum temperature does not permit a high carbon conversion rate [18]. Concentrated solar power methods never reached industrial scale due to their high cost, and non-thermal plasma technologies encountered technological problems on the industrial scale, producing a low-value carbon (Atlantic Hydrogen).
However, methane pyrolysis via thermal plasma has been recently gaining interest [19]. Thermal plasma converts electrical energy to thermal energy, reaching temperatures that cannot be achieved in traditional combustion processes, reaching several thousands of degrees [20-21]. They are particularly interesting for endothermic processes for their tuneable enthalpy and the absence of direct CO2 emissions in the process itself [20]. Among the main advantage of this technology for this particular application is the high methane conversion with hydrogen as a major product [18].

After discussing different methane pyrolysis technologies, it is important to note that very little information exists academically on the environmental benefits of hydrogen produced via thermal plasma pyrolysis of methane. In fact, most life-cycle assessment of hydrogen targeted green hydrogen, and to a lesser extent, blue hydrogen.

For the case of green hydrogen, the majority of studies focused on comparing water electrolysis to conventional production methods such as SMR. For instance, Bhandari et al. reviewed twenty-one LCA studies of hydrogen production methods, with most of the studies comparing electrolytic hydrogen to conventional methods such as SMR or coal gasification, with or without carbon capture and storage [6]. Values among the different reviewed LCA studies vary, partly due to using different assumptions, and partly due to data change among the years, but the common conclusions are that water electrolysis using a dirty electricity mix is the worst from an ecological point of view, and hydrogen from wind electrolysis or thermochemical water decomposition has the best environmental performance. Other than the variability in data and assumptions taken, the main limitation of these studies is that not all hydrogen production methods are taken into account, and in particular, methane decomposition/pyrolysis is largely disregarded. Few studies seemed to favor thermochemical water splitting/decomposition as the most attractive hydrogen production method [22-23]. However, both studies present data from around 10 years, and for Smitkova et al.'s study, data is taken from "hypothetical" plants with Aspen simulations.

For the case of blue hydrogen, an interest in LCA of blue hydrogen is growing in the recent years, with one recent example being the work of Howarth and Jacobson [9]. In fact, some researchers argue that several blue hydrogen configurations can help in reducing carbon emissions, with a 2020 study showing that reductions in CO2eq emissions were around 50 to 85% when using CCS for a 100-year global warming potential (GWP) [24]. On the other hand, Howarth and Jacobson argue that blue hydrogen is "best viewed as a distraction, something that may delay needed action to truly decarbonize the global energy economy" [9]. The large gap in results of both views lies in the assumptions used in modelling, mainly in taking a 100-year GWP vs a 20-year GWP, and in the choice of methane leakage percentage and carbon capture percentage.
Regarding the main two parameters affecting LCA performance of blue hydrogen, the GWP choice and the methane leakage percentage:

a. To take different GHG to assess their aggregated total contributions, the common approach is to express them as CO2 equivalent through a GWP. Thus, a duration over which a GWP is expressed needs to be carefully considered, balancing between short term and long-term warming target. The two most common durations are either a 100-years horizon (GWP-100) or a 20-years horizon (GWP-20), with the latter putting more weight on GHGs with shorter lifespan, such as methane, that has a mean lifetime of 11.8 years [1]. In fact, in the IPCC's working group I's contributions for the sixth climate change assessment report, methane is argued to have an 82.5 gCO2 equivalent using GWP-20, and a 29.8 gCO2 equivalent using GWP-100 [1]. Therefore, in the long term, GWP-20 overestimates the response. To conclude on the GWP choice, it is worthy to mention that the latest published contribution of the IPCC on the sixth climate change assessment report does not recommend a particular GWP metric, leaving the choice for a political decision, but the GWP-100 is the adopted one for the UNFCC and Paris Agreement studies.

b. As for the discrepancy in the methane leakage percentage reported in studies, this is largely due to the location of the study itself, and the assumptions used for several areas of the natural gas supply chain, such as production, processing facilities, pipelines, storage and local distribution. Even on a global level, values seem to differ. For the United States, a study conducted by the Environmental Defense Fund (EDF) from 2012 to 2018 estimates methane leaks to be around 2.3%, which is 60% higher than the Environmental Protection Agency (EPA)'s leak rate of 1.4% [25]. The discrepancy in the hypotheses is due to the fact that methane leaks are measured in "bottom-up" readings based on activity data and emission factors, and "top-down" readings based on satellites or aircrafts observations combined with transport models. The first tends to underestimate emissions, while the latter is not effective at pinpointing emission sources [25]. Thus, many uncertainties are present in the measurement of methane emissions, mostly due to the inaccuracies of measuring methods, and to the variability in emissions for different locations and supply chains. However, promises are being made to reduce these emissions by more than 70% by 2030 under the IEA's Sustainable Development Scenario with the IEA having developed a regulatory toolkit to help actors of the sector reach this goal [26].

Lastly, for the case of turquoise hydrogen, one disregarded method in LCA studies for hydrogen production is methane decomposition/pyrolysis with its several technologies, possibly due to a lack of key-data. Methane decomposition for hydrogen and carbon coproduction has been assessed throughout the years from an energetic and economic perspective, but rarely from an environmental perspective over a life cycle [27-28]. In fact, very few studies conducted an LCA for methane decomposition [29-34]. The main contributions came from a collaborative work between the ESCET of the Universidad Rey Juan Carlos and the National Institute of Aerospace Technology (INTA) with a series of studies published between 2009 and 2012 [29-32]. The first publication reports an LCA comparing both thermal and autocatalytic (where the produced carbon is used as a catalyst) decomposition of methane to SMR and SMR coupled with CCS [29]. Results favored the autocatalytic decomposition by having the lowest environmental impact. After this publication, a follow-up study reports an LCA to compare methane decomposition with carbonaceous catalysts with decomposition using metallic catalysts, and with thermo-catalytic decomposition and SMR with CCS [30]. The last two studies of this collaborative work assessed the life-cycle of methane decomposition compared to additional processes such as autothermal reforming of hydrocarbons and coal gasification [31] and water photo-splitting and solar thermochemical cycle [32]. However, there is no clear data in all these studies on the choice of parameters for the supply chain of the natural gas and on the methane leakage percentage used as this is one of the most debated parameters in such LCA studies.
Another technology of methane decomposition/pyrolysis, the liquid-metal technology for methane decomposition, was assessed from an LCA perspective in 2016 by Postels et al [33]. Compared to SMR, the authors argue that methane cracking can reduce the global warming impact by 64%. Results also show a better performance compared to water electrolysis using wind energy, but a higher fossil depletion impact. However, the main limitation of this study is in the choice of data and assumptions, as for the industrial scale data, a "hypothetical" scenario is used where assumptions are used for a future scale-up according to laboratory experimental data.
Finally, when it comes to methane pyrolysis via thermal plasma, the main contributions came from one study only [34] where an LCA for the GHG emissions and levelized costs of hydrogen is conducted for methane pyrolysis via plasma, molten metal, and thermal gas. The source of electricity, along with the natural gas supply chain parameters are found to be critical factors. Using a mean CO2 emission for the natural gas supply chain based on four main supply chains (Russia, Middle East, Norway, and LNG transport), and assuming a 1.7% methane leakage, results show that the lowest GHG emissions are for electrolysis using renewable energy, followed by plasma pyrolysis of methane using renewable energy.
However, it is worthy to note that in all LCA studies targeting methane pyrolysis, the co-produced carbon is assumed as non-usable, and thus the worst-case approximation is taken: all environmental impacts are weighted on hydrogen. The reason for that is the fact that only the process assessed in this study, the Monolith methane pyrolysis process, allows the co-production of industrial grades of carbon-black that are of value.

The review of the literature focuses mainly on the common methods for the production of green, blue, and turquoise hydrogen, as these methods are the most adopted when it comes to hydrogen production.
It is worthy to note however, that other emerging technologies for hydrogen production are being developed such as aqua-hydrogen (hydrogen extracted from oil sands (natural bitumen) and oil fields) [35]. Additionally, the focus of this study is on the environmental impact of the hydrogen production method, and not on its economic impact. Turquoise hydrogen, characterized by a relatively low cost of production, is attractive compared to other hydrogen production methods when comparing its production cost to the reported cost of blue and green hydrogen in Ajanovic et al.'s article [36].

Overall, based on the main findings of the literature, the following gaps and limitations are identified:

- There is a considerable variability in data among the studies, partly due to conducting studies in different locations and regions, and partly due to the change of data throughout the years
- No large-scale plants for electrolysis or methane pyrolysis were used to collect operating data and plant specifications
- The choice of methane leakage percentage is controversial, as some researchers argue that the average value is underestimated, and others, that it is overestimated
- More focus should have been given on the natural gas supply chain assessment and data choice in the studies (especially for methane leakage rate), as it considered a critical factor in the global GHG emissions of methane pyrolysis
- The co-produced carbon black is assumed in the studies to be non-usable, as only the process assessed in this study (the Monolith methane pyrolysis process) allows the co-production of industrial grades of carbon-black that are of value. Thus, all the environmental impacts in previous studies are ascribed on the hydrogen product
- Production of hydrogen through the pyrolysis of methane using thermal plasma is largely disregarded in the studies, even though it is the most promising methane pyrolysis technology
- The metric to measure the global warming potential is not unified, with each study choosing a metric that greatly impacts its conclusions

Based on the gaps and limitations of the literature, this paper reports the first LCA study of methane pyrolysis via thermal plasma, aiming to answer the following question: why turquoise hydrogen will be a game changer for the energy transition?

