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Biohydrogen production through biomethane steam reforming with CCUS for decarbonizing Spain's tile industry

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ABSTRACT

Renewable hydrogen production is a fundamental element in the pathway towards industrial decarbonization. While electrolysis is the primary method for producing renewable hydrogen, there is considerable potential in using renewable gases to complement this process. By upgrading biogas from anaerobic digestion of organic waste to biomethane and feeding it into a steam methane reforming facility where biogenic CO_2 is captured, biohydrogen with negative emissions (HyBECCS) can be produced. This study focuses on the decarbonization potential of HyBECCS, specifically in the Spanish tile sector, assessing HyBECCS/natural gas and biomethane/natural gas blends. Results show that HyBECCS blends save over 37 % of biomethane compared to biomethane/natural gas blends for the same emissions reduction. A 50 % HyBECCS/natural gas blend is proposed, which requires 4.7 TWh of biomethane to meet the tile sector's demand, representing less than 3 % of Spain's total biomethane production potential. The cost analysis reveals that this 50 % HyBECCS blend, achieving a 53.4 % reduction in emissions, is competitive with pure natural gas when natural gas prices exceed 16.5 €/MWh, when biomethane comes from the organic fraction of municipal solid waste. This blend always exhibits lower costs than natural gas if biogas comes from landfills.

Acronyms

BECCS	BioEnergy Carbon Capture and Storage
bio-CH_4	Biomethane
CAPEX	CAPital EXpenditure
CCS	Carbon Capture and Storage
CCUS	Carbon Capture, Usage and Storage
CEPCI	Chemical engineering plant cost index
EBA	European Biogas Association
EU	European Union
EUR	Euro
f-CH_4	Fossil methane
GDP	Gross Domestic Product
GO	Guarantee of Origin
HHV	Higher Heating Value
HOMER	Hybrid Optimization of Multiple Energy Resources
HyBECCS	Biohydrogen Carbon Capture and Storage
IEA	International Energy Agency
IEAGHG	IEA greenhouse gas R&D programme
IGME	Spanish Geological Survey
LCOH	Levelized Cost of Hydrogen
LHV	Lower Heating Value
NIECP	National Integrated Energy and Climate Plan
OFMSW	Organic Fraction of Municipal Solid Waste
OPEX	OPerational EXpenditure
SEDIGAS	Spanish Gas Producers Association
SMR	Steam Methane Reforming
TRL	Technology Readiness Level

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Symbols

Variable	Units	Meaning
CAPEX	€/kg H_2	Capital expenditure
$CELF_x$	p.u.	Constant escalation levelization factor of the x-th item
CRF	$year^{-1}$	Capital recovery factor
C_{x0}	€	Cost of x-th item in the year zero
DP	kg/day	Daily hydrogen production
INV	€	Investment of a SMR plant
INV_{CCS}	€	Investment of a SMR with CCS plant

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INV_{noCCS}	€	Investment of a SMR without CCS plant
N	years	Lifespan of the project
$OPEX_{bm}$	€/kg H_2	OPEX due to biomethane (biogas, upgrading and injection)
$OPEX_{CO_2}$	€/kg H_2	OPEX due to transport, injection and carbon tax of CO_2
$OPEX_{om}$	€/kg H_2	OPEX due to operation and maintenance
P	kg H_2/year	Annual hydrogen production
r_x	p.u.	Nominal escalation rate of the x-th item
$Wacc$	p.u.	Weighted average capital cost

1. Introduction

The decarbonization of European economies is crucial for meeting the goals outlined in the European Green Deal [1] approved in December 2019, aiming for climate-neutrality by 2050. In Spain, the industrial sector stands as the second-largest contributor to the Gross Domestic Product (GDP), accounting for approximately 17.6 % in 2022, after the service sector [2]. Natural gas currently represents 55 % of the total energy consumption in the Spanish industry [3], playing a fundamental role particularly in thermo-intensive subsectors like refining, chemical-pharmaceuticals, and construction. Furthermore, its role in facilitating the substitution of less environmentally sustainable fuels has been notable in the industry's gradual decarbonization efforts in recent years. However, further decarbonization of the sector is imperative to meet the targets and to safeguard the long-term competitiveness of businesses. Additionally, this effort will enhance the resilience of the Spanish economy and diminish its reliance on fossil fuels.

Electrification with renewable energies as a form of decarbonization is not always the best option, especially in the short-term. Fraunhofer ISI [4] assessed the electrification potential of the energy demand not yet electrified in the entire industrial sector until 2030 at 62.4 %, based on 2019 energy demands in the EU-27. This share falls to 35 % in the case of the non-metallic minerals industry, including cement, glass and ceramics. Current electrification technologies (resistance heating, plasma heating and shock-wave heating) require further technological development to be applied in these sectors. The same authors mentioned that the difficulties found in the electrification of these sectors put pressure on the transition towards carbon capture, thus delaying the decarbonization of the energy supply. Additionally, hydrogen from renewable electrolysis is proposed as an indirect electrification method, which should be taken into account. In this context, a review of water electrolysis technologies is performed by El-Shafie [5], focusing on prices and technical barriers associated with different electrolyzer types. An assessment by IRENA [6] indicated that electrification is expected to play a significant role in the future in hard-to-abate sectors. However, this will primarily involve technologies that are still in their early stages, although some have already reached or are close to technological maturity. Furthermore, indirect electrification through the production of renewable hydrogen is again recognized as an important factor in achieving emissions reductions in these sectors. De Bruin et al. [7] also determined that the technology for electrifying high-temperature heat in energy-intensive industries generally requires further development. They also highlighted an important point: if the electricity sector has not reached carbon neutrality, increasing electricity demand may delay the shutdown of fossil fuel-based power plants, which will not help achieve climate neutrality.

As an alternative to electrification by renewable energies, renewable gases such as biomethane and green hydrogen are expected to replace conventional natural gas in thermo-intensive industrial processes. This is stated by the Spanish Government in [8] for biomethane and in [9] for hydrogen, where not only hydrogen from renewable electricity is considered, but also biohydrogen from biomethane. The development of renewable gases is presented as an efficient lever to ensure a sustainable future for the industry, and could even be a key factor in attracting new industry seeking competitive sustainable energy. For energy-intensive industries, de Bruin et al. [7] suggested that hydrogen can be integrated into fossil fuel-fired furnaces with only minimal adaptations to the burner and fuel system. As an intermediate step, hydrogen and natural gas blends can lead to cleaner and more efficient combustion due to the properties of the mixture, potentially reducing emissions by half [10].

Targets of renewable hydrogen production have been defined at national and international levels. The EU objectives amount to 20 Mt of H_2 by 2030 [11]. Focusing on Spain, 4 GW of electrolyzers are planned to be installed by 2030 as stated in the National Hydrogen Roadmap [9]. In the June 2023 draft revision of the National Integrated Energy and Climate Plan (NIECP) this objective was revised and raised to 11 GW, within the same time frame [12].

On the other hand, Spain has significant potential for generating biogas and biomethane from organic waste. According to the latest comprehensive study conducted by the Spanish Gas Producers Association (SEDIGAS) [13] this potential is estimated at 163 TWh/year. This figure is roughly supported by other sources such as EBA, which projected the potential at up to 210 TWh/year [14]. Similarly, ENGIE estimated it at 133 TWh/year [15], and the European Commission at 122 TWh/year [16]. The current production (2024) of biomethane for natural gas grid injection in Spain is 407 GWh/year from 9 plants, according to [17], with 23 new facilities under construction that will be able to additionally produce 1040 GWh/year. The production of biogas occurs through anaerobic digestion, serving as the initial stage in biomethane production. Although this is a well-established process, typically achieving efficiencies between 72 and 85 % [18], extensive research is still being conducted. For instance, Nayeri et al. [19] demonstrated that by applying new physical, chemical, and biological pretreatment techniques, it is possible to improve the overall efficiency of the anaerobic digestion system, maximizing methane yield. Moreover, for distributed production, Zainab B. et al. [20] conducted a study using a small-scale biogas digester to enhance biogas production from the anaerobic digestion process. After the biogas is produced, it is converted into biomethane. This conversion, referred to as upgrading, involves removing impurities and CO_2 from the biogas, resulting in nearly pure methane, indistinguishable from natural gas.

Currently, most of the global hydrogen production originates from steam methane reforming (SMR) of natural gas. This method accounted for 62 % of the total global production in 2021 [21]. According to the European Hydrogen Observatory [22], in 2022 in Europe, SMR alone was responsible for 90.8 % of the hydrogen production, with three additional SMR plants incorporating carbon capture and storage (CCS), accounting for 0.2 % of the production. In Spain, production in 2022 was 614 kt/year, with SMR having a share of 93.5 %. There are no CCS facilities in Spain.

