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DOI

[10.1016/j.ecmx.2024.100695](https://doi.org/10.1016/j.ecmx.2024.100695)

Publication date

2024

Document Version

Final published version

Published in

Energy Conversion and Management: X

Citation (APA)

Baghirov, B., Voskov, D., & Farajzadeh, R. (2024). Exergetic efficiency and CO₂ intensity of hydrogen supply chain including underground storage. *Energy Conversion and Management: X*, 24, Article 100695. <https://doi.org/10.1016/j.ecmx.2024.100695>

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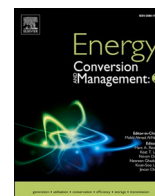
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Exergetic efficiency and CO₂ intensity of hydrogen supply chain including underground storage

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ARTICLE INFO

Keywords:

Hydrogen
CO₂ intensity
Exergy Analysis
Energy Storage
CCS

ABSTRACT

Hydrogen plays a crucial role in the transition to low-carbon energy systems, especially when integrated into energy storage applications. In this study, the concept of exergy-return on exergy-investment (ERoEI) is applied to investigate the exergetic efficiency (defined as the ratio of useful exergy output to invested exergy input) and CO₂ equivalent intensity of the hydrogen supply chain, with a specific focus on the underground hydrogen storage process. Our findings reveal that the overall exergetic efficiency of the electricity-to-hydrogen-to-electricity conversion process can reach up to 25 %. Among the hydrogen production methods, green hydrogen, produced via electrolysis powered by renewable energy, exhibits the lowest CO₂ equivalent intensity. Blue hydrogen, produced from natural gas with carbon capture, can significantly reduce the carbon footprint of electricity generation, though this advantage comes at the expense of decreased exergetic efficiency. Analysis further indicates that the exergetic efficiency of underground storage components ranges from 72 % to 92 %. A substantial fraction of the exergy is lost during compression and injection of the stored hydrogen. Nevertheless, the subsurface operations contribute a minimal CO₂ emission, between 1.46–4.56 grams of equivalent CO₂ per megajoule (gr-CO_{2eq}/MJ) when powered by low-carbon energy sources. Furthermore, it is found that hydrogen loss in the reservoir, along with methane and hydrogen leak during surface operations, notably affects the overall efficiency of the storage process.

Introduction

To effectively address climate change and secure a sustainable energy future, transitioning to low carbon, particularly, renewable energy sources is essential. In 2022, fossil fuels maintained a dominant position, constituting nearly 80 % of the global energy mix, while renewables only made up about 20 % of the total energy mix [1]. However, projections indicate a significant transformation, with fossil fuel's share anticipated to decline steadily, while renewable energy sources are forecasted to surpass 50 % of total energy consumption by 2050 [1]. Despite this growth, electricity generation from renewable sources, such as wind and solar, remains inherently dependent on weather conditions and time of day, posing challenges for establishing a reliable and scalable power network. A potential solution lies in the storage of renewable electricity during peak production periods for later use [2].

Hydrogen (H₂) has emerged as a promising storage medium and energy carrier, offering the potential to decarbonize the energy sector

effectively. Its capacity for long-term storage enables the utilization of excess renewable energy during periods of high demand. The high energy density of H₂ (118 MJ/kg) and clean combustion, producing only water vapor make it an ideal fuel for electricity and heat generation [3]. With the ability to be produced from renewable sources such as wind and solar power through processes like electrolysis (green H₂), hydrogen holds immense potential to facilitate the transition to a low-carbon economy. Furthermore, hydrogen produced from fossil fuels using carbon capture and storage (CCS) technology, commonly referred to as blue H₂, also contributes to the decarbonization of the energy portfolio, despite not being directly sourced from renewables [4]. However, realizing the full potential of H₂ as an energy carrier entails overcoming several challenges. Firstly, the production of hydrogen is currently energy-intensive and costly, with the majority produced from methane (CH₄) and other hydrocarbons (96 % of total produced H₂), leading to substantial greenhouse gas emissions. Only a small fraction (4 %) of H₂ is produced via low-carbon water electrolysis [5]. Moreover, the low

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<https://doi.org/10.1016/j.ecmx.2024.100695>

Received 4 June 2024; Received in revised form 28 July 2024; Accepted 21 August 2024

Available online 23 August 2024

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volumetric density of hydrogen necessitates expensive pressurization or liquefaction for storage, posing economic and safety challenges. Current storage methods, primarily, in pressurized or cryogenic containers as either gas or liquid, are insufficient to meet large-scale (TWh) storage demand. Therefore, establishing cost-effective, secure, and reliable H₂ storage systems is imperative to foster the growth of a hydrogen economy [6].

Underground storage offers a viable solution for long-term and large scale H₂ storage, leveraging various global geological formations. Different types of underground reservoirs, such as solution-mined salt caverns [7–9], saline aquifers [10–12], and depleted hydrocarbon fields [13,14] have been explored as potential storage sites for hydrogen. Integrating underground storage with green and blue H₂ production contributes to achieving decarbonization targets and reducing greenhouse gas emissions in the energy industry. While green H₂ aligns directly with renewable energy sources, blue H₂ offers a dependable energy resource to complement intermittent renewables. Furthermore, existing studies and industrial practices underscore the viability and significance of integrating blue H₂ production with geological storage options to facilitate the transition toward sustainable energy and reduce carbon emissions in the energy industry [4,15–17]. However, despite its potential, underground H₂ storage systems have not undergone a comprehensive evaluation concerning their thermodynamic efficiency and environmental impact within the entire H₂ supply chain, including underground storage options.

Several studies in the literature have conducted thermodynamic and environmental assessments of H₂ production and storage systems. Ozcan et al. [18] conducted an exergy analysis of a solar-based H₂ production and storage system, yielding overall energy and exergy efficiencies of 16.31 % and 17.6 %, respectively. The solar field accounted for the largest portion of the total exergy investment, approximately 64 %. Calderon et al. [19] proposed a system integrating photovoltaic (PV) and wind technologies with H₂ storage to meet the electrical energy demands of Badajoz, Spain. They found that PV modules in Badajoz exhibited a low exergy efficiency of 8.39 %. Khosravi et al. [20] investigated energy, exergy, and economic analyses for renewable hybrid energy systems utilizing H₂ storage, suitable for remote areas, achieving average energy and exergy efficiencies of 12 % and 16 %, respectively. The PV system exhibited the highest exergy destruction at approximately 65 %. Al-Zareer et al. [21] assessed the efficiency of two compressed H₂ storage systems, estimating overall exergy efficiencies of 92.9 % and 96.1 % for the first and second models, respectively. Neelis et al. [22] conducted a life-cycle analysis of H₂ production and storage systems for automotive applications using eight fuel chains. Compressed H₂ storage systems demonstrated the highest exergetic efficiency, while liquid H₂ systems exhibited lower exergetic efficiency due to the significant exergy requirements for liquefaction. Farajzadeh et al. [23] calculated the exergetic efficiency and CO₂ intensity of underground bio-methanation processes using renewable H₂ for electricity and heat generation. The biomethanation process achieved maximum exergetic efficiencies of 15–33 % for electricity and 36–47 % for heat generation, with H₂ production being the primary exergy consumer in both applications. Pérez et al. [24] compared the conversion efficiency of H₂ to renewable electricity within underground energy storage systems (adiabatic compressed air storage, hydrogen storage, and methanation). The conversion efficiency of H₂ storage in salt caverns was calculated to be 43–48 %.