2. Methods

2.1. Greet Description

Life-cycle assessment of methane pyrolysis via thermal plasma is conducted using "Greenhouse gases, Regulated Emissions, and Energy use in Technologies Model" (GREET), an analytical tool developed by Argonne National Laboratory [37-39]. GREET is the chosen interface to conduct a cradle-to-gate LCA of the process products, as it is widely recognized in LCA among several regulatory agencies such as the US Environmental Protection Agency (EPA) and the California Air Resources Board, notably in the Low Carbon Fuel Standard (LCFS) program [40-41]. Its main advantage compared to other LCA tools is the built-in models of several pathways for the US. A description of the canonical process conversion that GREET uses is presented in the "Method" section of the supplemental information document S1.

2.2. Process Flow Diagrams

The production of hydrogen through methane pyrolysis via thermal plasma is presented in Figure 1A.
A mixture of natural gas and other hydrocarbons is used as feedstock to the thermal plasma reactor. The reactor, using electricity to create a high-temperature plasma arc, pyrolyzes the feedstock, meaning that CH_4 is decomposed into carbon black "C" (a nanostructured material composed of small crystallites, and having a carbon content higher than 95% by elemental composition), and hydrogen "H_2".
Therefore, instead of combustion, the feedstock is pyrolyzed meaning that there is no direct CO_2 emissions, but rather a production of "C" by direct decomposition of methane.
Once the feedstock pyrolyzed, a very small quantity of metallurgical coke is produced, and the main resultant stream containing "C" and "H_2" is split into two chains:

- one goes to the "carbon black back-end" unit consisting of filtration, pelletizing, drying, and other processes, producing carbon-black
- the other chain goes to a hydrogen purification unit, producing purified hydrogen using a Pressure Adsorption Swing unit (PSA). The PSA technology uses the binding of gas molecules to adsorbent material so separate impurities from the main stream, leading to a high-pressure hydrogen stream and a flue gas that contains impurities and is then cleaned up.

A "Balance of Plant" (BOP) box is also portrayed in Figure 1A, aggregating all supporting systems and utilities that are not unit operations in the primary process flow (such as cooling towers, generators, fire-pumps, etc.).
The data for the whole process is detailed in the following subsection 2.3 "Data". The presented method consumes in total 364.6 Mgal of water per year, for the coproduction of 42.3 kilotons of hydrogen and 180 kilotons of carbon-black.

To assess the environmental performance of methane pyrolysis via thermal plasma, it is critical to compare hydrogen production with carbon black co-production to their respective conventional processes. Both conventional processes are presented in Figure 1B.

- For hydrogen, the reference process for comparison is Steam Methane Reforming (SMR). SMR catalytically converts hydrocarbons, in particular methane, and steam to hydrogen and carbon oxides. The process involves reforming (syngas generation), water-gas shift, and gas purification. As mentioned in the "Introduction" section, most of the global supply of hydrogen is produced using SMR, having a relatively low production cost, but poor environmental performance, as will be assessed in the coming sections.
- For carbon-black, the conventional process consists of thermally decomposing oil in a reactor at high temperatures using fuel and air, leading to an incomplete combustion forming carbon-black. Produced carbon-black is in powder form, and mixed with water inside a pelletizer, can be processed into pellets. By-product gases can be separated into filters and dried for reuse as fuel in the plant. It is worthy to note that most of today's carbon-black is produced using incomplete combustion.

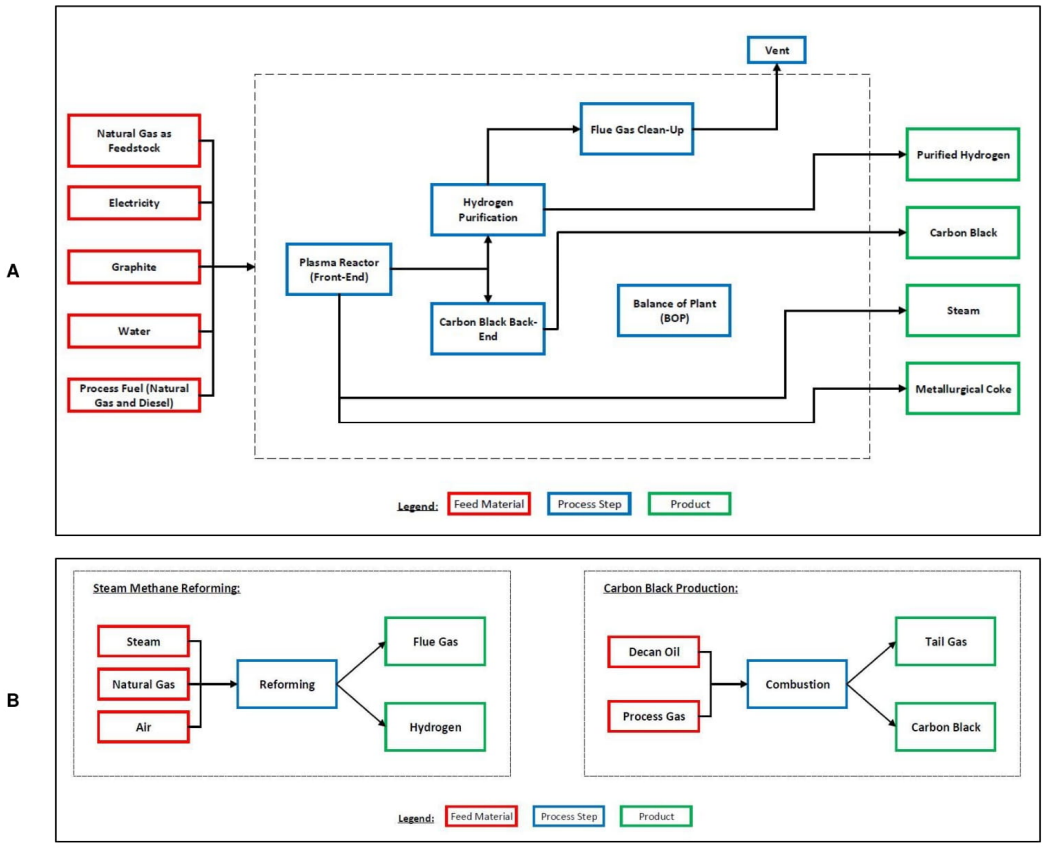

Figure 1. Process Flow Diagrams

(A) Methane Pyrolysis via Thermal Plasma
(B) Conventional Processes for Hydrogen and Carbon Black Production

2.3. Data

As mentioned in the synthesis of the introduction section, one of the main limitations of the assessed LCA studies is that the data used is mostly generated from laboratory-scale experiments or simulations. In practice, large-scale plants would have different values.

For this study, a commercial plant is used as the study case, with data taken from real-time operation for accuracy. The chosen plant is the only commercial plant available using thermal plasma to pyrolyze methane for hydrogen and carbon black production: Monolith Materials' Olive Creek plant.

Data used is presented in Table 1, with an annual production of 42.3 kilotons of hydrogen and 180 kilotons of carbon-black. Additionally, 10 kilotons of $CO2_e$ are added as direct emissions representing emissions emitted from the flare stack.

In theory, using molar masses of CH4, C, and H2 when pyrolyzing one mole of CH4 to co-produce 1 mole of carbon-black and 2 moles of hydrogen, the produced mass of hydrogen should be 1/3 of that of carbon-black. The reason for having a lower ratio of hydrogen to carbon-black in practice is, in addition to losses, the internal usage of part of the produced hydrogen in the process itself, such as for heat supply, etc.