The steam methane reforming process itself yields a gas mixture containing predominantly hydrogen (around 75 %) along with varying proportions of methane (2–6 %), carbon monoxide (7–10 %), and carbon dioxide (6–14 %) [23]. Additionally, the water–gas shift reaction is employed to convert carbon monoxide into hydrogen and carbon dioxide. The two aforementioned reactions are represented in Eqs. (1) and (2).

$$CH_4 + H_2O \rightarrow CO + 3H_2 \tag{1}$$

$$CO + H_2O \rightarrow CO_2 + H_2 \tag{2}$$

This type of hydrogen produced from SMR of natural gas is referred to as grey hydrogen, and emits approximately 9 kg of CO_2 per kg of hydrogen produced, considering an efficiency of the SMR process (without CCS) of 75.9 %, mainly due to the use of additional methane in combustion to maintain the required process temperature [24]. If the SMR is coupled with a carbon capture and storage (CCS) unit, those emissions are captured, and the obtained hydrogen is known as blue hydrogen. CCS processes can achieve over 95 % CO_2 capture as reported in [25].

To decarbonize the hydrogen production, the authors have proposed to replace natural gas with biomethane, resulting in what is known as biohydrogen [26]. This biohydrogen is classified as renewable [27] or green [28], and it could potentially lead to negative emissions [26]. This phenomenon arises from the fact that biomethane, obtained from organic waste, retains carbon originally absorbed from the atmosphere by plants. Eventually, these plants are transformed into organic waste, with both plants and livestock serving as intermediary agents in this process. In this sense, the capture might be denoted as "pre-combustion" technology. If the CO_2 captured during the SMR process is stored (CCS), negative emissions are achieved because the CO_2 is effectively removed from the atmosphere. These negative emissions might be used by hard-to-abate sectors to offset their unavoidable emissions. The hydrogen produced in this way is referred to as HyBECCS [29].

For the development of HyBECCS, the scalability and maturity of the technology associated to SMR with CCS play a pivotal role. Since 2003, when Lipman detailed investment costs for several plants of different scales [23], improvements in SMR with CCS have been notable. Three plants producing blue hydrogen have been reported by IEA from 2013 to 2020 [30]. Investment costs have substantially decreased, as corresponds with the high technological maturity. Rosa et al. [31] recently assigned high technology readiness levels to SMR (9) and SMR with CCS (7–8). As an example, a plant of 214,286 kg H_2/day of capacity would require in 2020 an investment of EUR 308.96 million [24], while a similar size plant would have required around 2.46 times more investment in 2003,[1] reflecting the learning process over nearly two decades [32].

Fig. 1 shows the most important processes involved to obtain HyBECCS, divided into four different stages: biogas production, upgrading to biomethane, hydrogen production and CCS.

The scheme proposed is based on a comprehensive examination of the different phases involved in HyBECCS production: biogas generation, subsequent upgrading, injection of biomethane into the natural gas grid, centralized hydrogen production hubs with CO_2 capture, and ultimately, the transportation and storage processes in geological formations. Leveraging existing infrastructure, mature technologies, and stable feedstock pricing, the model ensures economic feasibility. A thorough assessment of economic viability, using the levelized cost of hydrogen (LCOH), has been detailed in [32].

The concept of negative emissions associated to BECCS (bioenergy with carbon capture and storage) or, specifically when hydrogen is produced, HyBECCS, is drawing considerable interest from the research community. In the case of biomethane, EBA [33] proposed CO_2 capture and storage in both its production and use, as a technique to enhance the reduction of emissions, being a cost-competitive technique compared with other renewables. They proposed three ways to obtain these negative emissions: with special agriculture practices, storing the CO_2 captured in the upgrading process (both methods would be pre-combustion technologies) and capturing and storing the CO_2 released in the final combustion (post-combustion). Odunlami et al. [34] performed a review in this area, identifying advanced and modern techniques for capturing CO_2 and detailing their efficiencies and cost-effectiveness. Regarding HyBECCS, Rosa et al. [31] highlighted its potential in Europe when hydrogen is derived from biogas by SMR with CCS, assigning, as aforementioned, a TRL of 9 to SMR and a 7 to 8 to SMR with CCS and highlighting the maturity of this technology. The study also analyzed the integration of production and demand within the supply chain. Following a similar scheme, Antonini et al. [35] performed a life-cycle analysis, showing that if the digestate (by-product of the biogas production, Fig. 1) is utilized as fertilizer, the hydrogen production reaches negative emissions even if there is no CCS in the process. In this context, Moioli et al. [36] suggested the production of renewable gases using biomass through two distinct pathways: biomass gasification to yield syngas, subsequently converted into hydrogen, and anaerobic digestion of organic waste to produce biogas. Biogenic CO_2 is generated in both scenarios, offering the option for either storage to achieve negative emissions or integration with hydrogen derived from electrolysis using renewable sources to generate biogenic synthetic methane. Lefranc et al. [37] investigated the use of HyBECCS to fuel a bus fleet in Madrid. They demonstrated that the negative emissions from this process could save biomethane when buses equipped with fuel cells are powered by HyBECCS. Full et al. [29] compared the production of biomethane and biohydrogen with carbon dioxide capture and storage as BECCS approaches. While both have the potential to reduce GHG emissions and increase energy security, they reported up to five times higher carbon removal potential in the hydrogen-production approach (HyBECCS).

A cornerstone in this negative emissions scheme is the CO_2 supply chain management, as the European Commission recognized [38], outlining a strategy for managing CO_2 emissions. This strategy involves capturing and storing CO_2 from both fossil and biogenic sources, as well as direct atmospheric removal and utilization of captured CO_2 for industrial purposes. However, challenges remain, including regulatory issues and the lack of recognition for negative emissions within the European Trade System, hindering market incentives for the process. Additionally, a recent report from the Commission [39] highlighted the need for a transport network to manage captured CO_2, estimating a required investment range and recommending storage capacities to be located in specific regions to reduce costs. Cross-border issues, particularly concerning quality standards for transport and storage, are also addressed.

CO_2 capture, along with its use and storage (CCUS) [30], are key technologies in the global decarbonization strategy. These technologies not only allow the reduction of CO_2 emissions from large point sources but also offer the possibility of reusing captured CO_2 in various processes, including the manufacturing of building materials [40]. In Europe, there is a strong impulse for the development and implementation of this technology, being considered for incorporation into the National Energy and Climate Plans [41], with initiatives such as the European Union's proposal for an annual CO_2 injection target and the improvement of permitting procedures for CCUS projects [42]. The United Kingdom, for example, announced significant investment for the early deployment of CCUS projects [43]. Additionally, CO_2 carbonation is being investigated as a viable alternative for CO_2 management [44]. In order to achieve an appropriate scale of production, guarantee of origin certificates [45] may be employed to link the distributed generation of biomethane with centralized SMR production, situated close to the hydrogen consumer and at an intermediate distance to the CO_2 geological storage site or to the industrial applications that utilize it. Sun et al. [46] identified fifteen highly favorable and feasible geological structures in Spain after conducting an analysis to assess the potential and feasibility of CO_2 storage.

Other methods for producing renewable hydrogen are well documented in the literature. Aravindan et al. [47] reviewed different technologies, including biomass and renewable water splitting. For biomass, they focused on solid biomass and concluded that the technology is still immature, encountering difficulties with carbon capture. However, they noted advantages in biohydrogen. Nemitallah et al. [48] expanded on this review, examining costs and presenting case studies. Regarding SMR, they highlighted production scale as a key cost factor. It is notable that these reviews do not include biomethane as a feedstock for SMR. Both reviews consider CCS, but only for capturing fossil CO_2 in the SMR process or producing biogenic CO_2 from solid biomass.

Currently, hydrogen from water electrolysis using renewable energies is proposed as a decarbonization option, requiring the study of its integration with the electrical grid. Tan et al. [49] proposed a day-ahead dispatch strategy for electricity-hydrogen systems under the solid-state transportation mode of hydrogen energy. They addressed the uncertainty issue using the information-gap decision theory envelope

[1] Using correlations derived from Lipman [13] and scaling it to 2020.