Furthermore, from an environmental perspective, our analysis primarily focuses on the CO₂ footprint of hydrogen production and storage systems. Bhandari et al. [25] conducted a comprehensive analysis of 21 studies examining the life cycle assessment (LCA) of various H₂ production methods, focusing on ecological considerations. They found a consistent global warming potential (GWP) of 0.97 kg CO_{2eq}/kg H₂ for wind-based electrolysis, while steam methane reforming of natural gas showed GWP values ranging from 8.9 to 12.9 kg CO_{2eq}/kg H₂. Sebastian et al. [26] examined the life-cycle greenhouse gas emissions of H₂

production via different CH₄ decomposition methods (plasma, molten metal, and thermal gas), and found that the plasma system using renewable electricity generated the lowest emissions, with 43 g CO_{2eq}/MJ. G. Kubilay et al [27] focused on the environmental implications of different hydrogen storage technologies. Their findings indicated that liquid hydrogen storage resulted in the lowest emissions, averaging 3.5 kg CO_{2eq} per kg of stored H₂, whereas metal hydride storage tanks exhibited the highest emissions, reaching 113.6 kg CO_{2eq}. Bicer et al. [28] evaluated four green ammonia production methods and estimated greenhouse gas emissions for each option. Municipal waste sources had the lowest CO₂ equivalent emissions (0.34 kg CO_{2eq}). Other studies explored different H₂ production methods in terms of production cost and CO₂ emissions [29–32].

In summary, various studies have focused on the thermodynamic or environmental analysis of H₂ production and storage systems, separately. However, while a thermodynamically optimized system might excel in one aspect, it might not be optimal in terms of environmental considerations, or vice versa [23,33,34]. Despite the potential for optimizing systems based on thermodynamic or environmental criteria alone, a holistic approach is necessary for viable engineering solutions. Thus, it is imperative to simultaneously consider all or a subset of these criteria to identify a more optimal solution [23,33,34]. A key feature of our study is the integration of thermodynamic (exergy) and environmental (CO₂ intensity) analyses into a unified framework. Furthermore, there exists a substantial gap in the literature concerning the evaluation of thermodynamic efficiency or exergetic efficiency, and particularly CO₂ intensity throughout the full cycle of the H₂ supply chain, including the option of underground hydrogen storage (UHS). Therefore, our study aims to bridge this gap by developing exergy-based method to define the exergetic efficiency and CO₂ intensity (grams of CO₂ per MJ of electricity) of the full life-cycle H₂ supply chain with encompassing underground storage.

Exergy serves as a helpful metric for evaluating and comparing the sustainability of energy storage systems [35–37]. It is the maximum portion of the energy that can be converted to useful work [23,38]. Exergetic efficiency, also known as Exergy Return on Exergy Invested (ERoEI), denotes the percentage of exergy input (exergy investment) transformed into productive work (exergy return), with the remaining “lost” or “wasted” exergy attributable to irreversibilities described by the second law of thermodynamics, practically contributing to CO₂ emissions [23,39,40]. In the study, exergy analysis is done with several common H₂ production methods with high technical readiness level (TRL). The results are then compared to the case of producing electricity from CH₄, whose CO₂ has been captured. By comparing various hydrogen production (green and blue H₂) methods and calculating ERoEI and CO₂ intensity, our research offers novel insights into optimizing the H₂ supply chain and its environmental footprint. Importantly, the analysis considers CH₄ and H₂ leakage and their global warming potential in the study as well to reflect their environmental impact accurately. The results of this paper provide insights into contribution of each stage of the process on the exergetic efficiency and leveled CO₂ footprint of the H₂ supply chain, including UHS system, which can then be used for optimization purposes.

The paper is structured as follows: Initially, the H₂ supply chain and its boundaries are precisely defined for assessment. Next, the work and material streams of the system are calculated based on underlying assumptions. These data are then used to determine the ERoEI and CO₂ intensity of the UHS process with different H₂ production methods. A sensitivity analysis is performed to quantify the impact of different parameters on the results. The results are presented in two distinct sections: the first section discusses the findings of overall H₂ supply chain process, and the second part focuses exclusively on the UHS process. The paper concludes with insightful remarks summarizing the findings.

System and Methodology

Method of analysis

In this study, the method based on the concept of Exergy Return on Exergy-Investment (ERoEI) is applied to investigate the exergetic efficiency and CO₂ intensity of the H₂ supply chain for electricity application including underground storage process. The intermediate and final results of the exergy analysis serve as valuable tools for decision-makers to identify the most optimal scenario for the studied process while minimizing environmental impact [23,35,37,38]. The ERoEI is defined as

$$ERoEI = \frac{Ex_{returned}}{Ex_{invested}} \quad (1)$$

where, $Ex_{returned}$ represents the gained exergy (in MJ) from the produced fuel (H₂ and CH₄) and $Ex_{invested}$ signifies the total exergy investment in different stages of the process. Theoretically, ERoEI can range from 0 to $+\infty$. For the system considered in this study, eq. (1) is expanded to:

$$ERoEI = \frac{Ex_{H_2}^{ch} \times \eta}{Ex_{H_2prod.} + Ex_{tran.} + Ex_{UHS} + Ex_{end_use}} \quad (2)$$

Here, the returned exergy is the chemical exergy of H₂ ($Ex_{H_2}^{ch}$) discounted by the efficiency (η) of the fuel cell. The invested exergy includes exergy requirement for H₂ production ($Ex_{H_2prod.}$), transportation ($Ex_{tran.}$), underground storage (Ex_{UHS}), and the end-use application streams (Ex_{end_use}). To perform the exergy analysis, the material ($Ex_{returned}$) and work ($Ex_{invested}$) streams of the system depicted in Fig. 1 are elaborated in more detail [23,38,40].

After estimating the consumed exergy, the CO₂ intensity of the process can be calculated. The carbon intensity is defined as the mass of CO₂ released per unit of energy (gr-CO₂/MJ). To estimate the carbon intensity, the invested exergy of the system is multiplied by the specific carbon emission of the energy resources (Table 1) [23]. The specific CO₂ emission of CH₄ and wind power are considered 55 and 7 gr-CO₂/MJe, respectively [41,42]. To provide a reliable quantitative analysis, the leveled CO₂ equivalent emission is calculated by considering CH₄ (extraction and transportation to H₂ production side) and H₂ leakages (throughout the entire supply chain, including H₂ production, transportation, underground storage, and end-use application) and their global warming potential (GWP) for a selected period of 100 years, i.e.,

$$CO_{2eq,int} = \frac{Ex_{invested} \times W + M \times L \times GWP}{Ex_{returned}} \quad (3)$$

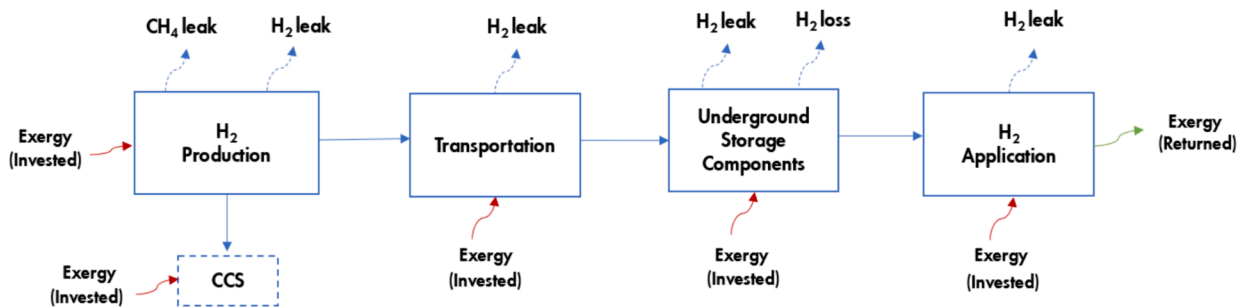


Fig. 1. Schematic view of the system boundary and main components for the full H₂ supply chain, including H₂ production, transportation, underground storage, and end-use application.