Table 1. Olive Creek Plant Data (Gathered from Monolith Materials)

Process Stage	Consumption
Plasma Reactor (Front-End)	Natural Gas: 259 kT/y Various Consumables*: 803 T/y Electricity: 1077 GWh/y Water: 22 Mgal/y
Carbon Black Back-End	Electricity: 34 GWh/y Water: 65 Mgal/y
Hydrogen Purification	Electricity: 96 GWh/y
Flue Gas Clean-Up	Fuel: 626 mmbtu/y Natural Gas
Balance of Plant (BOP)	Electricity: 43.2 GWh/y Water: 277.6 Mgal/y Fuel: 545.6 mmbtu/y natural gas and 85.6 mmbtu/y Diesel

**various consumables include diverse reactor pieces including insulation materials, graphite elements, etc.*

2.4. Hypotheses

The modelled process is the process presented in Figure 1A, with the GREET2020 version used for calculations.
Several hypotheses and assumptions were made, based on the GREET2020 average values for the US [38-39], and based on the California Air Resources Board's Low Carbon Fuel Standard program's models [40-41], as the chosen location is California.
Below are the hypotheses and assumptions for each phase of the process:

1. Natural Gas (NG) Supply Chain:

- Regarding the natural gas composition, a mix of 50.2% of shale formations and 49.8% conventional wells is used, based on the California Air Resources Board's models [41]
- Using the California Air Resources Board's hypothesis that 10% of natural gas is produced in state and 90% is imported from basins from Western Canada to Texas, the assumed average pipeline transportation distance is 575 miles
- The pipeline transport fuel share is taken as 98% natural gas and 2% electricity, being the default value for the US natural gas pipeline transport used in GREET [42]
- When required, the electricity in the whole natural gas supply chain is used from the average US grid mix electricity, as the global chain passes through different states. The GREET average US grid mix has a carbon intensity (CI) of around 0.14 kgCO2e/MJ of electricity, with the US grid mix taken from the EIA's 2021 annual energy outlook [43]
- Methane content is taken as 92% of the gas, and the emissions of methane based on the literature review's "life cycle assessment" section is taken for the base case as 1.5% of methane emitted from the total methane produced

2. Plasma Reactor:

- Graphite used for the electrodes is assumed to be produced according to GREET's only available graphite model made primarily out of pet coke, having a carbon intensity of around 5 kgCO2e/kg graphite [44]

3. Electricity:

- For all stages of the process, electricity used in the plant is taken as a low-carbon renewable electricity (RE), notably "wind power", assumed to have a 10 gCO2e/kWh carbon intensity based on Bhandari et al.'s sensitivity analysis [45]

4. Process Fuels:

- The natural gas used as fuel in some of the processes (such as in the flue gas clean-up unit) is taken as the same natural gas that arrives to the Olive Creek plant, but has a higher carbon intensity due to combustion
- Diesel used as process fuel in the "balance of plant" aggregated processes (that includes supporting systems and utilities that are not unit operations in the primary process flow) is taken as the conventional diesel from crude oil for the US, a model already designed on GREET

2.5. Allocation Methods for Carbon Intensity Calculations

As mentioned in the synthesis part of the introduction, the co-produced carbon black is assumed in all assessed studies to be non-usable, therefore putting all the environmental impacts on hydrogen, as only the process assessed in this study (the Monolith methane pyrolysis process) allows the co-production of industrial grades of carbon-black.
In fact, carbon-black is a growing market with a compound annual growth rate (CAGR) of 5.15%, valued at $15.5 billion with several applications in tires, rubber plastics, toners, paints and textile fibers [46].
However, as previously mentioned, the mass of carbon-black produced is more than three times more than that of hydrogen. Therefore, industrial applications for carbon-black may not be enough on their own. For this, several other applications of carbon-black are currently under assessment. While this study focuses on the first generation of the process where carbon-black is directly used for its current industrial applications (tires, rubber plastics, toners, paints and textile fibers), in the near

future, carbon-black can be used in building materials and infrastructures (cement, roads, etc.), and on the long-term, carbon can be used as an amendment in soils.

Thus, by producing a usable coproduct (carbon-black), and like any life-cycle analysis of a process having more than one output, allocation methods need to be implemented to distribute the total emissions among the product and coproducts.
To remove any bias from this study towards one product or coproduct, both economic and mass allocations methods are used for the base case.
However, with the current rapid change of hydrogen and carbon black prices due to the effect of Covid-19 on the supply chain, the economic allocation method is relatively unstable. Thus, the mass allocation method is emphasized in the alternative scenarios, leading to more stable number allocation.

1. Economic Allocation:

The economic allocation is based on both the market price and the produced mass of each product/coproduct, distributing the environmental impact among them according to equation (1):

$$w_i = \frac{P_i . M_i}{\sum_j^n P_j . M_j} \quad (1)$$

With w the allocation factor of the product/coproduct, P_i the price of product/coproduct i, M_i its mass, j the product/coproduct and "n" the total products/coproducts.
The data used along with the weight results is presented in Table 2:

Table 2. Economic Allocation Data (from Monolith Materials) and Results

Item	Unit	Price	Allocation Factor
Hydrogen	$/ton	1,800	23.94%
Carbon Black	$/ton	1,343	76.01%
Coke	$/ton	50	0.05%

2. Mass Allocation:

The mass allocation method is only based on the mass of the products and coproducts, therefore distributing the environmental in an equal way among the outputs, as is shown in equation (2):

$$w_i = \frac{M_i}{\sum_j^n M_j} \quad (2)$$

The data used along with the weight results is presented in Table 3:

Table 3. Mass Allocation Data (from Monolith Materials) and Results

Item	Unit	Mass	Allocation Factor
Hydrogen	ton	42,300	18.77%
Carbon Black	ton	180,000	79.89%
Coke	ton	3,000	1.33%

3. Carbon Intensity Calculation:

Finally, once the allocation method chosen, the respective allocation weight factor for each product and coproduct "w" is used to find its allocated emissions. Dividing these allocated emissions to the product mass output gives its carbon intensity, using equation (3):

$$CI_i = \frac{E_i}{M_i} \quad (3)$$

$$E_i = w_i . E_t \quad (4)$$

with "CI_i" the carbon intensity in kgCO2e/kg of product "i", "E_i" the GHG emissions in kgCO2e of product "i", "M_i" the mass of the product/coproduct, and "E_t" the total process emissions in kgCO2e.

2.6. Environmental Metrics

To assess the environmental impact of the process, a common approach is to convert greenhouse gases such as CH4 and N2O to CO2 equivalent according to a global warming potential (GWP) metric. Several GWPs were proposed throughout the years, as discussed in detail in the introduction, with the most common GWP duration being a 100-years horizon or a 20-years horizon. The difference in the metric duration mainly affects the CH4 conversion, as methane has a mean lifetime of around 11.8 years [1], therefore its environmental impact for a 100-years horizon is much different than for a 20-years horizon.
As discussed in Introduction section, the common approach is to use the GWP-100, as most studies are conducted using this metric, making it easier to compare this study's results with other studies. Moreover, the GWP-100 is adopted in major studies and reports, such as the UNFCC and Paris Agreement studies.
However, to remove any bias towards hydrogen production through methane pyrolysis, the GWP-20 is also used in a sensitivity analysis that is presented in this work.
Both the GWP-100 and the GWP-20 metric values are taken from the latest IPCC's working group I's contributions for the sixth climate change assessment report, AR6, and are presented in Table S1 of the Supplemental Information Document S1.

2.7. Scope Definition

An important part of any LCA study is the definition of the scope. The International Organization for Standardization highlights the importance of well defining the scope based on parameters such as the function of the system, the system's boundaries, the data and assumptions, etc. according to the ISO 14040 [47], whereas the World Business Council for Sustainable Development and the World Resources Institute, through the GHG Protocol [48], identify three scopes for LCA studies, with scope 1 including direct emissions, scope 2 including purchased electricity and heating and cooling resources, and scope 3 including indirect emissions.
For this study, scopes are defined as follows:

1- Scope 1: including the process' direct emissions only
2- Scope 2: including indirect emissions related to the electricity purchased
3- Scope 3: including indirect emissions associated with natural gas sourcing

2.8. Scenarios

As a baseline case, methane pyrolysis via thermal plasma is assessed, as presented previously in Figure 1A, using fossil natural gas (shale and conventional). The baseline case is then compared to conventional processes of hydrogen and carbon-black production, using SMR and incomplete oil combustion respectively, as presented in Figure 1B.
For the alternative scenario, fossil natural gas (FNG) in the methane pyrolysis via thermal plasma is replaced with renewable natural gas (RNG). A sensitivity analysis is conducted, changing the percentage of renewable natural gas used as feedstock in the process from 0 to 100%. Other parameters to which a sensitivity analysis is conducted are the environmental metric, going from GWP-100 to GWP-20, and the methane emissions percentage that is initially taken as 1.5% as discussed in the "Hypotheses" subsection.
A summary of the different scenarios and sensitivity parameters is presented in Figure 2.