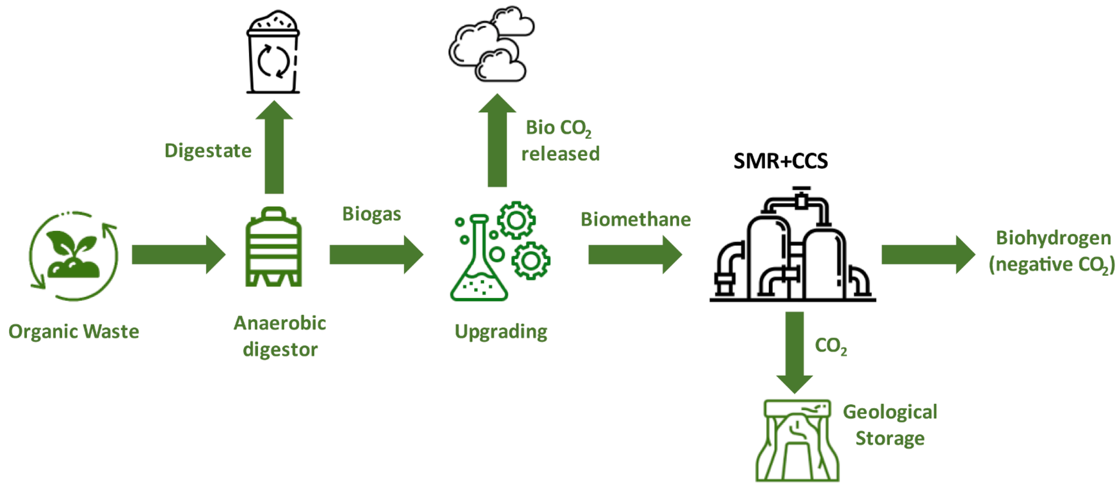


Fig. 1. Biohydrogen with negative emissions (HyBECCS) production stages [32].

constraint. It is also possible to take advantage of using electricity to produce hydrogen, as proposed by Xia et al. [50] with the co-supplying of a remote micro-community with electricity and hydrogen. They used HOMER (Hybrid Optimization of Multiple Energy Resources) to analyze the system, including its economic feasibility. In another study [51], Xia et al. assessed the co-production of electricity and hydrogen from wind, taking Lutak (Iran) as a case study. They showed a detailed procedure to calculate the levelized costs of electricity and hydrogen from wind, including environmental and technical aspects of wind turbines deployment. In the case of hydrogen, the focus is on the production itself, with the remainder of the supply chain reserved for future analysis.

Regarding the long-term integration of hydrogen in energy systems, Quion et al. [52] proposed a multi-objective optimization model for integrated energy hubs and multi-energy networks that incorporates hydrogen energy. This model focuses on minimizing total annual energy costs and CO_2 emissions, demonstrating the benefits of dynamic hydrogen pricing and integrated demand response to enhance economic and environmental performance. Similarly, Yanbin et al. [53] developed a collaborative operational model for shared hydrogen energy storage within a park cluster, employing a distributionally robust optimization approach. This model quantifies economic, environmental, and reliability benefits, proving its effectiveness in enhancing energy utilization efficiency and reducing system operating costs while addressing uncertainties in renewable energy output. Additionally, Plazas-Niño et al. [54] provided a comprehensive techno-economic assessment of hydrogen pathways in Colombia, presenting an updated proposal for the National Hydrogen Roadmap. This work highlights hydrogen's potential to significantly reduce greenhouse gas emissions and foster socio-economic benefits through the deployment of low-emission technologies such as electrolysis and biomass gasification. Furthermore, Gabbar et al. [55] enhanced the SWITCH model by incorporating a module for optimizing hydrogen plants, balancing water demands, and integrating with existing optimization platforms for comprehensive energy planning, emphasizing sustainable deployment of hydrogen infrastructure. The reviewed studies collectively emphasized the critical role of hydrogen energy storage in enhancing energy utilization efficiency, reducing greenhouse gas emissions, and fostering economic benefits through various optimization models. They explored the techno-economic assessments, integration strategies, and multi-value quantifications of hydrogen within energy systems, focusing on innovative frameworks for optimizing hydrogen production, storage, and deployment. Unlike these studies, our research does not propose an additional optimization model. Instead, it provides a practical application of hydrogen implementation in a specific industrial sector, namely the tile sector in Spain, showcasing real-world deployment and operational insights. This approach aims to bridge the gap between theoretical models and actual industrial practice, highlighting the feasibility and tangible benefits of biohydrogen energy in a targeted industry context.

In this paper, a case study has been conducted to study the effectiveness in reducing emissions of biomethane and HyBECCS in hard-to-electrify sectors in Spain. Thus, the model developed by the authors has been applied to a specific industrial sector: the tile sector in Spain, which is a suitable representative of the thermo-intensive sectors. In [3], it is demonstrated how the increase in natural gas prices caused a sharp drop in the tile sector's production in 2023, highlighting the urgency of transitioning to more sustainable energy solutions.

The study focuses on assessing and comparing the required biomethane in biomethane/natural gas and HyBECCS/natural gas blends to achieve the same level of decarbonization. Then, a specific assessment of the required biomethane and natural gas to meet the demand of the tile sector is conducted for the two blends. Considering that there will be a technical limit in the tile furnaces of around 50 % hydrogen concentration in the short term, this blend is assumed as the base case for a sensitivity analysis of natural gas price. As the price of HyBECCS largely depends on the source of the biogas, two cases are analyzed: biogas from the organic fraction of municipal solid waste and from landfills.

Despite the significant potential of renewable hydrogen and carbon capture technologies, several gaps remain in the current scientific understanding. The adoption of CCS technologies in hydrogen production from biomethane is limited, primarily due to regulatory challenges and the lack of market incentives for negative emissions. Additionally, there is insufficient research on the scalability and economic feasibility of large-scale HyBECCS production, as well as the effective integration of renewable hydrogen into existing natural gas infrastructure. This manuscript addresses these gaps by proposing hydrogen production from biomethane with CCS to achieve negative emissions. This concept is applied to the Spanish tile sector, providing a novel case study that assesses the decarbonization potential and economic feasibility of natural gas blends with this type of renewable hydrogen, compared to blends of natural gas with biomethane producing the same decarbonization effect. The comprehensive economic analysis carried out, including a sensitivity analysis on natural gas prices, illustrates the conditions under which this hydrogen can be a viable and cost-effective solution, providing valuable insights for policymakers and industry stakeholders.

The paper is organized as follows: Section 2 outlines the methodology, with further details provided in [32], where the costs model is fully developed. Section 3 describes the results, beginning with general findings, applying these findings to the tile sector's demand, and concluding with a sensitivity analysis on natural gas prices. Section 4 summarizes the conclusions.

2. Methodology

One of the main characteristics of the Spanish tile industry is its high geographical concentration in the province of Castellón, configuring an industrial cluster. Approximately 94 % of the national production originates in this province, where 80 % of the sector's companies are located [56]. This location concentration facilitates the implementation of a hydrogen valley, integrating the production and demand of hydrogen. Additionally, within a distance of less than 250 km, there are potential CO_2 geological storage sites with an estimated capacity of at least 1.7 Gt CO_2 [57]. Specifically, the "Maestrazgo-1″ geological storage site, located 70 km from the Castellón refinery, is a potential candidate for storing captured CO_2. With a storage capacity of 86 Mt of CO_2, it has been assigned a medium-quality rating by the Spanish Geological Survey (IGME). For higher quality and larger capacity needs, the "Zona Enlace–Bunt" storage site in Teruel province, also approximately 70 km from Castellón, could be considered. This site has a capacity of 1212 Mt of CO_2 and is rated as high quality. It could also serve as a storage location for CO_2 captured from other industrial hubs, such as the chemical hub in Tarragona. Both storage sites are part of IGME's selected areas and favorable geological structures for CO_2 storage in Spain, which are considered safe for both humans and the environment. While there might be social opposition to the location of CO_2 storage sites, it should be noted that the process for selecting these locations involves a comprehensive procedure, including seismic evaluations, as detailed in [46] for Spain.

Regarding biomethane production, a distributed scheme is assumed, leveraging the natural gas grid to transport the biomethane using guarantees of origin certificates, recognized in Spain from 2023 [58]. In this way, the current natural gas grid is used to transport biomethane to the SMR+CCS facility, avoiding the necessity for the facility to be located in close proximity to biomethane producers, thereby enabling the integration of small producers. The entire scheme is illustrated in Fig. 2.

The primary method for creating negative emissions involves capturing the biogenic CO_2 produced by the SMR+CCS process when biomethane is used. This concept is somehow transformed when guarantees of origin certificates are incorporated into the supply chain. Fig. 3 illustrates this process: if a fossil methane (f-CH_4) molecule is used to produce hydrogen, but the SMR+CCS facility pays through the guarantee of origin certificate for a biomethane (bio-CH_4) molecule, this molecule is not used to produce hydrogen, but remains in the natural gas grid. This approach results in a CO_2 void in the atmosphere, producing negative emissions and generating a carbon credit. Later, when a combustion facility needs to burn natural gas and purchases a carbon credit for compensation, the biogenic CO_2 from the biomethane stored in the grid is released, filling the previous CO_2 void and achieving a net-zero overall process. In essence, the guarantee of origin certificate is utilized by the SMR+CCS facility to create a carbon credit for another entity, with the natural gas grid serving as temporary storage (with redemption possible within one year).

Regarding the HyBECCS production, 18.54 t H_2/GWh-HHV CH_4 is obtained, considering 69.1 % efficiency in the SMR with CCS. Assuming a 90 % carbon capture efficiency, 8.64 t CO_2/t H_2 is expected [32]. The higher heating value for methane is taken as 11.04 kWh/Nm^3, whereas the lower heating value is 9.952 kWh/Nm^3.