Table 1

Main input values used for calculating CO_{2eq} intensity. The “blue” refers to H₂ production from methane, whose associated CO₂ has been captured.

Input	Value	References
CH ₄ consumption (blue H ₂ production option)	3–3.8 kg/kg H ₂	[30,43]
H ₂ consumption	1 kg	–
Leak rate during CH ₄ consumption (mass fraction)	0–1.5 %	[44–48]
Leak rate during H ₂ consumption (mass fraction)	3.5–9 %	[49]
GWP-100 of CH ₄	28	[50]
GWP-100 value of H ₂	12	[51,52]
Specific CO ₂ emission of CH ₄	55 gr-CO ₂ /MJe	[41]
Specific CO ₂ emission of wind power	7 gr-CO ₂ /MJe	[42]

where, W represents the specific carbon intensity of energy source (gr-CO₂/MJ), M denotes the mass of the utilized fuel (including CH₄ consumption in the hydrogen production process and H₂ consumption across the full H₂ chain), and L shows the leaked mass fraction of gases (H₂ or CH₄) occurring at various stages of the process.

System definition

Fig. 1 depicts the main components of a H₂ supply chain including hydrogen storage process. The overall system consists of the H₂ production, transportation, underground storage, and end-use application subsystems. For this study, the system conditions are modelled based on a candidate depleted gas reservoir. The initial stage involves H₂ production through various methods outlined in Table 2. For blue options, steam methane reforming (SMR), autothermal reforming (ATR), and partial oxidation (POX) methods are explored due to their widespread usage and high technical readiness levels (TRL) [30,53]. Water electrolysis, utilizing electricity sourced from low-carbon sources (in this case, windmills), is chosen for green H₂ production. The required water undergoes desalination and treatment before entering the electrolysis unit. It is assumed that the water source is near the plant, with negligible production and transportation exergy requirements.

Afterwards, the produced H₂ is compressed to the necessary pipeline pressure, transported, and stored in a gas reservoir. Since the produced gas stream contains native reservoir gas (assumed to be pure CH₄) along with H₂, separation from other gases is necessary before final use. Regarding the produced CH₄ stream, two scenarios are considered: In Scenario 1, the produced CH₄ is reinjected back into the reservoir, whereas in Scenario 2, CH₄ is exported to a near-by power plant to generate electricity. The produced CO₂ from the power plant is captured and stored in a near-by storage site. The exported H₂ is further purified

Table 2

Exergy investment in the form of heat and electricity for different H₂ production methods. CCS stands for Carbon Capture and Storage. The final exergy consumption is equal to sum of electricity and heat demand of each production method.

H ₂ production methods	Abbreviation	Invested exergy (MJh/kg-H ₂)	Invested exergy (MJe/kg-H ₂)	Total invested exergy (MJ/kg-H ₂)	References
Steam methane reforming with CCS	SMR+CCS	151–215	20–81	171–296	[30,54,55]
Auto-thermal reforming with CCS	ATR+CCS	150–185	34–110	184–295	[54,55]
Partial Oxidation with CCS	POX+CCS	139–209	26–119	165–328	[53]
Electrolysis	–	–	169–252	169–252	[64–67].

for generating electricity in fuel cells. Fig. 1: Schematic view of the system boundary and main components for the full H₂ supply chain, including H₂ production, transportation, underground storage, and end-use application.

Additionally, the following assumptions are applied in the calculations:

- The outlet pressure of the produced H₂ is 15 bar after the electrolysis process, 30 bar after the SMR, ATR, and 50 bar after the POX processes [30,53–55].
- Transportation pressure of H₂ is assumed to be 50 bar.
- The pressure of the gas reservoir is 350 bar.
- The distance from H₂ production site to the reservoir is 300 km.
- The transportation distance from the reservoir to the H₂ end-use application is 30 km.
- The gas power plant is positioned close to the reservoir (~20 km).
- The distance between CO₂ capture and storage facilities is 50 km.
- The export pressure of CH₄ is regulated at 50 bar.
- CH₄ is recompressed after separation from 50 bar to the reservoir pressure.
- 3–18 % H₂ loss [56] is considered in the reservoir due to mixing, structural trapping, solubility, biochemical and geochemical conversion processes.
- 3.5–9 % H₂ [49] and 0–1.5 % CH₄ [44–48] leakage rates are assumed in overall surface operations.
- For the system considered in this study, the main produced fuels are H₂ and CH₄, with the chemical exergy values of 118 MJ/kg and 51.8 MJ/kg, respectively [57].

In the following, different stages of the system are explained in more details.

Hydrogen Production. In this work, blue (SMR+CCS, ATR+CCS, POX+CCS) and green (water electrolysis using a low carbon power source) methods are considered as H₂ production technologies. The so-called grey option is currently the most common method for producing H₂, usually produced from the natural gas by reforming or oxidation process but without collecting the generated greenhouse gases [54,55]. In the blue H₂ option, the produced CO₂ is captured and stored in geological formations or utilized as feedstock in other processes. However, the addition of CCS stage to the process increases its energy requirements [54,55]. The magnitude of the exergy required to separate CO₂ from a gas mixture depends on its concentration or partial molar volume. Typically, monoethanolamine (MEA) solvent is used in fossil-fueled power plants to absorb CO₂ from flue gas streams with high capture efficiency (80–95 %) [40,53–55]. The required exergy with MEA-based method for capturing CO₂ can vary between 2.5–6 MJe/kg CO₂ [37,58,59]. It is important to realize that when the energy source of the CCS stage is not zero carbon, additional CO₂ is produced during the process [54,55].

In contrast to the SMR method, production of H₂ from the ATR and POX processes requires 3.2–6.6 kg of pure oxygen (O₂) per kg of H₂, thereby increasing the invested exergy for H₂ production from these methods [54,55]. The air separation unit (ASU) produces O₂ using the cryogenic distillation method and uses electricity as the primary energy input, necessitating 0.9–5.1 MJe/kg-O₂ [53]. The O₂ demand for the

ATR and the POX options rises significantly when a CO₂ capturing unit is added to the system, since more natural gas and, by extension, O₂ are required by the power generating system to meet the exergy demand of CO₂ separation [54,55]. Furthermore, it is worth mentioning that both oxidation and reforming processes require 3–3.8 kg of CH₄ as feedstock per kg H₂ production, and that the CH₄ extraction requires 3–8 MJ of exergy per kg [23,30,43].