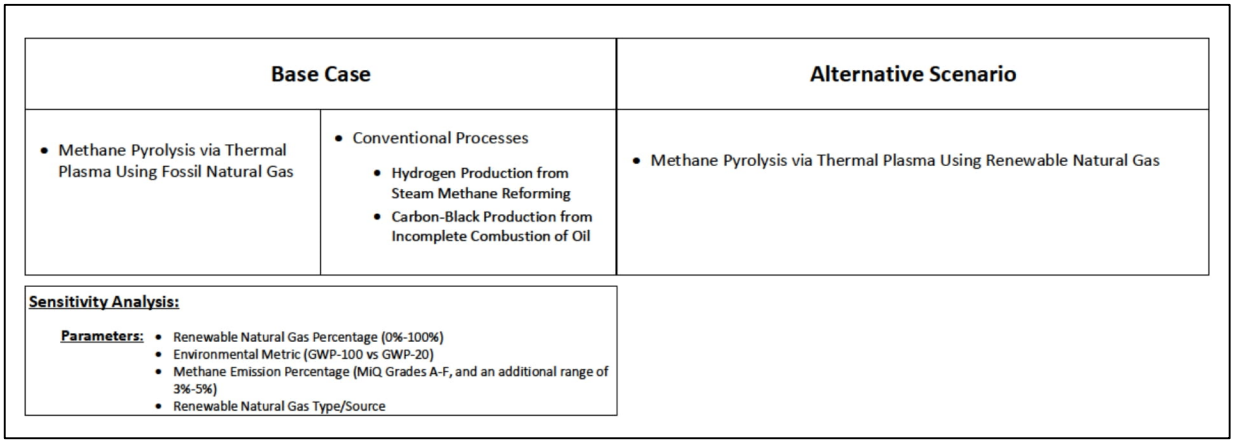

Figure 2. Scenarios Assessed

3. Results

3.1. Base Case

Using the data and hypotheses presented in the "Methods" section, methane pyrolysis via thermal plasma is modelled using GREET2020. The carbon intensity of the main product, hydrogen, along with the main coproduct, carbon-black, and the small

co-produced quantity of coke, is calculated using both the economic allocation method and the mass allocation method. Both allocation methods were presented in the "allocation methods for carbon intensity calculations" subsection, and calculations are carried on using both methods for the base case to remove any bias towards methane pyrolysis via thermal plasma. For the alternative cases and sensitivity analyses, only the mass allocation method is retained as, in light of what was previously discussed in the "Allocation Methods for Carbon Intensity Calculations" subsection, with the current rapid change of hydrogen and carbon black prices due to the effect of Covid-19 on the supply chain, the economic allocation method is relatively unstable.

First, the carbon intensity (CI) is calculated for scopes 1, 2, and 3, as described in the "Scope Definition" subsection, and results are presented in Figure 3A and figure 3B using the economic and mass allocation method respectively, using the baseline assumptions previously discussed such as having 1.5% methane emissions and using the GWP-100.

For the economic allocation method, hydrogen has the biggest share of emissions among the other outputs, as despite having a low allocation weight factor compared to carbon-black for instance, it has the highest weight-factor-to-mass ratio, leading to the highest carbon intensity among the process outputs, as can be shown using equations (3) and (4).

For the mass allocation method, as expected, considering that the weight factor is only related to the product/coproduct's mass (equation (2)), and the carbon intensity then divides the emissions by the respective masses, then all the output will have the same carbon intensity.

Using both methods, scopes 1 and 2 lead to very low hydrogen carbon intensity, reaching a maximum of 0.15 kgCO2e/kg for scope 2 for the economic allocation method, with scope 1 + scope 2 CI almost the double of scope 1 CI.

On the other hand, the CI of scope 1 and scope 2 together is around 13% of the aggregated CI of all three scopes together for both methods, with scope 3 containing emissions associated with natural gas sourcing. Thus, we can conclude that the largest share of emissions for methane pyrolysis via thermal plasma is due to the sourcing of the feedstock natural gas. For the rest of this study, the cumulative emissions of the process are used (scope 1 + scope 2 + scope 3).

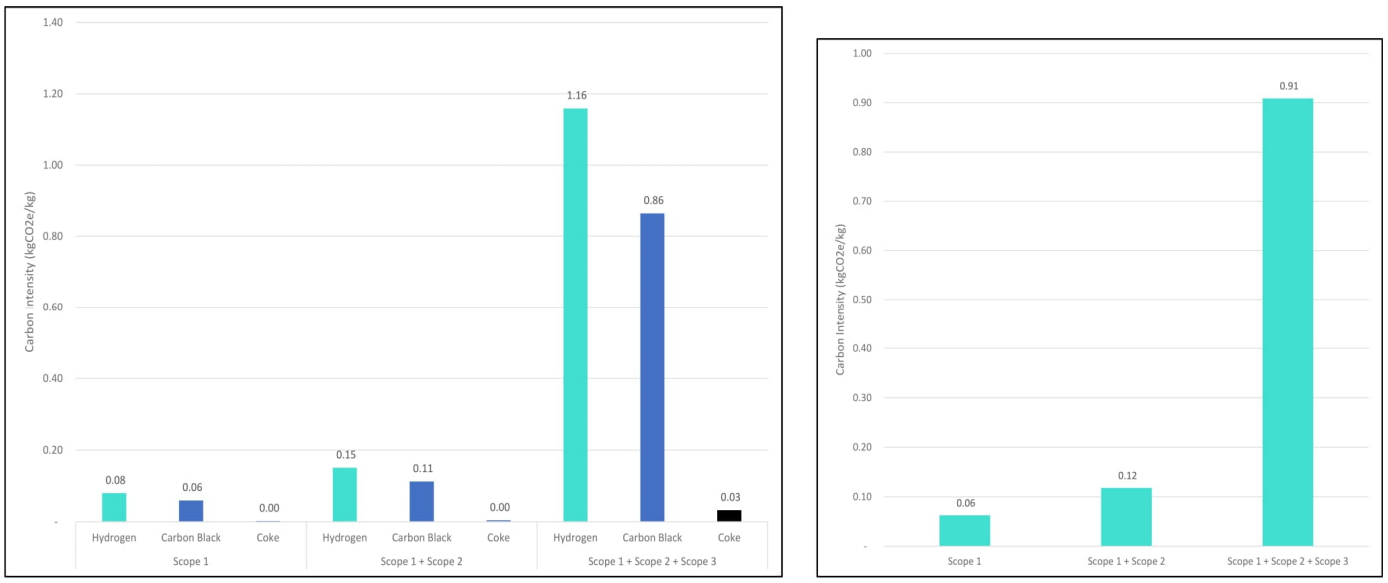

Figure 3. Carbon Intensity of Hydrogen, Carbon-Black, and Coke, for Methane Pyrolysis via Thermal Plasma

(A) Using the Economic Allocation Method Based on Scope 1, Scope 2, and Scope 3
(B) Using the Mass Allocation Method Based on Scope 1, Scope 2, and Scope 3
Detailed results per process phase are reported in Table S2 of the supplemental information document S1

In addition to calculating the carbon intensity of hydrogen and carbon-black for Methane Pyrolysis via thermal plasma based on the defined scopes, these carbon intensities are also compared to conventional processes as described in figure 1B, notably SMR for hydrogen, and coke production from coil.

Conventional processes are modelled on GREET using the default US average GREET models and using the same hypotheses used for methane pyrolysis via thermal plasma for comparison.

For the conventional carbon-black process, emission values for the furnace black process with thermal treatment are taken from the "IPCC Guidelines for National Greenhouse Gas Inventories" third volume [49].

Results are shown in Figures 4A and 4B for economic and mass allocation methods respectively.

For both cases, using the baseline assumptions such as having 1.5% methane emissions and using the GWP-100, ethane pyrolysis via thermal plasma performs significantly better from an environmental perspective, as the produced hydrogen has 88.3% less GHG emissions compared to SMR using the economic allocation method, and 90.8% less GHG emissions using the mass allocation methods. The co-produced carbon black also has a better environmental performance, with 67.0% less emissions using the economic allocation method, and 65.3% less emissions using the mass allocation method.

This can be traced back mainly to the fact that the heat supplied to the methane pyrolysis is provided using thermal plasma

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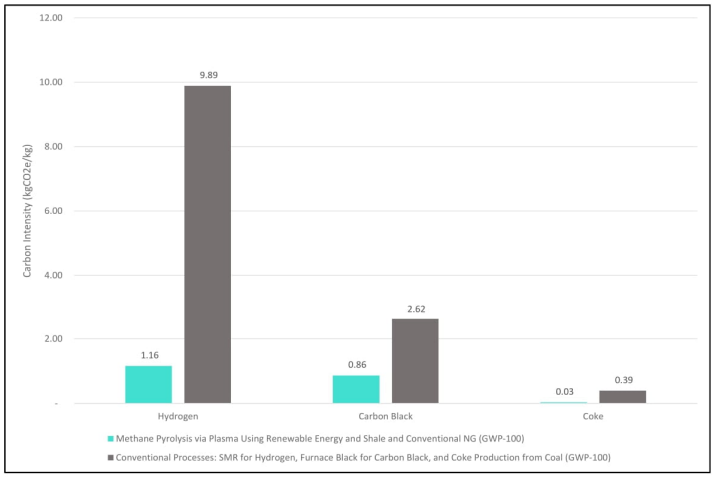

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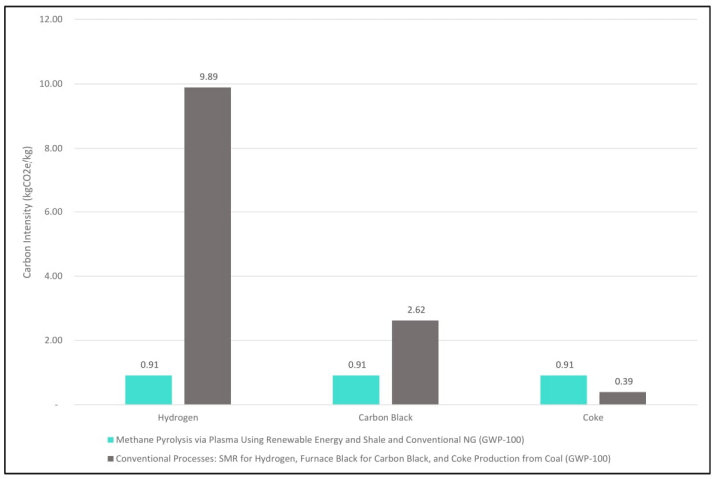

Figure 4. Carbon Intensity of Hydrogen, Carbon-Black, and Coke, for Methane Pyrolysis via Thermal Plasma and for Conventional Processes

(A) Using the Economic Allocation Method
(B) Using the Mass Allocation Method

produced from a low-carbon electricity. Furthermore, methane is pyrolyzed in the process and not combusted, leading to the carbon in CH4 being converted to carbon-black and not CO2, reducing the overall GHG emissions of the process.