The levelized cost of hydrogen (LCOH) has been determined using Bejan's formulation [59]. It is divided into capital expenditure (CAPEX) and operational expenditure (OPEX), as described by Eq. (3).

$$LCOH = CAPEX + OPEX \tag{3}$$

The CAPEX is calculated using Eq. (4), where *INV* represents the investment, *P* denotes the annual hydrogen production and *CRF* the capital recovery factor.

$$CAPEX = \frac{INV \cdot CRF}{P} \tag{4}$$

The capital recovery factor is determined by considering the weighted average capital cost (*wacc*) and the lifespan of the project (*N*), according to Eq. (5).

$$CRF = \frac{wacc \cdot (1 + wacc)^N}{(1 + wacc)^N - 1} \tag{5}$$

The OPEX considers the costs of biomethane (*bm*), operation and maintenance (*om*) and the total CO_2 management (CO_2) cost, as described in Eq. (6).

$$OPEX = OPEX_{bm} + OPEX_{om} + OPEX_{CO_2} \tag{6}$$

Each component of the OPEX is obtained from the specific cost of the *x-th* item in the year 0 (C_{x0}), corrected by the cost escalation levelization factor ($CELF_x$), as shown in Eq. (7).

$$OPEX_x = C_{x0} \cdot CELF_x \tag{7}$$

The constant escalation levelization factor is determined by Eq. (8), where k_x represents the ratio defined in Eq. (9). In this equation, r_x denotes the nominal escalation rate for the *x-th* item. It is assumed to be zero for all the components except for the carbon tax. When r_x equals to zero the *CELF* becomes equal to one. Consequently, the OPEX corresponds to the cost at year zero.

$$CELF_x = \left[\frac{k_x \cdot (1 - k_x^N)}{1 - k_x}\right] \cdot CRF \tag{8}$$

$$k_x = \frac{1 + r_x}{1 + wacc} \tag{9}$$

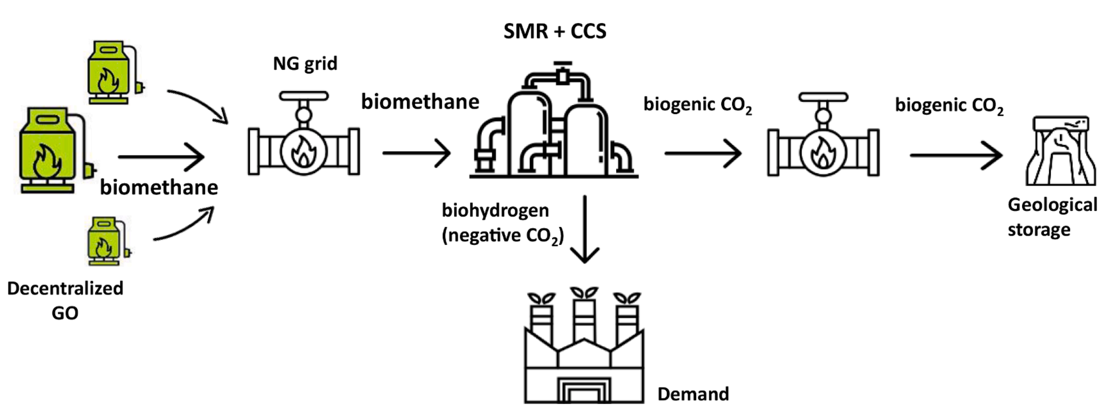


Fig. 2. Supply chain of HyBECCS in Castellón (adapted from [32]).

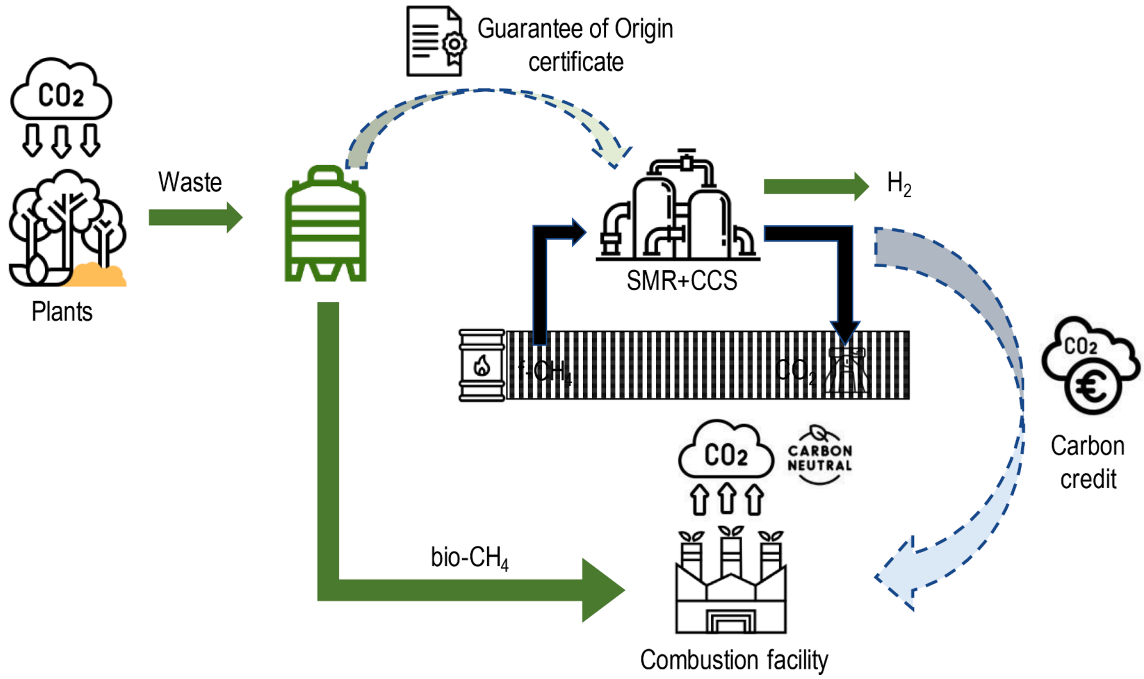


Fig. 3. Transformation of the guarantee of origin certificates in carbon credits with the proposed supply chain.

According to [32], the lifespan of the plant has been assumed to be 25 years, with a weighted average capital cost of 8 % and all nominal rates set to zero except for the carbon tax, which is set at 8 %.

The investment cost, in EUR, for a SMR+CCS plant for hydrogen production is determined based on data from Lipman [23]. Table 1 shows the investment costs for two types of SMR facilities: one without CCS (INV_{noCCS}) and one with CCS (INV_{CCS}) [32]. In Table 1, *DP* refers to the daily hydrogen production.

Based on Table 1, Eq. (10) represents the fit curve for the investment in SMR facilities without CCS, whereas Eq. (11) describes the fit curve for the incremental cost of CCS. The combined use of these equations allows for calculating the investment required for an SMR facility with CCS, as shown in Eq. (12).

$$INV_{noCCS} = 44,068 \cdot DP^{0.713} \tag{10}$$

$$INV_{CCS} - INV_{noCCS} = 6,132 \cdot DP^{0.8592} \tag{11}$$

$$INV_{CCS} = 44,068 \cdot DP^{0.713} + 6,132 \cdot DP^{0.8592} \tag{12}$$

To reflect current conditions, the scaling laws from Eqs. (10) and (12) need to be updated. Instead of using a time scaling factor such as the CEPCI (Chemical Engineering Plant Cost Index) or a similar metric, data from the IEA Greenhouse Gas R&D Programme (IEAGHG) [24] were used. This report provides the 2002 cost of an SMR plant producing 100,000 Nm^3/h (214,286 kg/day) of hydrogen with and without CCS. The investment without CCS was EUR 175.35 million, and with CCS, it was EUR 308.96 million. By maintaining the scale factor and updating the constants in Eqs. (10) and (12), Eq. (13) is derived, with the currency values (euros) adjusted to 2020.

$$INV_{CCS} = 27,727 \cdot DP^{0.713} + 3,511 \cdot DP^{0.8592} \tag{13}$$

Table 2 provides a summary of the operational and maintenance costs for a plant with a capacity of 100,000 Nm3/h, also obtained from the IEAGHG report [24]. Only fixed costs are considered in this table. Since variable costs (such as chemicals, methane, catalysts and makeup water) are primarily due to methane (99.4 %), only this cost has been considered for simplicity. The process typically requires steam, which is often generated by a cogeneration plant that also supplies electricity to the grid. However, the revenue from this electricity supply has not been accounted for. Consequently, the costs from Table 2 result in a relative cost of 0.148 €/kg H_2 for SMR facilities with CCS.