The exergy investment for production of H₂ from water electrolysis is 169–252 MJe/kg-H₂. The carbon footprint of this method largely hinges on the electricity source powering the electrolysis. These values also encompass water desalination and treatment, which involve various operations such as fine screening, coagulation-flocculation, filtration, and desalination, depending on the selected water source for the electrolysis process [60,61]. H₂ production from water electrolysis consumes from 10.0 to 22 kg water per kg of H₂. This study focuses on using seawater for electrolysis, which demands 1–6 MJe/kg-H₂, factoring in the energy for desalination and purification processes [60–63]. For the purpose of our analysis, the electricity for green H₂ production is sourced from wind turbines, associated with a CO₂ intensity of 1–3 kg-CO₂/kg [64–67].

Table 2 provides the magnitude of required exergy in the form of heat (MJh) and electricity (MJe) for different H₂ production methods.

Transportation. The transportation phase involves initially compressing H₂ and transporting it to the field site via a pipeline. For the case considered here, H₂ is initially compressed to 50 bar and transported by pipeline to the storage site located 300 km away from the production site. The exergy requirement for the initial compression varies depending on the outlet pressure of the H₂ production method. For example, the outlet pressure of the POX method is already at 50 bar and therefore it does not require further compression at this stage [53]. Compression is assumed to be an isentropic (adiabatic) process, which means that the entropy of the streams remains constant. CoolProp is used to perform the thermodynamic calculations [68].

To compute the practical exergy, expressed as:

$$Ex_{comp}^{prac} = \frac{Ex_{comp}^{th}}{\eta_{comp}\eta_d} = \frac{H_2(S_1(T_1, P_1), P_2) - H_1(S_1(T_1, P_1), P_1)}{\eta_{comp}\eta_d} \quad (4)$$

the efficiencies of the compressor (0.50–0.90) [69,70] and the electrical drive (0.8–0.9) [23,71] are considered in Eq. (4) [40]. It is assumed that all required electricity is supplied from a low-carbon renewable (wind) source. The efficiency of the wind power plants is in the range of 0.30–0.55 [72]; however, this is not considered in the calculations. The practical exergy investment for initial compression is provided in Table 3.

Table 3
Calculated compression exergy for transportation and injection of H₂.

Work Stream	Theoretical Exergy (MJe/kg H ₂)	Practical Exergy (MJe/kg H ₂)
Transportation Compression (from 15 to 50 bar) Electrolysis output	1.8	2.2–4.5
Transportation Compression (from 30 to 50 bar) SMR, ATR output	0.7	0.86–1.75
Injection Compression (from 50 to 350 bar)	3.0	3.7–7.5

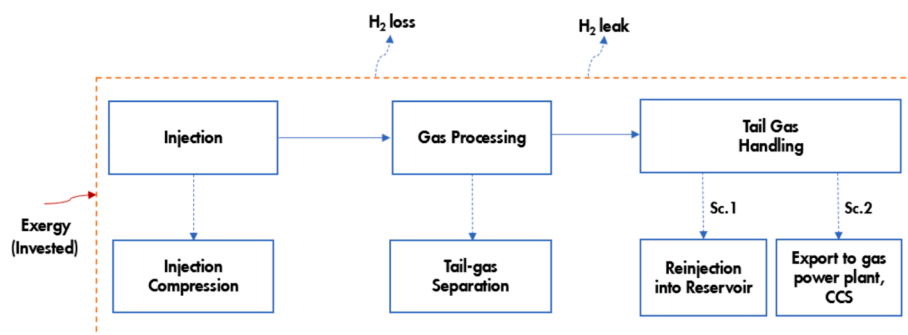


Fig. 2. Schematic overview of the underground hydrogen storage components. The blue dashed lines highlight subcomponents of the underground storage components. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Additionally, during transportation, a pressure drop of 0.15–0.30 bar/km and an exergy consumption of 1.44–3.96 MJe/kg H₂ for pipeline transport are considered [73,74].

Underground Hydrogen Storage Components (UHS). A UHS system typically consists of three main parts: injection, gas processing, and tail gas handling parts, as shown in Fig. 2.

Injection Component. In this stage, H₂ is recompressed to a pressure of 350 bar, which corresponds to the reservoir pressure, and subsequently injected into the reservoir. The compression exergy values are provided in Table 3.

Gas processing. During the storage period, H₂ mixes with in-situ gases in the gas reservoir. Therefore, it is crucial to separate H₂ from the produced tail gas before transferring it to next stage. The gas processing segment of the UHS process involves separating H₂ from the produced fluids (mixture of gases and possibly water), and transporting it to the application side. The chosen technology for H₂ separation from the H₂/CH₄ mixture is the pressure swing adsorption (PSA) method. The exergy requirement for gas separation is indicated in Table 4. It is assumed that the produced gas mixture is mainly composed of H₂ and CH₄ with proportion of 85–100 % H₂ and 0–15 % CH₄ in-line with some simulation and field results [56]. In addition, it is assumed that H₂ end-use application is located close to the gas field (~30 km). Therefore, the exergy investment in this transportation stage can be neglected.

Tail gas handling. The separated tail gas, primarily consisting of methane (CH₄), is managed through either reinjection into the reservoir or exportation to a power plant for electricity production. For reinjection (Scenario 1), the exergy cost of compressing and injecting CH₄ is calculated to range 0.51–1.0 MJe/kg CH₄, as detailed in Table 4. Alternatively, in the Scenario 2, where CH₄ (which may be mixed with H₂) is exported to a power plant, it is essential to also capture and transport CO₂ emissions generated during electricity production (refer to Table 4 for specifics).

H₂ loss in the reservoir. During the storage period of H₂ in reservoirs, a fraction of the injected H₂ may be lost due to various factors such as the complex geology of the reservoir, lateral movements, dispersion and mixing with in-situ gas, dissolution in water, and biochemical and geochemical conversion processes. Studies indicate that the lost fraction of H₂ could range between 3–18 % [56].

Table 4 provides the main exergy investments of the subsurface components of the UHS system depicted in Fig. 2.

Table 4

Summary of the exergy requirements for the work streams of the UHS system depicted in Fig. 2.

Underground Storage Components	Practical exergy Investments [MJe/kg gas]	References
Injection	3.7–7.5	[68]
Gas Processing	3–6.9	[75,76]
Tail gas handling (Sc.1)	0.51–1	[68]
Tail gas handling (Sc.2)	2.5–6	[37,58,59]

H₂ to Electricity. Fuel cells are devices that directly convert the chemical exergy of H₂ into electricity. The only byproduct of a perfect hydrogen–oxygen fuel cell is water, although a portion of the input exergy is lost as heat during the process. The efficiency of fuel cells depends on various factors including the type of reactants, the electrolyte used, and the temperature of the reactants [77,78]. Currently, H₂ fuel cells have a conversion efficiency ranging from 40–60 % [79]. Following the gas-processing stage, there may still be some contaminants present in the transported gas, which could be acceptable within the pipeline. However, for electricity generation in H₂ fuel cells, H₂ needs to undergo further treatment to ensure its purity exceeds 99 % [78]. Various methods, including chemical and physical absorption, adsorption, pressure swing adsorption (PSA), and polymer membrane technology, can be employed for the purification process [77]. In the analysis, membrane-based purification technology is considered due to its superior recovery capability, with a required exergy of 10.8–22.1 MJe/kg-H₂ [80].