3.2. Alternative Scenario (Using Renewable Natural Gas)

As discussed in the "Scenarios" subsection, and shown in Figure 2, an alternative case is also modelled and assessed, identical to the base case, but using renewable natural gas instead of the shale and conventional natural gas.
The chosen renewable natural gas is the renewable natural gas produced from food waste using anaerobic digestion, as it has a great potential of supplying renewable natural gas in the future. The GREET's available model is chosen [50-51]. Before being transported, the renewable natural gas passes through a biogas upgrading unit, after which it leaves with a high carbon ratio of 72.4%. The biogas upgrading unit is used to upgrade the biogas quality so that it becomes fully interchangeable with conventional natural gas.
Similar hypotheses are used for RNG to compare results, and leak is also accounted for in the process (transportation, etc.). However, in addition to the methane leakage, biogenic CH_4 is also released in the atmosphere.
Renewable natural gas percentage is varied from 0 to 100% (with the remaining percentage, when needed, taken as the baseline case's shale and conventional natural gas).
As previously discussed, only the mass allocation method is used as it leads to more stable numbers with the current instability of the supply chain. Results are shown in Figure 5.
When blending the feedstock with RNG having a negative carbon footprint (- 4.55 kgCO2e/kg natural gas), the carbon intensity of the process products becomes negative at a relatively low renewable natural gas percentage (between 14% and 15% for hydrogen).
The reason for this is that, as shown in the base case results, most of the process emissions occurs at the natural gas supply chain level, due to the production and transportation of natural gas, and due to the methane leakage. Replacing the natural gas supply chain with a sustainable, renewable natural gas produced by anaerobic digestion of food waste greatly reduces the overall process emissions and even leads to negative emissions depending on the renewable natural gas percentage.

Going to a fully renewable natural gas feedstock leads to a hydrogen carbon intensity of -5.22 kgCO2e/kgH2 for the mass allocation method with the GWP-100, meaning that producing hydrogen using methane pyrolysis via thermal plasma with renewable natural gas contributes in reducing GHG in the atmosphere. For every kilogram of hydrogen produced using this method, 5.22 kgCO2e are removed from the atmosphere.

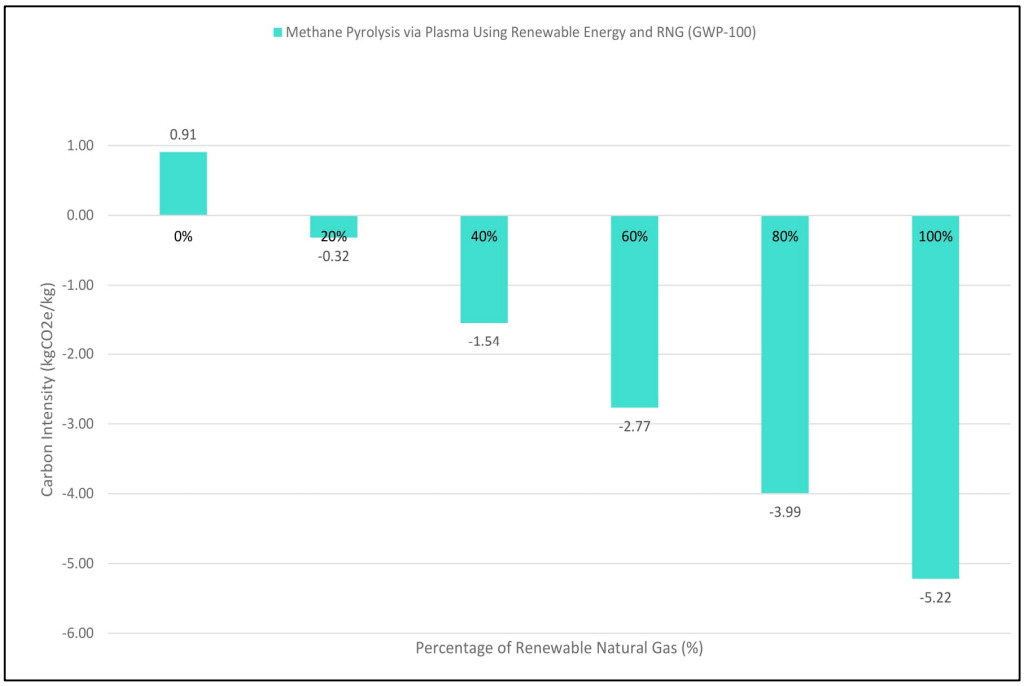

Figure 5. Variation of Carbon Intensity of Methane Pyrolysis via Thermal Plasma Products with The Variation of Renewable Natural Gas Percentage

Using the Mass Allocation Method

3.3. Sensitivity Analysis of Methane Emissions and GWP Metric

When it comes to LCA studies, many uncertainties arise, especially for emergent technologies, due to the assumptions inherent to data incompleteness on one hand [52-53], and due to the forecasting nature of such technologies on the other hand [54]. One of the strengths of this study is that it uses data gathered from an actual operating plant, Olive Creek, having a high technology readiness level, despite the fact that on a more general level, methane pyrolysis via thermal plasma can still be considered an emergent technology. However, a major source of uncertainty when performing LCA on such a system in particular, and on any process that includes natural gas in general, is the choice of methane emission percentage and of the environmental metric time horizon, as discussed in the introduction.

In light of the information presented in the literature review, both the base scenario and alternative scenario used the GWP-100 and 1.5% methane emissions out of the total produced methane.

However, to account for uncertainty and variability in methane emissions, a sensitivity analysis is conducted and presented in this section, varying the base case scenario's methane emissions for both GWP-100 and GWP-20.

Regarding the choice of the GWP time horizon, both the GWP-100 and GWP-20 are used in this part, based on the IPCC's AR6 values presented in Table S1 of the supplemental information document S1. As previously discussed, the choice is critical for such applications as methane, having a lifetime of around 11.8 years [1], has a significantly higher CO2e value when going from a 100-year horizon to a 20-year horizon. The reason for choosing GWP-100 for the base case is for its long-term suitability and for the fact that it is adopted for global reports including the UNFCC and the Paris Agreement studies. However, GWP-20 is also assessed, as some researchers such as Jacobson and Howarth argue that GWP-20 is better suited for applications including natural gas to account for its near-term impact [9].

Regarding the methane emission percentage, the percentage of methane emitted to the total methane produced, initially taken as 1.5% previously, is varied on one hand according to the MiQ certification standard grades A to F [55], and on the other hand, from 3% to 5% in steps of 0.5%.

The reason for choosing the MiQ emission standard grades is the popularity it is gaining among oil and gas operators, such as ExxonMobil and Bloomenergy [56-57], with MiQ certifying already 1.8% of the global gas market in their first year, with an aim of certifying 100% of the market in the next decade [46]. The MiQ standard grades are based on upstream carbon intensity, calculated as the percentage of methane emitted to the natural gas produced. For consistency with previous sections' calculations, MiQ's emission percentage is converted to a percentage of methane emitted to total methane produced, using the same methane content of 92% used in previous calculations. Emission percentage is shown in Table 4.

Table 4. MiQ Methane Emission Percentage

Grade	Upstream Emissions Threshold (methane emitted to natural gas produced)	Upstream Emissions Threshold (methane emitted to methane produced)
A	0.050%	0.054%
B	0.100%	0.109%
C	0.200%	0.217%
D	0.500%	0.543%
E	1.000%	1.087%
F	2.000%	2.174%

The main reason in assessing a better sourcing of natural gas is in the aim of reducing process emissions, in particular for hydrogen, in order to reach a target level of carbon intensity for hydrogen lower than 0.4-0.45 kgCO2e/kg, a target that is of particular interest for policy makers.