The production of biomethane involves a multi-step process. Initially, organic waste undergoes anaerobic digestion, where microorganisms break down the material in the absence of oxygen, resulting in biogas. The next phase is upgrading, which purifies the biogas by removing impurities and carbon dioxide to increase the methane concentration. The final product, biomethane, is similar to natural gas and can be injected into the natural gas grid. The cost of biogas is influenced by the source of organic waste. Landfill degasification presents the lowest cost range, between 6 and 10 €/MWh-LHV, in contrast with biogas produced from organic fraction of municipal solid waste (OFMSW), which ranges from 30 to 40 €/MWh-LHV [60]. The cost of upgrading depends on the volume flow rate of biogas upgraded. For the average-sized biomethane plant in Spain, 11.96 €/MWh-HHV is

Table 1
Investment costs of SMR facilities without CCS (INV_{noCCS}) and with CCS (INV_{CCS}) [32].

Hydrogen Production (DP) [kg/day]	INV_{noCCS} [USD_{2003}]	INV_{CCS} [USD_{2003}]	INV_{CCS}-INV_{noCCS} [USD_{2003}]
480	3,846,130		
24,000	50,448,185	86,032,633	35,584,448
609,000	605,214,704	1,177,984,909	572,770,205
609,000	601,471,108		
609,000	602,510,996		

Table 2
Operation and maintenance costs for SMR plants with a hydrogen production capacity of 100,000 Nm^3/h (currency in euros, adjusted to 2020 values) [24].

Concept	SMR with CCS [€/year]
Direct labor	2,580,000
Administrative	1,323,590
Insurance	3,053,280
Maintenance	4,579,920
Total	11,536,790

obtained [32]. The costs of injection into the natural gas grid exhibit a similar dependence, and it is estimated in 5.68 €/MWh-HHV [32]. Table 3 summarizes the operational expenditures of biomethane.

Regarding CO_2 management, three steps must be considered: transport to geological storage, injection into the storage and carbon tax. For transportation, a rate of 6.90 €/t CO_2 is considered [42], while 11 €/t CO_2 is taken for injection into deep saline aquifers (the most common structure in Spain) [42]. Considering the ratio of 8.64 tonnes of CO_2 captured per tonne of hydrogen, the combined cost of these two steps represents 0.16 €/kg H_2. A carbon tax of 80 €/t CO_2 has been assumed, with a nominal rate of 8 % [30]. The final step of CO_2 management costs reaches −1.619 €/kg H_2, with the negative value attributed to the biogenic origin of CO_2.

Taking into account the operational costs described, the OPEX of HyBECCS ranges from 0.029 €/kg H_2 when the biomethane derived from landfill gas to 1.34 €/kg H_2 when OFMSW is used. This demonstrates the competitiveness of this alternative compared to hydrogen produced via the conventional route, primarily steam methane reforming of natural gas, which has an estimated cost of 1.5 $/kg H_2, as mentioned by Medhat A et al. [61].

Table 4 summarizes all the model variables and their references. The daily hydrogen production (*DP*) is calculated for 87 kt H_2/year, based on a 50/50 blend of biohydrogen and natural gas to meet a demand of 14 TWh-HHV of natural gas (see Fig. 7).

In 2022, the tile sector in Spain consumed 14 TWh of natural gas, leading to 2.5 Mt CO_2 emissions [56]. Of this gas consumption, 58 % is used in kilns and dryers, 38 % in cogeneration, and the remaining 4 % in atomization [62]. Replacing natural gas with biomethane represents a significant challenge due to the high consumption levels. SEDIGAS estimates an overall biomethane potential of 163 TWh [3], but current and near-term production is well below this potential, amounting to less than 1.5 TWh. On the other hand, considering only landfill and OFMSW as substrates due to their lower feedstock costs, the potential is reduced to 8.81 TWh for landfill and 7.92 TWh for OFMSW [3]. To optimize biomethane use, blending natural gas with a renewable gas is proposed. Two blends have been analyzed in this work: biomethane/natural gas as the baseline and HyBECCS/natural gas. The analysis assesses the biomethane and natural gas amounts needed to achieve equivalent decarbonization per thermal unit delivered for both blends.

3. Results

First, the general results of using HyBECCS/natural gas compared to biomethane/natural gas blends are presented. The quantities of biomethane and natural gas required in the blends are assessed, along with the decarbonization achieved. Then, a detailed application to the tile sector in Spain is conducted, focusing on the use of a 50/50 HyBECCS/natural gas blend, compared to the biomethane/natural gas blend required to achieve the same level of decarbonization. The required biomethane is evaluated and compared to the lower-cost feedstocks (landfill and OFMSW). The required production of hydrogen is also calculated, and contextualized with the size of existing SMR plants in Spain. Finally, a detailed cost analysis is performed, highlighting the contributions of different factors and identifying natural gas price as a key factor. This leads to the inclusion of a sensitivity analysis regarding natural gas prices for the two biogas feedstocks considered.

Table 3
Costs associated with the stages of biomethane production and final OPEX. Energy is based on higher heating value of methane.

	Biogas [€/MWh]	Upgrading [€/MWh]	Injection [€/MWh]	Biomethane [€/MWh]	$OPEX_{bm}$ [€/kg H_2]
Landfill	7.21	11.96	5.68	24.85	1.340
OFMSW	31.55	11.96	5.68	49.19	2.653

Table 4
Variables of the model and their corresponding assumptions.

Variable	Value	Unit	Reference
DP	238,356	kg H_2/day	Own calculations
N	25	Years	[32]
$OPEX_{bm}$ (biogas)	8 - 35	€/MWh-LHV	[60]
$OPEX_{bm}$ (injection)	5.68	€/MWh-HHV	[32]
$OPEX_{bm}$ (upgrading)	11.96	€/MWh-HHV	[32]
$OPEX_{CO_2}$ (carbon tax)	80	€/t CO_2	[30]
$OPEX_{CO_2}$ (injection)	11	€/t CO_2	[42]
$OPEX_{CO_2}$ (transport)	6.90	€/t CO_2	[42]
OPEXom	0.148	€/kg H_2	[24]
r_{CO_2}	8	%	[32]
r_x (other than carbon tax)	0		[32]
Wacc	8	%	[32]

3.1. Resources and decarbonization potential

Fig. 4 depicts the relative CO_2 emissions (blue line) of a blend of HyBECCS with natural gas, with x representing the volume concentration of HyBECCS. It is observed that HyBECCS concentrations exceeding 71.8 % result in negative emissions, which can offset additional emissions in the factory. The red line represents the required volume concentration (y) of biomethane in a blend with natural gas to achieve emissions equivalent to those of the blend of HyBECCS with natural gas. This plot is provided for HyBECCS concentrations lower than 71.8 % because negative emissions cannot be achieved with biomethane alone. For example, as it is shown, a 50/50 blend of HyBECCS with natural gas yields emissions of 92 g CO_2/kWh-LHV, identical to a blend consisting of 53.6 % biomethane and 46.4 % natural gas. The emissions from HyBECCS are −8.64 kg CO_2/kg H_2 (−771.43 g CO_2/Nm^3 H_2), while the emissions from natural gas (assumed to be pure CH_4) are 1964.29 g CO_2/Nm^3 CH_4. Thus, in a 50 % blend, the net emissions are 596.43 g CO_2/Nm^3 of the blend. Taking into account the lower heating values of hydrogen and methane (3 kWh/Nm^3 and 9.952 kWh/Nm^3, respectively), the lower heating value of the blend is 6.48 kWh/Nm^3 and the emissions amount to 92 g CO_2/kWh-LHV. These emissions represent a 53.3 % reduction compared to pure fossil methane (197.38 g CO_2/kWh-LHV). Since biogenic CO_2 from biomethane is considered neutral, the natural gas in a biomethane/natural gas blend has to be diluted to 46.6 % (92/197.38) to achieve the same specific emissions level.

Fig. 5 illustrates the required amounts of biomethane and natural gas (MWh-HHV) per unit of heat released (MWh-LHV) for each type of blend as a function of the concentration of HyBECCS. The higher heating value (HHV) is used to represent the overall energy content of the resources, while the heat released is expressed as the lower heating value (LHV) because the flue gases temperature is typically higher than the dew point. The solid lines demonstrate the reduction in biomethane usage due to the negative emissions in blends with HyBECCS. As hydrogen exhibits a lower volume heating value than methane (approximately 1/3), HyBECCS blends require more natural gas, as indicated by the dashed lines.