Fugitive CH₄ and H₂ emissions

CH₄ leakage. Methane is a highly potent greenhouse gas, emitting approximately 28 times as much warming as CO₂ over a 100-year time frame following a pulsed emission [50]. Achieving leak-free production and utilization of natural gas in industry poses significant challenges. Especially, both oxidation and reforming processes require 3–3.8 kg CH₄ per kg H₂ production [30,43]. OGI (Oil and Gas Climate Initiative) member companies in the energy sector have committed to a 2025 target intensity of well below 0.20 %, with an aim to achieve near zero methane emissions by 2030 [48]. However, the measurement of methane emissions is characterized by significant uncertainty and shows large variations depending on the specific kind of fossil fuel production, such as shale oil and gas, conventional oil and gas, oil sands. The observed disparity exists in several worldwide locations, where the complex characteristics of each production process contribute to the difficulty of measuring emissions. In this study, we assume 0–1.5 % methane leakage rate for the processes consuming CH₄. Nevertheless, certain regions exhibit emissions that may extend up to 3 % [44–48].

H₂ leakage. H₂ is an indirect greenhouse gas that interacts with tropospheric hydroxyl radicals, influencing the distribution of CH₄ and ozone in the atmosphere. Moreover, it enhances the stratospheric

Table 5

H₂ leak rate (%) in various stages of the system shown in Fig. 1.

Process	Leak Rate (%)	Impacted stage in Fig. 1	References
Blue H ₂ Production	1–1.5	H ₂ Production	[49]
Green H ₂ Production	2–4	H ₂ Production	[49]
Compression, Transportation and Injection	1–2	UHS and Transportation	[49]
H ₂ End Use	1.5–3	H ₂ Application	[49]

concentration of water vapor, contributing to warming by hindering outgoing infrared radiation [46,81]. According to research data from Columbia University's Energy Center, the full implementation of the H₂ supply chain is associated with H₂ leakage rates ranging between 3.5–9 % throughout various stages, including production, transportation, subsurface storage (compression, injection, and separation), and end-user applications [49]. H₂ leakage has a crucial impact on the exergy and carbon efficiency of the overall H₂ chain process. The leakage rates for each stage of the process are detailed in Table 5.

Results and Discussions

In this section, we will conduct a thorough analysis of the full-cycle exergetic efficiency and CO_{2eq} intensity of the underground hydrogen (H₂) storage process, taking into account different H₂ production methods. Our primary objective is to assess these metrics across two key areas: the complete H₂ supply chain, which encompasses H₂ production, transportation, underground storage, and end-use applications, as well as the specific process of underground storage.

1. Full H₂ supply chain

A significant portion of the total exergy is consumed during the H₂ production phase. Depending on the chosen tail-gas handling approach (Scenario 1 or 2), subsurface storage and the application of H₂ at the end-use stage account for approximately 6 % and 25 % of the total exergy investment, respectively. With fuel cells' conversion efficiencies, 48–87 MJ (MJ) of electricity can be generated from 1 kg of H₂. Utilizing Eq. (1), we calculate that the maximum exergetic efficiency of the entire system, ranges from 0.07 to 0.25. This reveals that a substantial part of the invested exergy (75–93 %) is lost throughout the cycle of converting electricity to H₂ and back to electricity.

Fig. 3 illustrates the exergetic efficiency (ERoEI) in the full H₂ supply chain for different H₂ production techniques. Among these, the Partial Oxidation (POX) method emerges with the highest overall exergetic efficiency, marked at ERoEI=0.25. Since the output pressure of H₂ obtained by the POX method is higher (50 bar) compared to the other methods, less exergy is spent on the compression stage. Furthermore, integrating Carbon Capture and Storage (CCS) with any H₂ production method – resulting in so-called blue H₂ – decreases the overall exergetic efficiency. This decline is primarily due to the additional exergy required for CO₂ capture.

As previously discussed, once the exergy invested in the process is determined, its corresponding CO₂ intensity can also be calculated by considering the specific CO₂ emissions of the energy source. Fig. 4 presents the CO₂ equivalent intensity for the entire supply chain through various H₂ production methods.

The production of grey H₂ results in a significantly high level of CO₂ emissions. To mitigate this, the blue H₂ option incorporates Carbon Capture and Storage (CCS) technologies to capture the CO₂ produced during operations. However, it is important to acknowledge that not all emitted CO₂ can be captured, and part of the generated CO₂ is released. The implementation of CCS in the blue H₂ production significantly reduces the CO₂ intensity of the generated electricity by 34–76 %, as illustrated in Fig. 4. The CO₂ capture efficiency is estimated to be between 80–90 % for Steam Methane Reforming (SMR) and 90–100 % for Autothermal Reforming (ATR) and Partial Oxidation (POX) methods [53–55]. The higher capture efficiency in ATR and POX is attributed to the injection of pure oxygen (O₂) into the system, which prevents nitrogen dilution in the syngas and flue gas streams. This efficiency allows a single CCS unit to capture a large volume of emissions effectively. In contrast, the SMR method may require multiple CCS units to achieve up to 90 % capture of onsite emissions. Thus, ATR and POX methods are preferred for carbon capture due to their simpler CO₂ removal processes than the SMR method [50,51].

Our analysis further reveals that H₂ production via the POX+CCS and ATR+CCS methods exhibit, on average, 20 % lower CO₂ intensity compared to the SMR+CCS method when generating electricity. Nevertheless, the most environmentally friendly approach for electricity generation from H₂ chain is through green H₂, produced via water electrolysis, which eliminates methane (CH₄) emissions and utilizes low-carbon power sources. The CO_{2eq} intensity for this method ranges between 22.8–123 gr-CO_{2eq}/MJe. In terms of CO₂ intensity, electricity generation from blue methane (CH₄ + CCS) is comparable to green H₂, emitting nearly 2.5 times less CO₂ than the blue H₂ option. The data for the blue CH₄ route is adapted from the work of Farajzadeh et al. [23], with an adjustment to include 0–1.5 % of fugitive CH₄ emissions for consistency. Moreover, the CH₄ + CCS option also demonstrates higher exergetic efficiency compared to the H₂ supply chain, with ranging from 0.76 to 2.58 for electricity generation. This enhanced efficiency is primarily because CH₄ serves as a primary energy source, with relatively low production costs from an exergy perspective [23].

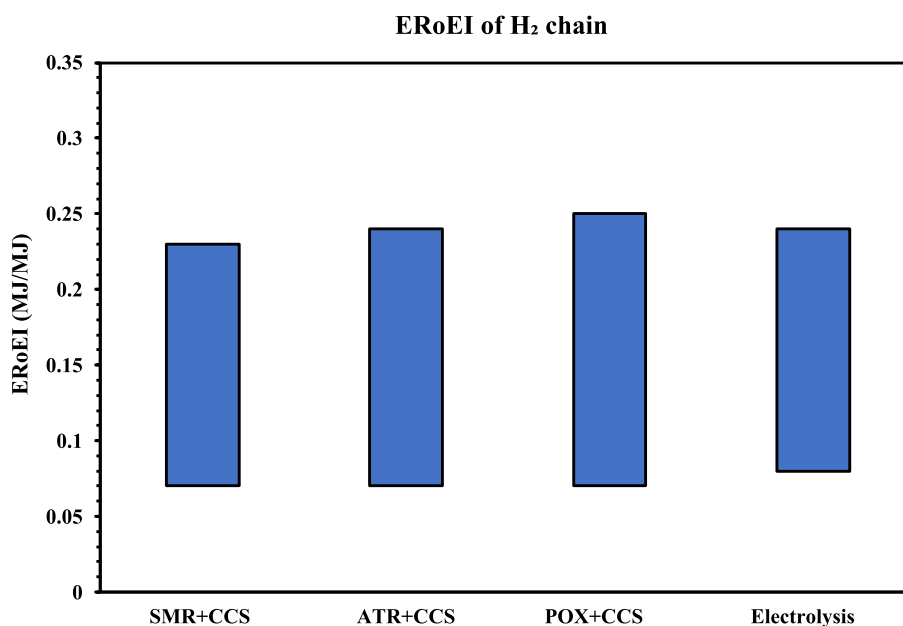


Fig. 3. The exergetic efficiency (ERoEI) of the full H₂ supply chain shown in Fig. 1 including H₂ production, underground storage, and application processes considering different H₂ production methods.