As for the reason in varying the methane emission percentage from 3 to 5%, it is to account for the uncertainty and variability regarding this percentage that has been discussed in the introduction. To reiterate, the Environmental Defense Fund estimates methane leaks to be around 2.3%, 60% higher than the Environmental Protection Agency's leak rate of 1.4%. On the higher end of the range, Howarth and Jacobson estimated the percentage of methane emissions as 3.5% of consumption [9], and argued that a 4.3% of consumption can be calculated from a sensitivity analysis study presented by Howarth for shale gas [58]. On the lower end of the range, a lower value can be found in a study reported by Alvarez et al. in 2018 [59], using a bottom-up approach to estimate emissions. An even lower value can be traced using satellite data in Maasakkers' study [60]. Regarding future projections, promises are made to reduce these emissions by 70% by 2030 under the IEA's Sustainable Development Scenario [26], and in the 2021 World Energy Outlook, it is argued that using leak detection and repair (LDAR) equipment and alternative technologies and policies could reduce these emissions by even 75% [61]. The bottom range values are already taken into account with the relatively low MiQ grade percentages, thus only the higher-end of the range is assessed separately.

Results for the MiQ Grades using the mass allocation method are shown in Figure 6A, and those for the upper range are shown in Figure 6B. Considering that it is hard to achieve low emission percentages on GREET without modifying the process, the CI values of MiQ Grades A to D were extrapolated.

As predicted, the use of GWP-20 leads to significantly higher carbon intensity for the products than when using the GWP-100. This is due to methane having a higher carbon dioxide equivalent value for a 20-year horizon compared to a 100-year horizon, as shown in Table S1 (82.5 gCO2e vs 29.8 gCO2e). Having a lifetime of around 11.8 years, methane's short-term impact is bigger than its long-term impact, which is translated in the carbon intensity results.

Using the MiQ Grade A's threshold emission percentage of 0.05% gives the lowest hydrogen carbon intensity for GWP-100, reaching 0.45 kgCO2e/kg hydrogen. When going from Grade A to Grade F, this value increases by a factor of around 2.5.

Furthermore, regarding the methane emission percentage, increasing this value from MiQ Grade A to MiQ Grade F, and from 3 to 5%, has a significant increase on the overall carbon intensity, due to two main reasons:

1. The environmental impact of methane, having 29.8 to 82.5 times the equivalent of CO2 for a 100-year horizon and a 20-year horizon respectively
2. The share of natural gas supply chain from the total process emissions, as when using a low-carbon electricity for methane pyrolysis via thermal plasma, the largest share of the emissions come from the natural gas supply chain

Additionally, a linear increase can be seen when going from the lowest methane emissions to 5% for GWP-100 and GWP-20, as the only parameter varying is the percentage of methane emitted to the atmosphere.

What is worthy to note, is that using the largest emission percentage (5%) with GWP-20 gives the highest carbon intensity for hydrogen of 4.8 kgCO2e/kgH2, that is still 51.5% less than the carbon intensity of hydrogen produced through SMR. Thus, even when taking the extreme values that overestimate the emissions, hydrogen production through methane pyrolysis via thermal plasma still performs better than hydrogen produced through SMR by a significant margin.

Moreover, using the GWP-20 is particularly interesting for the alternative scenario where renewable natural gas is used, as by removing methane from the atmosphere for every kilogram of hydrogen produced, and by attributing more weight for methane using the GWP-20, the carbon intensity of hydrogen reaches -15.70 kgCO2e/kg using the mass allocation method for a 100% renewable natural gas percentage.

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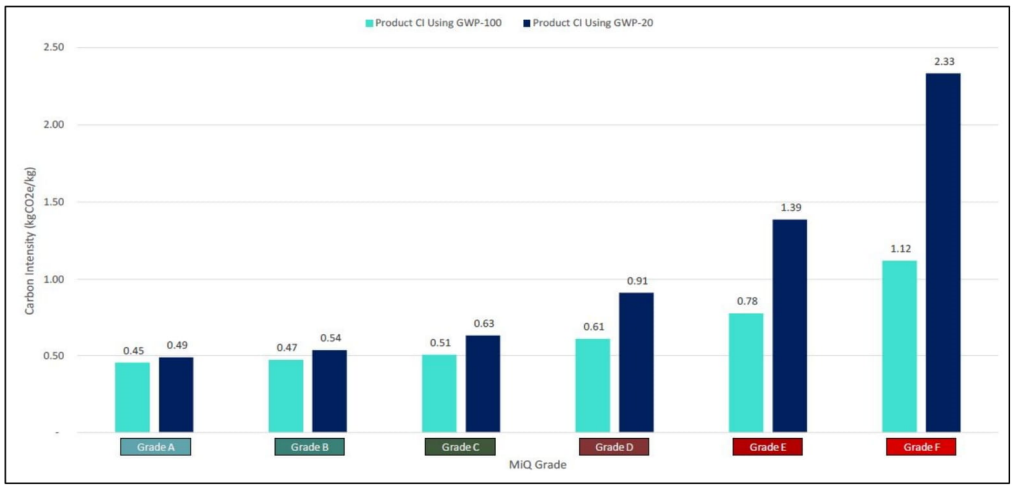

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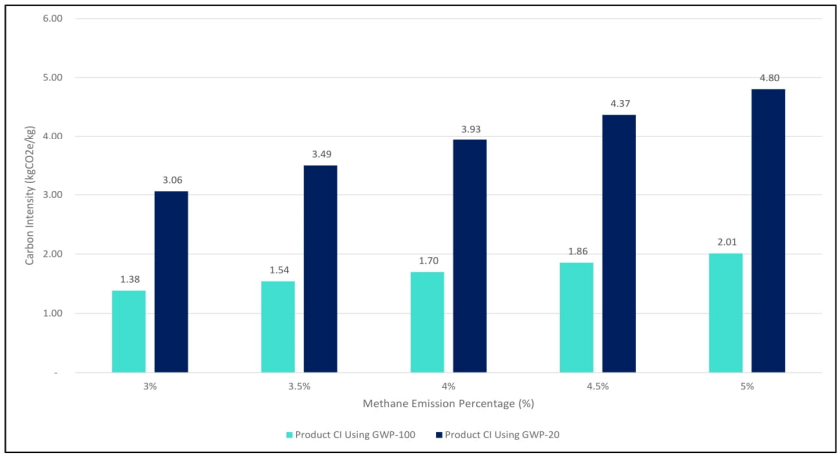

Figure 6: Variation of Products' Carbon Intensity with Methane Emission Percentage

(A) Using MiQ Grades and the Mass Allocation Method
(B) Using a 3-5% Range and the Mass Allocation Method

3.4. Blending with Different Types of RNG

As seen in the alternative scenario, blending the feedstock with renewable natural gas has a significant impact on the process emissions. Thus, it is therefore important to assess different types of RNG in order to determine at which percentage each RNG type leads to a hydrogen CI less than 0.4 kgCO2e/kg, a target that is of particular interest for policy makers.
For this, three types of RNG are assessed:

- RNG from food waste via anaerobic digestion (CI of -4.55 kgCO2e/kg RNG)
- RNG from fats, oil, and grease (FOG) via anaerobic digestion (CI of -3.70 kgCO2e/kg RNG)
- RNG from swine manure via anaerobic digestion (CI of -8.47 kgCO2e/kg RNG)

All three RNG are modelled using GREET's available models [50-51], and three different percentages are taken for each type:

1- 100% of the feedstock as the respective RNG type
2- The percentage of the RNG for which the CI of hydrogen is 0.4 kgCO2e/kg
3- The percentage of the RNG for which the CI of hydrogen is 0.0 kgCO2e/kg

Results are shown in figure 7.
RNG from swine manure is the best performing RNG type from an environmental perspective by a significant margin, with RNG from food waste in second place. In order to drop below 0.4 kgCO2e/kgH2, a percentage as low as 4.5% of the feedstock is required using RNG from swine manure, whereas 8.3% of feedstock are required for RNG from food waste, and 10.2% for RNG from FOG.