In 1 Nm^3 of a 50 % of HyBECCS/natural gas blend (6.48 kWh-LHV/Nm^3), there are 44.64 g H_2, meaning that 0.04464/18.54 MWh-HHV of biomethane is required. This results in 0.373 MWh-HHV of biomethane per 1 MWh-LHV of heat released in the blend. Since a biomethane/natural gas blend requires 53.4 % biomethane to reach the same specific emissions (92 g CO_2/kWh-LHV), the biomethane requirement is 0.534 × 11.04 kWh-HHV/Nm^3 of blend, that is, 0.534 × 11.04/9.952 MWh-HHV of biomethane/MWh-LHV of heat released by the blend. Thus, a 53.4/46.6 biomethane/natural gas blend requires 0.5924 MWh-HHV of biomethane per MWh-LHV of heat released, exceeding the demand by 58.8 % in a 50/ 50 blend of HyBECCS/natural gas. Therefore, decarbonization through HyBECCS results in a 37.2 % reduction in biomethane usage compared to biomethane/natural gas blends. In this sense, Lou et al. [26] compared biohydrogen production from biomass gasification and from biomethane via SMR, claiming that the latter should be

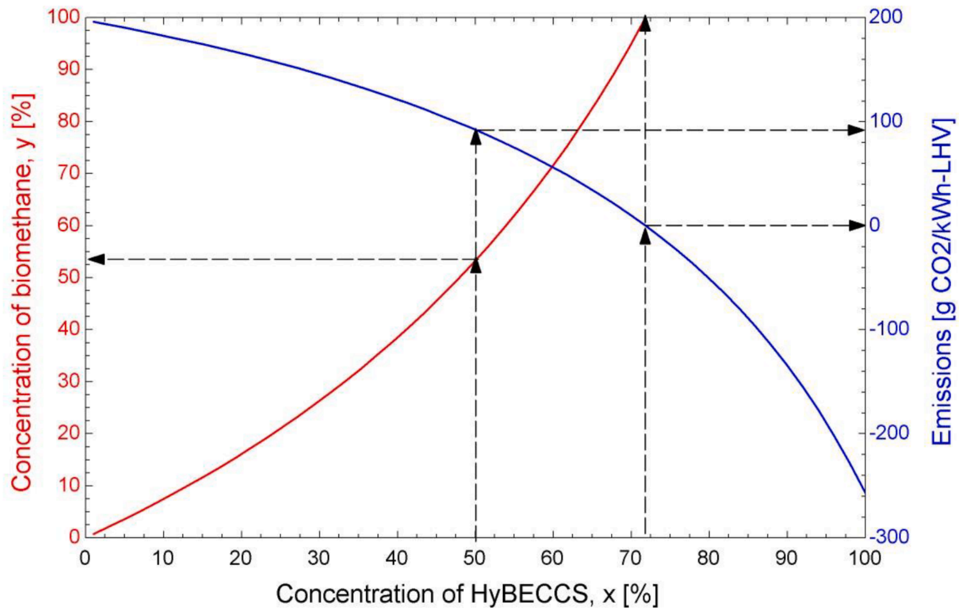


Fig. 4. Required concentration of biomethane (y, red line in vertical left axis) in a biomethane/natural gas blend to achieve the same level of relative emissions (blue line in vertical right axis) than a HyBECCS/natural gas blend of a given concentration (x, horizontal axis) of HyBECCS.

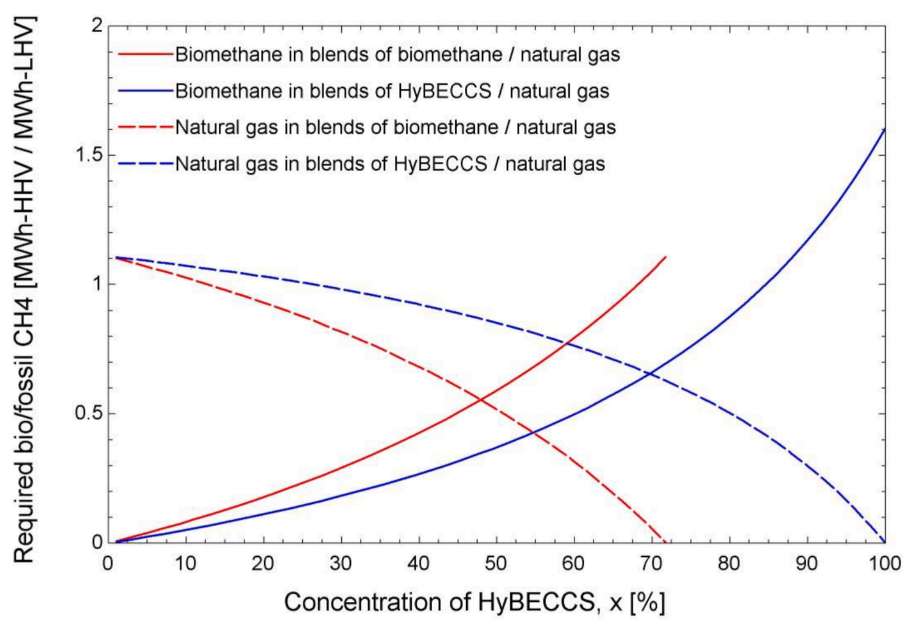


Fig. 5. Use of biomethane and natural gas in blends with the same level of decarbonization.

prioritized for substituting natural gas rather than producing biohydrogen. The reduction in biomethane usage, however, allows for biohydrogen production via SMR+CCS to be a more efficient utilization of biomethane, provided that hydrogen combustion conditions are met. This will be further discussed in the context of the tile sector.

3.2. Application to the tile sector in Spain

Fig. 6 is obtained scaling the curves from Fig. 5 to cover the entire demand of the tile sector (14 TWh), whereas Fig. 7 depicts the required hydrogen production and the resulting emissions. It can be seen that a 50/50 HyBECCS/natural gas blend requires 4.693 TWh-HHV of biomethane and 10.76 TWh-HHV of natural gas, emitting 1.162 Mt CO_2. This blend results in a 53.4 % reduction in CO_2 emissions and a 23.1 % reduction in natural gas usage. In comparison, a 53.4/46.6 biomethane/natural gas blend requires 7.467 TWh-HHV of biomethane (2.774 TWh-HHV more than using HyBECCS) and 6.533 TWh-HHV of natural gas (4.227 TWh-HHV less than using HyBECCS) to achieve the same CO_2 emissions. Given the current limited production of biomethane, the advantage of using HyBECCS in blends instead of direct biomethane is clear. The biomethane required for the HyBECCS production results in 87 kt H_2, which is in accordance with the size of existing SMR plants in Spain, ranging from 8 kt to 147 kt of annual capacity [22].

Figs. 8 and 9 provide the breakdown of the energy balance and emissions. Figs. 8a and 8b reveal that even when considering losses in the SMR with CCS process (1.31 TWh-LHV) the consumption of biomethane is lower with HyBECCS (4.23 TWh-LHV) compared to direct biomethane use (6.731 TWh-LHV). Regarding emissions, Figs. 9a and 9b demonstrate that the negative emissions from HyBECCS (−0.75 Mt CO_2) offset the additional emissions from natural gas (1.91 Mt CO_2 using

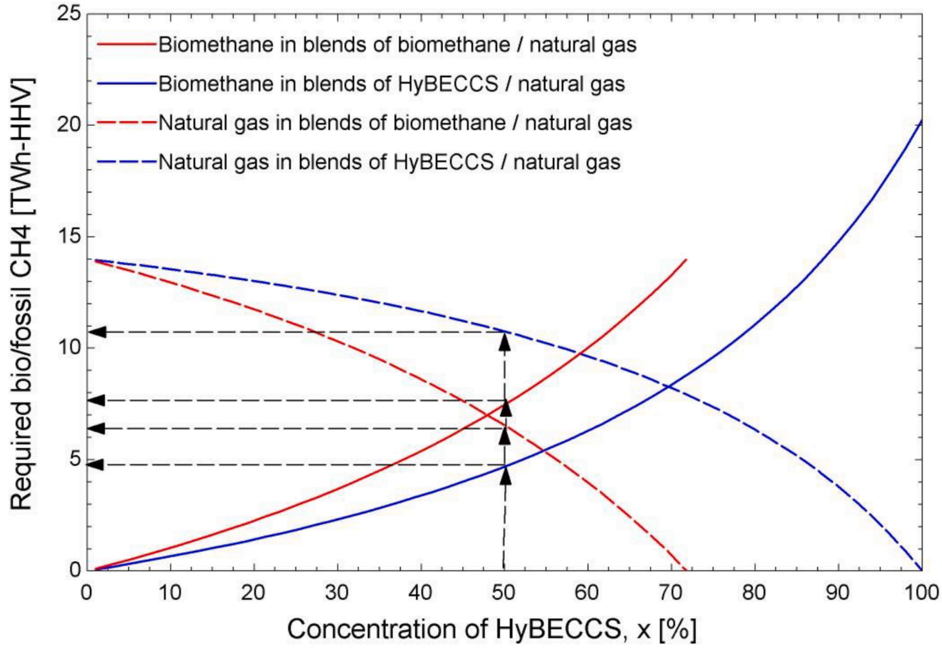


Fig. 6. Required biomethane and natural gas in blends with the same level of decarbonization to cover the entire thermal demand of the tile sector in Spain (14 TWh-HHV).