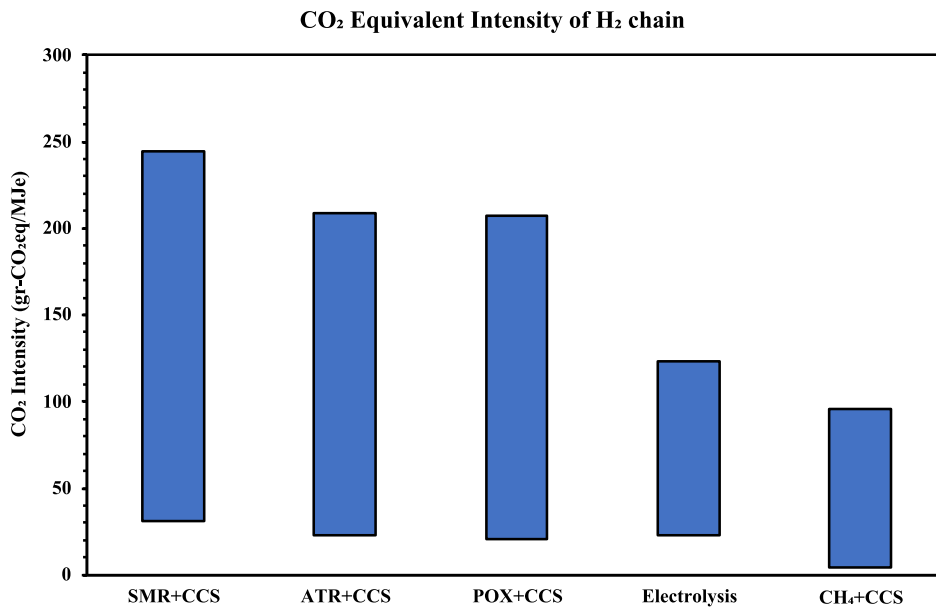


Fig. 4. CO₂eq Intensity of the full H₂ supply chain including H₂ production, underground storage, and application processes for different H₂ production methods and considering CO₂, CH₄ and H₂ emissions. Results are compared with the CH₄ + CCS option.

Fig. 4 incorporates the impact of fugitive methane (CH₄) and hydrogen (H₂) emissions on the system’s CO₂eq intensity, applying Eq. (3) for calculation. Despite CH₄ leaks occurring less frequently than H₂ within the system, the global warming potential (GWP) of CH₄ is significantly higher, leading to a greater effect on greenhouse gas emissions. Specifically, a fugitive emission rate of 1.5 % for CH₄ can augment the CO₂eq intensity of electricity generation by 11–25 %. Should the CH₄ leakage rate escalate to 3 %, this increase in total CO₂eq intensity could range from 24–49 % over a 100-year period. Additionally, the impact of CH₄ leakage is also evaluated over shorter durations, such as a 20-year period in some studies, where the GWP of CH₄ is recognized to be 86 [45]. In such a scenario, 1.5 % CH₄ leakage could elevate the CO₂eq intensity by 48–69 %. In comparison, leakage rates of 3.5–9 % H₂ in the system contribute to an increase the total CO₂eq intensity by 4–20 gr-CO₂eq/MJe, translating to an increase of 5–23 % over a 100-year period.

Fig. 5 and Fig. 6 present the average contribution of the invested exergy and the corresponding specific CO₂ emissions with blue and green H₂ production methods, respectively. In each case, the predominant share of total exergy is allocated to the H₂ production phase, comprising 56 % for blue H₂ and 67 % for green H₂. The elements of

subsurface storage and electricity generation are responsible for about 6 % and 25 % of the overall exergy investment, respectively. Moreover, within the blue H₂ pathway, 13 % of the exergy is expended during the CO₂ capture process. A similar contribution is observed for the CO₂ footprint of the system. The emission from the H₂ production stage accounts for a big portion of the total emission in both green and blue H₂ options, with 76 % and 84 %, respectively. The CCS stage in the blue H₂ option emits 13 % of the total CO₂ emissions. Operations involved in subsurface storage, including compression, separation, and handling of tail-gas, are estimated to contribute 6–7 % of the emissions. The analysis also indicates that transportation components emit a comparatively small amount of CO₂, ranging from 1–2 %.

2. Underground Storage Process

As depicted in Fig. 2, the underground hydrogen storage (UHS) process is composed of three primary components: injection, gas processing, and tail-gas handling. The term “loss” refers to the amount of H₂ that remains trapped within the reservoir due to either mixing or hydrodynamic/geochemical interactions. This retained H₂ reduces the exergy return but does not contribute to H₂ emissions. Eq. (1) illustrates how H₂ loss within the reservoir directly impact on the Exergy Return on Exergy Invested (EROEI), indicating that any reduction in exergy return

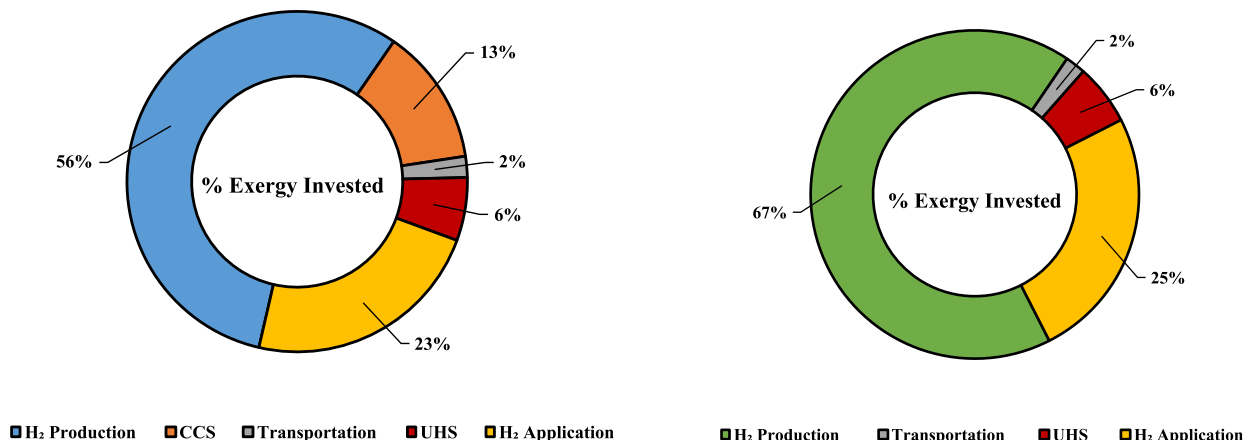


Fig. 5. Fraction of the exergy investment with blue (left) and green H₂ (right) production methods.