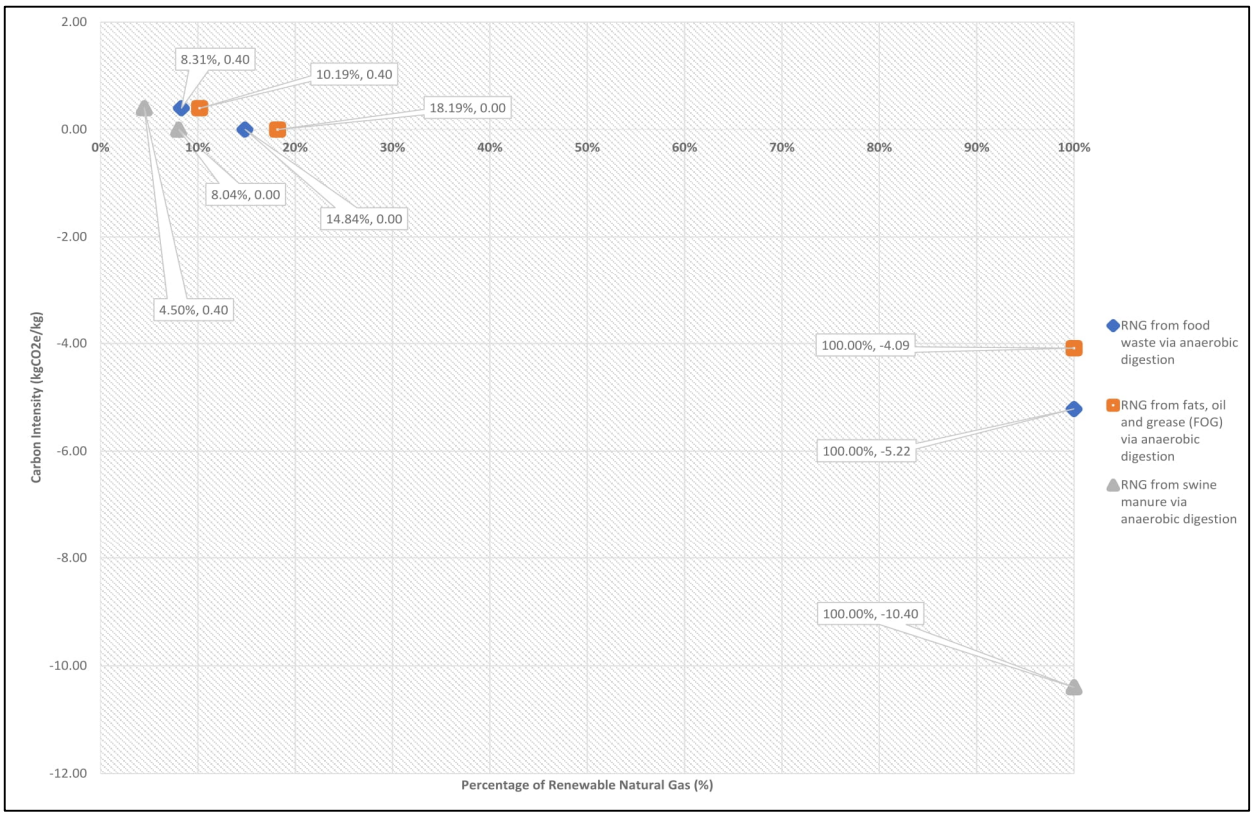

Figure 7. Variation of RNG Percentage with Product CI for Several Types of RNG

Detailed results on the emissions of the renewable natural gas supply chain are reported in Table S3 of the supplemental information document S1

3.5. Comparison of Grey, Blue, Green, and Turquoise Hydrogen

In this subsection, the carbon intensity of hydrogen produced from methane pyrolysis via thermal plasma is compared to other production methods, notably grey hydrogen (using SMR), blue hydrogen (using SMR coupled with carbon capture and sequestration (CCS)), and green hydrogen produced from electrolysis powered by renewable sources.

Regarding blue hydrogen in particular, Howarth and Jacobson's calculations [9] using SMR with carbon capture and flue gas capture are reproduced. The base-case's 3.5% emission percentage used in their study is taken on the value chain up to consumption, with 3.4% of emissions up to production. Therefore, to compare with our model that calculates the carbon intensity up to production, Howarth and Jacobson's values are recalculated up to production phase with a 3.4% emission percentage. Moreover, the latest GWP-20 values (Table S1) are used for all calculations in this case to be able to compare. On the other hand, the lowest end of Howarth and Jacobson's sensitivity analysis range is also taken (1.45% emission percentage up to production), using the latest GWP-100 values (Table S1) instead of GWP-20.

Furthermore, the gross caloric calorific heat content of hydrogen used in the calculations and conversion is taken the same as the one used by Howarth and Jacobson: is 0.286 MJ per mole [9].

Results are presented in Figure 8.

Compared to Howarth and Jacobson's base case blue hydrogen value, and using the same 3.4% methane emission percentage, hydrogen produced from methane pyrolysis via thermal plasma emits 81.4% less GHGs using the mass allocation method.

Even with lower methane emissions (1.45%), and with less weight attributed to these emissions (GWP-100), turquoise hydrogen produced using plasma and renewable energy emits significantly less GHGs than Howarth and Jacobson's blue hydrogen.

Thus, from an environmental perspective, methane pyrolysis via thermal plasma seems to present significant advantages compared to SMR coupled with CCS. However, Howarth and Jacobson's blue hydrogen CI is greater than other blue hydrogen studies due to the authors' assumptions and hypotheses that some criticized to be overestimating the impact of methane emissions.

As a result, GREET's carbon capture pathway is instead used for the same baseline assumptions to calculate blue hydrogen's carbon intensity [62]. GREET's default model for SMR with CO_2 sequestration is used as it provides a benchmark model for the US for blue hydrogen, and is based on the same model of gray hydrogen assessed in this study. Therefore, GREET's default hypotheses for blue hydrogen and CO_2 capture rates are used.

It is shown that adding carbon sequestration reduces the carbon intensity calculated for grey hydrogen from 9.89 kgCO2e/kg to 3.32 kgCO2e/kg.
Furthermore, the carbon intensity of green hydrogen, produced from either wind electrolysis or photovoltaic electrolysis, is taken from Suleman, Dincer, and Agelin-Chaab's study [63]. The authors' values are calculated up to production, thus the carbon intensity of electrolytic hydrogen, when using renewable sources, is very small.
Grey, blue and green hydrogen's carbon intensity are compared to that of hydrogen produced by methane pyrolysis via thermal plasma both using fossil natural gas and renewable natural gas presented in previous sections. Results are portrayed in Figure 9, with the mass allocation method used for turquoise hydrogen. It is worthy to note that the focus of the study is on turquoise hydrogen, and thus all sensitivity analysis is conducted on the assessed process only. Blending the feedstock with RNG is only done for turquoise hydrogen produced by methane pyrolysis via thermal plasma and not for grey and blue hydrogen.
It is shown that using fossil natural gas, turquoise hydrogen performs significantly better than both grey and blue hydrogen, but not as good as a wind electrolysis, with the latter having almost negligible production emissions (0.0325 kgCO2e/kg hydrogen).
However, when using renewable natural gas, turquoise hydrogen is the "greenest" production method, performing largely better than wind electrolysis, removing 5.22 kg of GHGs from the atmosphere for every kilogram of hydrogen produced. This leads to a negative carbon intensity for turquoise hydrogen compared to a close-to-zero carbon intensity for green hydrogen using wind electrolysis.

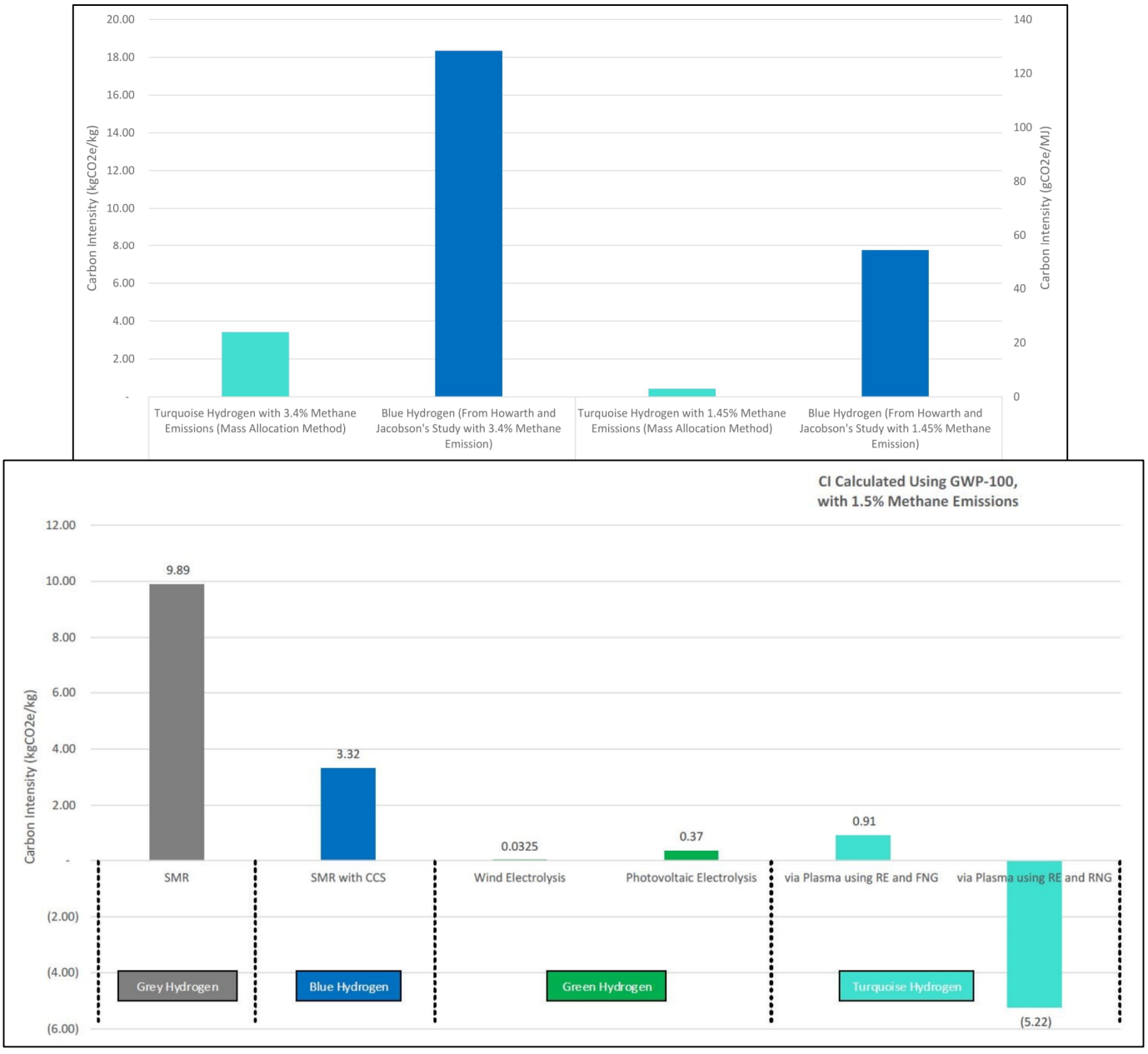

Figure 9. Comparison of Grey, Blue, Green, and Turquoise Hydrogen Carbon Intensity