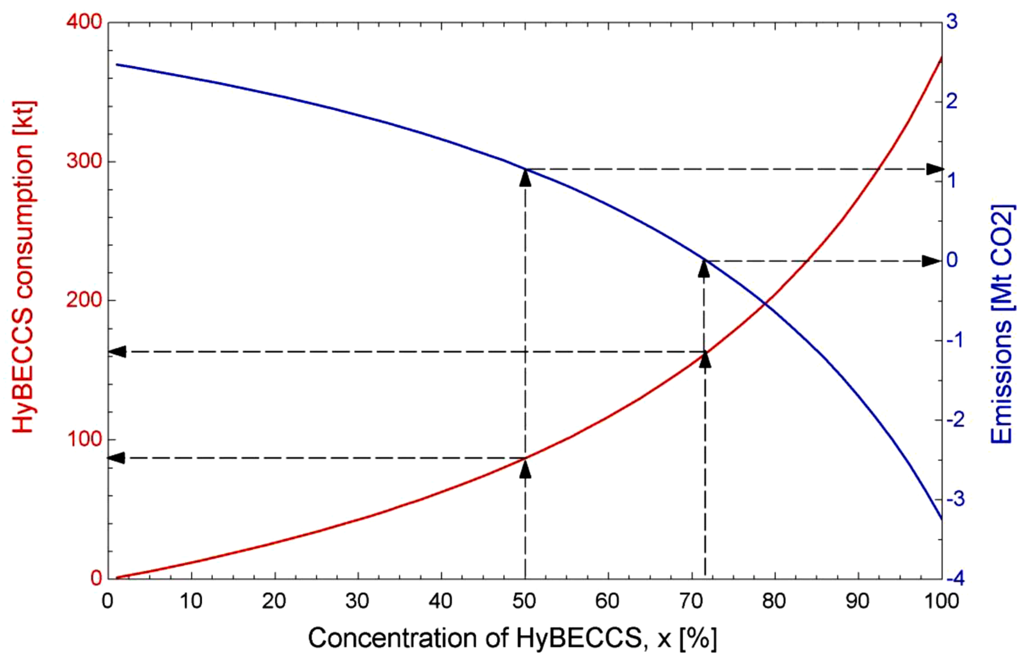
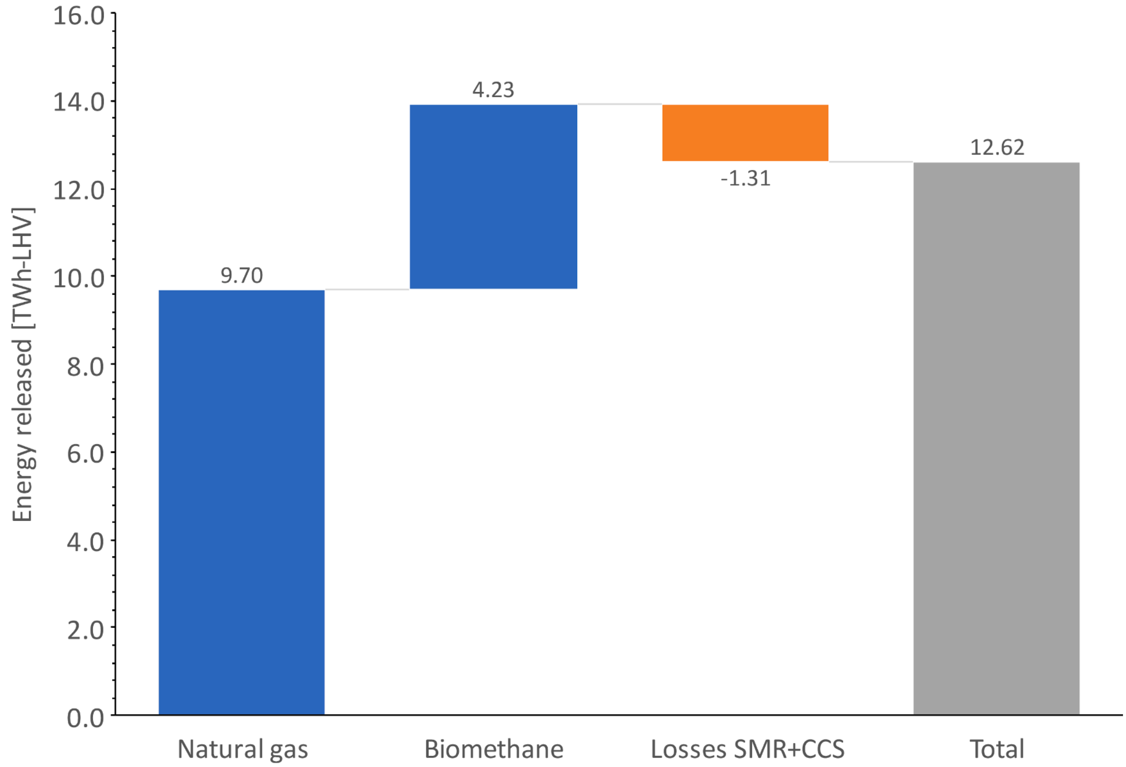


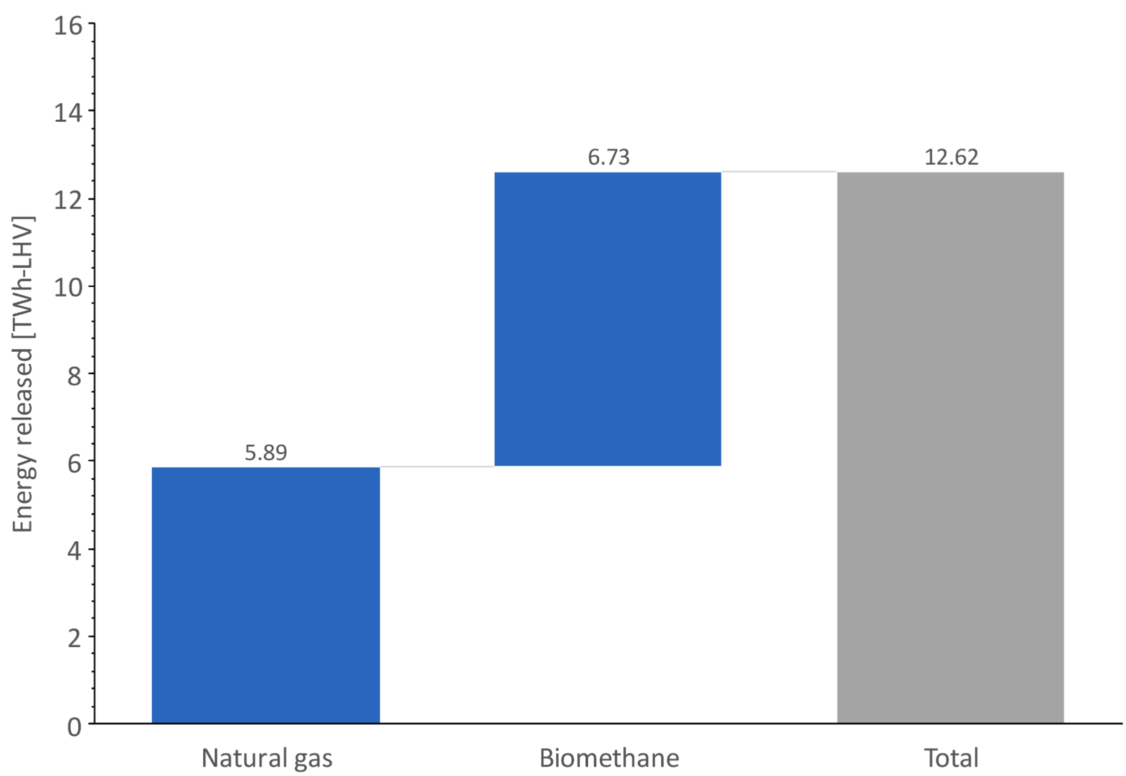
Fig. 7. Required HyBECCS production (red line in left axis) and CO_2 emissions generating (blue line in left axis) with blends of HyBECCS (x concentration) to cover the entire thermal demand of the tile sector in Spain (14 TWh-HHV).

HyBECCS compared to 1.16 Mt CO_2 using direct biomethane). These results reveal that it is more efficient to use biomethane as feedstock to produce HyBECCS rather than using it directly in combustion, as Lou et al. [26] proposed, especially given the limited biomethane production. The explanation for this apparently contradictory result lies in the negative emissions from the SMR+CCS process, which outweigh the efficiency losses in the process.

The consumption of gas in the tile sector is mainly divided between fuel for furnaces (58 %) and fuel for gas turbine operating in cogeneration (38 %) [62]. In relation to the furnaces, the Technological Ceramic Institute (ITC) in Spain conducted tests on blending hydrogen with natural gas in the Hidroker project, achieving positive results with blends containing up to 20 % hydrogen (in volume) [63] using conventional burners. The objective of this project is to maximize the share of hydrogen in the blend, aiming for 100 % utilization. Similarly, the Hydeploy project in the UK is conducting comparable tests, finding that concentrations higher than 75 % require new burners design due to changes in combustion patterns [64]. As previously mentioned, a significant portion of the natural gas in the tile sector is consumed by cogeneration gas turbines, making it important to explore whether hydrogen can be burned in these turbines. In this context, the potential use of hydrogen in gas turbines has attracted interest from both



(a)



(b)

Fig. 8. Energy balance breakdown of a 50/50 HyBECCS/natural gas blend (a) and 53.4/46.6 biomethane/natural gas blend (b) to meet the entire thermal demand of the tile sector in Spain (14 TWh-HHV of natural gas).