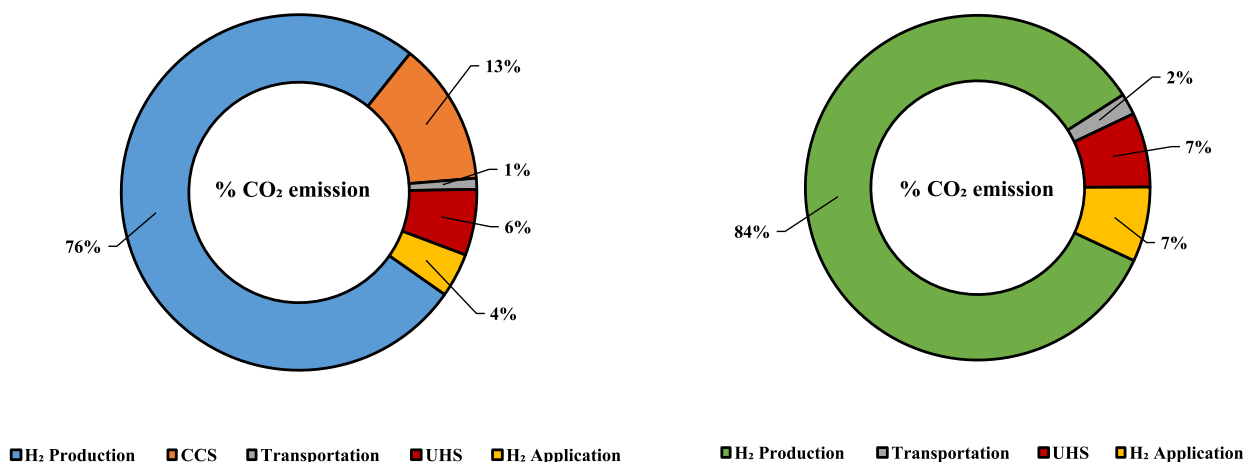


Fig. 6. Fraction of the CO₂ emission with blue (left) and green H₂ (right) production methods.

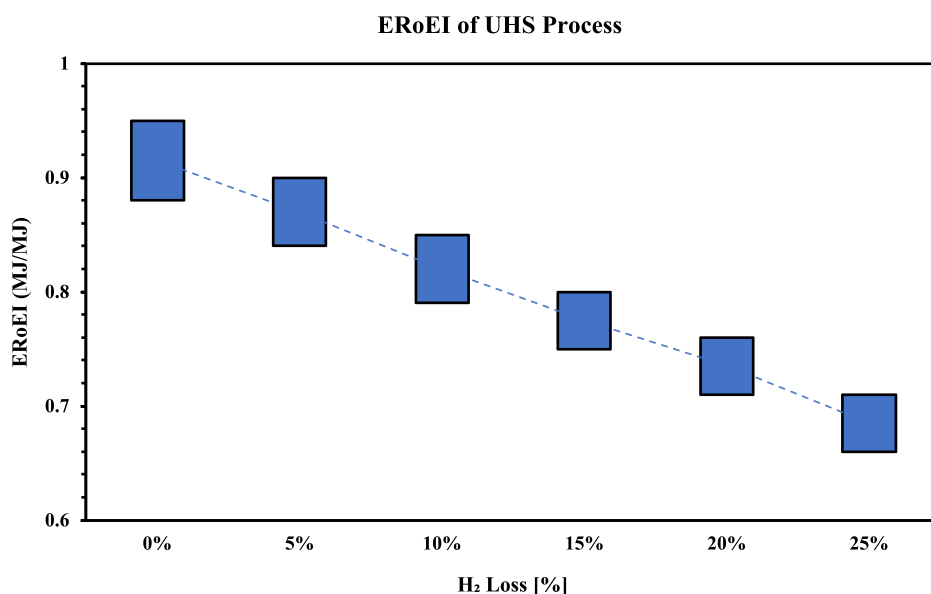


Fig. 7. Exergy return on the exergy investment (ERoEI) as a function of H₂ loss for the UHS process.

due to H₂ loss leads to a decrease in ERoEI. The exergetic efficiency for the UHS component is estimated to be between 0.72 and 0.92.

Fig. 7 further elucidates the influence of H₂ loss on the exergetic efficiency of the UHS process. It is evident that the exergetic efficiency diminishes as H₂ loss increases. For instance, the average ERoEI decreases from 0.91 – with no H₂ loss (equating to 100 % H₂ recovery factor) to 0.78 for a 15 % H₂ loss (equating to 85 % H₂ recovery factor).

Corresponding to the exergetic efficiency, the CO_{2eq} intensity of the UHS is calculated to range 1.46–4.56 gr-CO_{2eq}/MJ, under the assumption that low-carbon energy sources, such as wind power, are utilized for all subsurface operations. Studies indicate that surface operations typically experience a 1–2 % H₂ leakage rate (Table 5).

Fig. 8 shows the relationship between CO₂ intensity and different H₂ leak rates within UHS process. The results indicate that H₂ leak has a considerable impact on the CO_{2eq} intensity, which rises significantly with higher leakage rates. For example, the CO_{2eq} intensity increases from 2.4 gr-CO_{2eq}/MJ to 14 gr-CO_{2eq}/MJ as the H₂ leak rate increases from 1 % to 10 %.

The choice of tail gas handling does not have a significant impact on the exergetic efficiency of the overall system. Scenario 2, i.e., which involves export of CH₄ to a gas power plant, yields a slightly higher exergy gain but also a slightly increased CO₂ intensity compared to

Scenario 1, where CH₄ is re-injected into the storage reservoir. The reason for this is that in Scenario 2 the conversion of the chemical exergy of CH₄ into electricity results in a larger exergy gain; however, this conversion process is accompanied by additional CO₂, which needs to be captured. Fig. 9 and Fig. 10 present the average distribution of the exergy investment and the unit CO₂ emitted over the scenarios in a comparative manner. In both scenarios, the injection phase, particularly the compression stage, accounts for the largest portion of exergy consumption, with figures at 49 % and 48 %, respectively. The gas processing segment, which includes separation, is the second-largest contributor, with 42–44 %. When comparing scenarios, in Scenario 2 (exporting CH₄ to gas power plant and capturing generated CO₂ from combustion) CCS process demands a slightly higher exergy investment than in Scenario 1 (reinjection of CH₄ back into reservoir). Furthermore, larger exergy investment corresponds to a larger fraction of CO₂ emission. Consequently, the injection stage produces the highest CO₂ emission in Scenario 1. Nonetheless, despite the lower exergetic cost of tail-gas handling in Scenario 2, this stage accounts for 41 % of the total CO₂ emissions. This arises from the CCS process and combustion of the exported CH₄.