4. Discussion

4.1. Environmental Performance of Turquoise Hydrogen via Thermal Plasma

The environmental performance of hydrogen produced by the pyrolysis of methane via thermal plasma is to a strong degree dependent on the emissions associated with the supply chain of the feedstock natural gas, and on the emissions associated with electricity. Using a low-carbon electricity, as is the case of this study, the emissions of the natural gas supply chain account for around 87% of the carbon intensity of hydrogen (Figure 3B). These results are for the base case using the GWP-100 and 1.5% of methane emitted from total methane produced. Using these hypotheses, the carbon intensity of hydrogen is 90.8% less than grey hydrogen using the mass allocation method (Figure 4B). This value is further reduced either by improving the sourcing of natural gas, reaching 0.45 kgCO2e/kg for MiQ Grade A natural gas (Figure 6A), or by blending the feedstock with renewable natural gas, reaching -5.22 kgCO2/kg for a 100% feedstock of RNG from food waste.
With renewable natural gas, hydrogen produced by the pyrolysis of methane via thermal plasma is by a significant margin the best environmental performant compared to grey, blue, and green hydrogen production methods.
Even when using fossil natural gas, particularly with a high MiQ Grade, hydrogen's carbon intensity using this process is only higher than that of green hydrogen produced by water electrolysis using renewable energy by a small margin.
What is worthy to note here, is that the main advantage of methane pyrolysis via thermal plasma not only lies in the reduction in carbon intensity, but also in the energy consumption compared to green hydrogen. In fact, based on thermodynamics, hydrogen produced from water electrolysis needs in theory 285 kJ/mole, whereas hydrogen produced from methane pyrolysis needs around 38 kJ/mole, i.e. 7.5 times less energy than water electrolysis, in theory[50]. In practice, based on the quality of the coproduced carbon, methane pyrolysis using plasma consumes between 10 and 30 kWh per kg H2, significantly less than water electrolysis that consumes 55 kWh per kg H2 on average [64].
Additionally, methane pyrolysis via thermal plasma has the potential to also disrupt industries that rely on hydrogen as a feedstock, such as the ammonia industry. Recent studies show interest in thermal and nonthermal plasma applications for ammonia synthesis [65-66]. For instance, in assessing the conversion of biomass to hydrogen using anaerobic digestion and thermal plasma, and then hydrogen to ammonia using nonthermal plasma, Sarafaz et al. showed that such a process can be self-sustaining, self-sufficient, environmentally friendly and resilient [66].

4.2. Feedstock Impact

With the majority of emissions associated to the natural gas supply chain when using low carbon electricity, blending the feedstock with renewable natural gas is of particular interest for turquoise hydrogen production methods, as replacing the relatively high carbon intensity of the feedstock by a negative carbon intensity for renewable natural gas is shown in Figure 7 to decrease the carbon intensity of hydrogen by a significant margin, leading to a negative value.
The study reports results for an RNG feedstock blend up to 100%.
However, it is worthy to note that even a very small percentage of renewable natural gas will have a considerable effect on the carbon intensity of the produced hydrogen. For instance, for the alternative case renewable natural gas, produced by anaerobic digestion of food waste, every 1% of renewable natural gas blended in the feedstock decreases the carbon intensity by 0.061 kgCO2e/kg on average, as 1% of fossil natural gas is replaced by natural gas with a negative carbon intensity (-4.55 kgCO2e/kg natural gas).
The negativity in emissions comes from the fact that renewable natural gas takes more CO2e from the environment than it produces as the emissions associated with the breakdown of organic sources are captured and converted to RNG instead of escaping to the environment.
The study's results show that using even a small percentage of renewable natural gas (10-20%) is interesting for methane pyrolysis via thermal pyrolysis, leading to a better environmental performance than low-carbon hydrogen production methods such as water electrolysis using renewable energy.

4.3. Carbon Credits

It is important to reiterate that all calculations conducted in this study used the economic and mass allocation method: no displacement method was used.
Therefore, if carbon credits were allocated from carbon-black to hydrogen, the carbon intensity of hydrogen will be even lower in the methane pyrolysis method via thermal plasma.
For instance, for the base case and using the economic allocation method, carbon black is produced with less carbon intensity than using conventional processes, as was presented in Figure 4B. The reduction in GHGs can be allocated as carbon credits from carbon-black to hydrogen, increasing the overall environmental performance of hydrogen production using methane pyrolysis.

5. Conclusions

Overall, this study aims to assess environmentally the life-cycle of methane pyrolysis via thermal plasma.
Its novelty lies in being the first LCA study conducted on methane pyrolysis via thermal plasma that uses real-time plant data, and allocates the emissions among the co-products that previous studies assumed to be non-usable.

GREET2020 is used to simulate the process, comparing the carbon intensity of hydrogen and carbon-black to conventional processes. Furthermore, renewable natural gas is assessed in an alternative scenario, and varied from 0 to 100%, along with a sensitivity analysis conducted on the environmental metric and the methane emissions percentage, and on the type of renewable natural gas.
The main conclusions can be summed up as follows:

1- Hydrogen produced by methane pyrolysis via thermal plasma has a low carbon intensity, reaching 0.06 kgCO2e/kg using the mass allocation method with the GWP-100 for scope 1 where only the process' direct emissions are assessed
2- The two main factors contributing to total emissions are electricity and sourcing of the natural gas. With this, scope 2 includes purchased electricity emissions, and scope 3 includes indirect emissions associated with natural gas sourcing.
3- Using renewable electricity, notably wind power, the cumulative carbon intensity of hydrogen using this novel process for scope 1 and scope 2 together is 0.12 kgCO2e/kg for the mass allocation method
4- When using low carbon electricity, the main contribution of total emissions comes from the natural gas sourcing, with the aggregated carbon intensity of hydrogen (scope 1 + scope 2 + scope 3) reaching 0.91 kgCO2e/kg for the mass allocation method
5- Methane pyrolysis via thermal plasma not only performs better environmentally than water electrolysis, but also consumes 3-5 times less energy
6- Using renewable natural gas produced from anaerobic digestion of food waste gives a negative carbon intensity for hydrogen at a renewable gas percentage as low as 14-15%
7- Improving natural gas sourcing up to grade A (MiQ) reduces the carbon intensity of hydrogen to 0.45 kgCO2e/kg
8- Blending the natural gas feedstock with different types of renewable natural gas gives a carbon intensity of hydrogen less than 0.4 kgCO2e/kg for RNG feedstock percentages of 8-18% based on which RNG type is used
9- Even when overestimating the emissions and impact of methane by using an emission percentage of 5% and a GWP-20, hydrogen produced from methane pyrolysis via thermal plasma still has a better environmental performance than that produced by conventional processes, having a 51.5% less carbon intensity
10- Turquoise hydrogen performs better environmentally than grey and blue hydrogen, and using renewable natural gas, performs even better than green hydrogen by a significant margin

Future work needs to include economic parameters to highlight the economic benefits of this method compared to other low-carbon methods such as water electrolysis, as well as to assess the benefits of using this process with additional units to produce ammonia and compare it to conventional ammonia processes.
figur
Compared to grey, blue, and green hydrogen, turquoise hydrogen using thermal plasma and renewable natural gas is by a significant margin the best environmental performer, making it the game changer for the energy transition.

Supplemental Information

Document S1 is the main supplemental PDF, containing a section detailing the canonical methodology used in GREET
Table S1 of the supplemental information document S1 reports the adopted environmental metric values
Table S2 of the supplemental information document S1 reports the greenhouse gas emissions per process phase of the base case
Table S3 of the supplemental information document S1 reports the greenhouse gas emissions of the supply chain of the different types of renewable natural gas used in this study supply chain

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Author Contributions

Jad Diab: methodology, investigation, software, formal analysis, visualization, writing - original draft. Laurent Fulcheri: conceptualization, project administration, supervision, data curation, formal analysis, investigation. Vandad Rohani: investigation, validation, writing – review & editing, Volker Hessel: formal analysis, validation, writing – review & editing, Michael Frenklach: formal analysis, validation, writing – review & editing.

Declaration of Interests

The authors declare no competing interests.

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