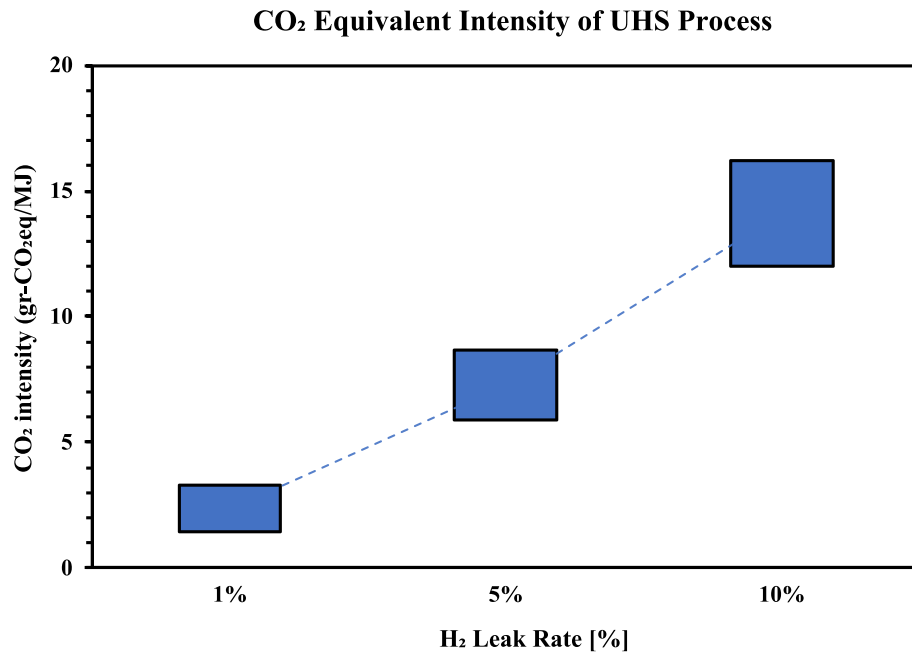


Fig. 8. Impact of H₂ leak rate on CO₂eq intensity in UHS process.

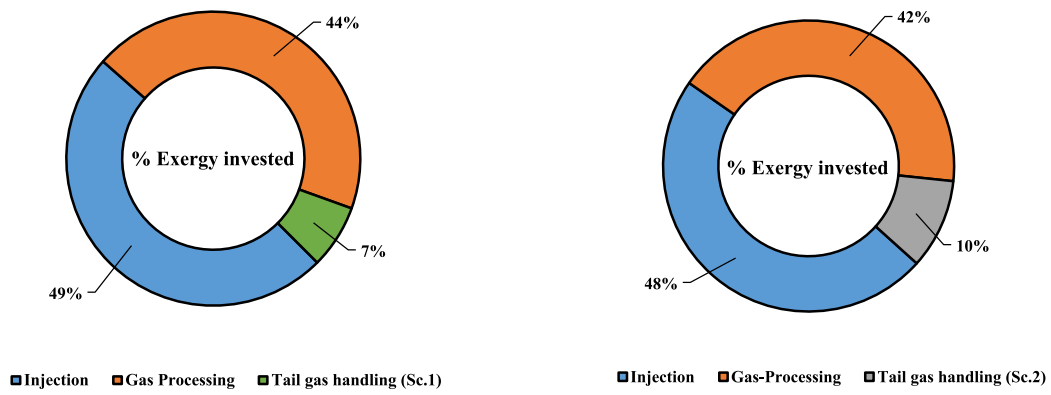


Fig. 9. Fraction of exergy investment in the UHS process with different tail-gas handling scenarios (referring to Fig. 2).

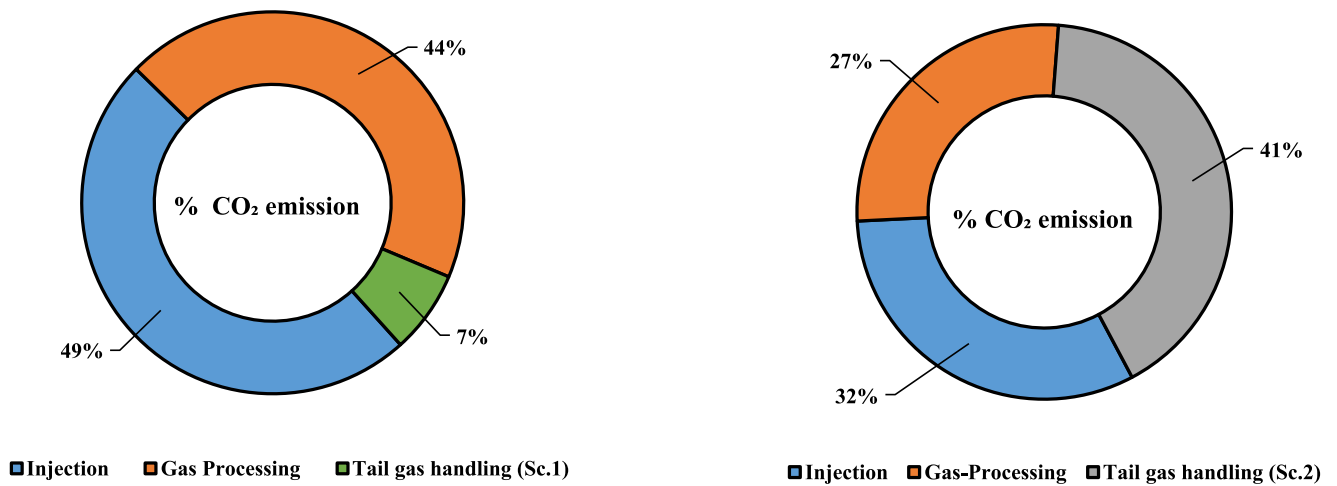


Fig. 10. Fraction of the CO₂ emission in the UHS process with different tail-gas handling scenarios (referring to Fig. 2).

Conclusions

In this study, the concept of exergy-return on exergy-investment (ERoEI) is used to estimate the exergetic efficiency of electricity generation via underground H₂ storage process, examining different H₂ production methods. Through detailed analysis, the ERoEI and CO₂ equivalent intensity (gr-CO_{2eq}/MJe) are calculated for the entire H₂ supply chain, including the underground storage option. Based on the system's assumptions and defined boundaries, we draw the following conclusions:

- The Partial Oxidation (POX) and Autothermal Reforming (ATR) methods exhibit the highest exergetic efficiency (ERoEI) throughout the process. These methods also offer lower CO₂ intensities in comparison to the Steam Methane Reforming (SMR) method.
- Although electrolysis using renewable power being an energy intensive process, it has the lowest CO₂ intensity for electricity generation among the H₂ production methods.
- H₂ loss at any stage of the process significantly reduces ERoEI of the system.
- Integrating a CO₂ capture plant during the production phase of blue H₂ significantly reduces the CO₂ intensity of the entire supply chain, by around halving it, albeit at the expense of reduced exergetic efficiency (ERoEI).
- The blue methane (CH₄ + CCS) option demonstrates higher exergetic efficiency and lower CO₂ intensity compared to the blue H₂ option. However, blue H₂ is a promising route for electricity generation, which can help decarbonize the energy sector.
- The inclusion of CH₄ emissions increases the overall CO₂ equivalent intensity of blue H₂, with the magnitude depending on the leakage rate. For 1.5 % CH₄ leakage rate, the CO₂ intensity increases by 25 % (100 years' time frame).
- The exergetic efficiency of the underground hydrogen storage process is calculated in the range of 72–92 %. The largest exergy investments are consumed for injection components.
- Utilizing low-carbon power sources in subsurface storage operations contributes minimally to CO₂ emissions, estimated between 1.46–4.56 gr-CO_{2eq}/MJ.

CRedit authorship contribution statement

Boyukagha Baghirov: Writing – original draft, Visualization, Methodology, Investigation, Conceptualization, Formal analysis. **Denis Voskov:** Writing – review & editing, Supervision. **Rouhi Farajzadeh:** Writing – review & editing, Supervision, Project administration, Conceptualization, Methodology, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgement

The authors thank Shell Global Solutions International for granting permission to publish this paper. The authors also thank Prof.dr. Hadi Hajibeygi for reviewing the initial draft of this manuscript.

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