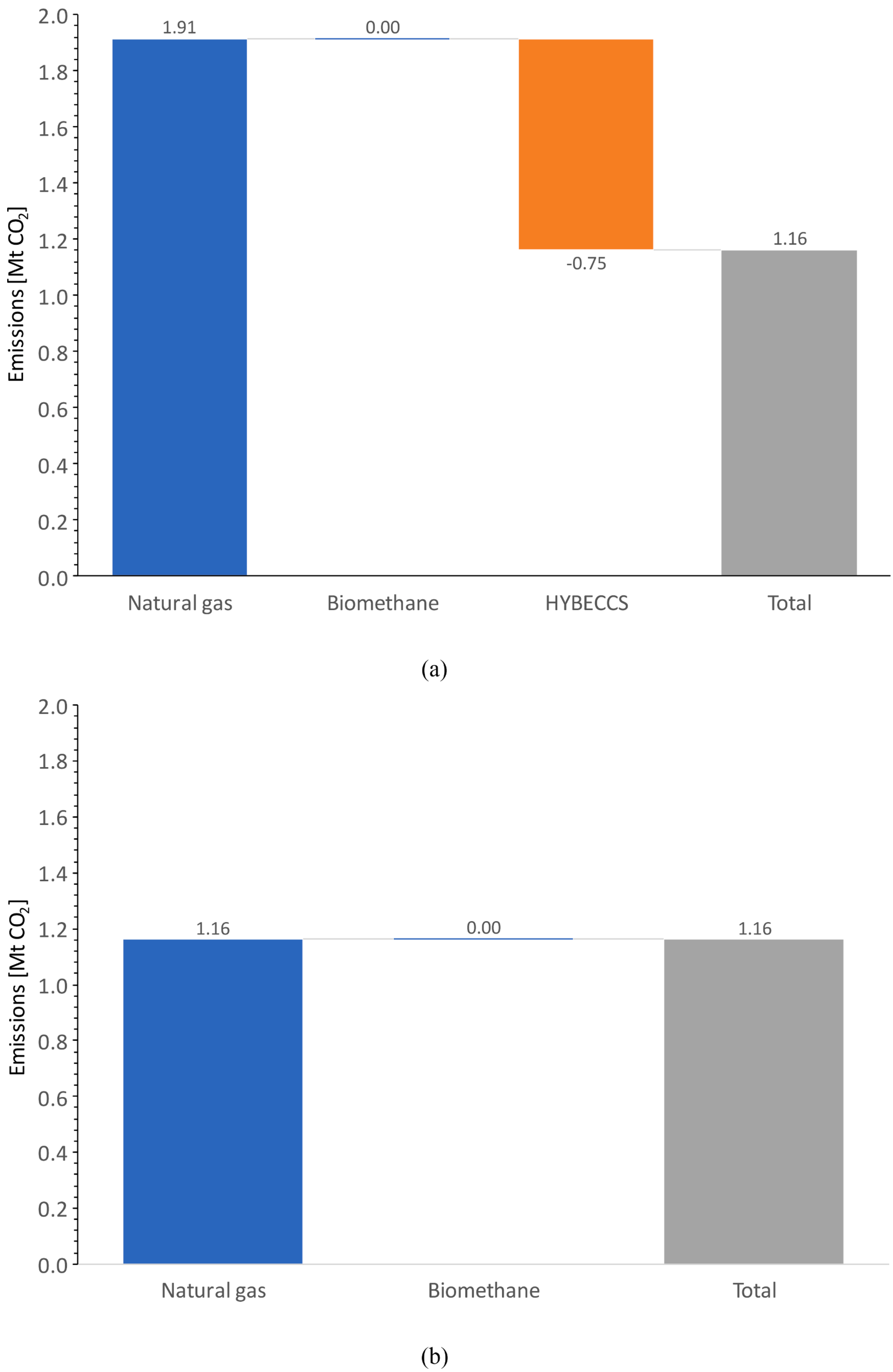


Fig. 9. Emissions breakdown of a 50/50 HyBECCS/natural gas blend (a) and a 53.4/46.6 biomethane/natural gas blend (b) to meet the entire thermal demand of the tile sector in Spain (14 TWh-HHV of natural gas).

researchers and turbine manufacturers. Pashchenko [65] analyzed the use of hydrogen/natural gas blends and steam methane reforming products (without water-gas shift), focusing on emissions reduction through gas turbine performance simulation. Ali et al. [66] reviewed a hydrogen use in gas turbines, especially with intercooling, and examined operational parameters and the impact of hydrogen share in the fuel. Mitsubishi reported that current gas turbines can operate with 20 % hydrogen blends, with challenges existing to reach 30 %. and they are converting three turbines to 100 % hydrogen at Magnum Power Plant in the Netherlands [67]. General Electric claims experience operating gas turbines fueled by hydrogen since the mid-1990s, with more than 100 gas turbines accumulating over 8 million hours of operation. They have current models with specific capabilities for burning hydrogen, some up to 100 %. Existing turbines can also be retrofitted [68].

Table 5 displays the performance of biohydrogen and natural gas blends in meeting the entire thermal demand of the tile sector in Spain (14 TWh-HHV) at two key hydrogen concentrations: 20 %, a currently achieved value, and 50 %, which is technically feasible in the short term. Table 6 provides the same information for blends of biomethane/natural gas with equivalent CO_2 emissions. It is observed that in both cases, the required biomethane can be obtained using the potential of only one of the proposed substrates (8.81 TWh from landfill or 7.92 TWh from OFMSW). Even with the near-term biomethane production (1.5 TWh) a blend with 20 % biohydrogen might be employed, although this would require all substrates, not just OFMSW or landfills. Regarding emissions reduction, a blend containing 50 % HyBECCS achieves a reduction of 53.4 %, whereas only 16.1 % reduction is achieved with 20 % HyBECCS. Considering this, a blend of 50 % HyBECCS and natural gas is considered, assuming its short-term technical viability in tile furnaces and gas turbines while limiting biomethane consumption.

Although new facilities are required, existing ones might provide a starting point for developing a scale demonstration. The SMR facility at the Castellón refinery has a hydrogen production capacity of 81.9 kt/year and a current production of 65.5 kt/year [22]. Thus, 16 kt of hydrogen could be available for the tile sector. This production would require 863 GWh of biomethane to be supplied to the SMR and would meet 2.58 TWh of the sector's demand (18.4 %). Additional infrastructure for carbon capture, as well as transport and storage management, would be needed, along with the logistics for transporting hydrogen to the tile factories.

Fig. 10 illustrates the cost of a 50/50 blend of HyBECCS /natural gas, based on a natural gas price of 25 €/MWh-HHV (excluding CO_2 tax). This gas price is indicative of the market situation in 2019, before the Ukraine war and without the effects of the COVID19 pandemic. For reference, the cost of natural gas including CO_2 tax is 64.71 €/MWh-LHV. The analysis considers two biomethane substrates: landfill gas (a) and OFMSW (b). In both scenarios, it is evident that the cost of the blend is primarily determined by the cost of natural gas including CO_2 tax (49.72 €/MWh-LHV). Natural gas accounts for 94.89 % of the blend cost when biogas comes from landfill and 80.91 % when it comes from OFMSW. On the other hand, the CAPEX and maintenance costs of the SMR+CCS plant are very low (3.51 €/MWh-LHV). The carbon credits (a revenue) fully offset the cost of biomethane when it is derived from landfill gas, while more than half is offset when OFMSW is used as a substrate. In both scenarios, the blend cost (52.40 €/MWh-LHV if landfill biogas is used or 61.45 €/MWh-LHV if biogas comes from OFMSW) is lower than the natural gas cost (64.71 €/MWh-LHV). The EBA assigns up to −26.5 kg CO_2/kg H_2 to biohydrogen from SMR+CCS accounting for the methane emissions avoided if manure is used as substrate to produce biomethane [69]. That level of negative emissions could result in carbon credits valued at up to 30.86 €/MWh-LHV for the blend, reducing the cost of biomethane to 36.57 €/MWh-LHV (compared to 70 €/MWh-LHV for biogas). While this suggests that manure could be a viable substrate, managing the captured CO_2 from the SMR facility is not sufficient in its own; certified sustainability calculations (proofs of sustainability) for methane emissions avoided are also needed. However, as such certificates are not yet part of Spanish regulation [58], the implementation of the HyBECCS process might be delayed. Therefore, the current proposal is considered more robust because of the cost advantages and regulatory frameworks for landfills and OFMSW.

Table 5
Absolute performance in HyBECCS/natural gas blends to meet a 14 TWh-HHV demand.

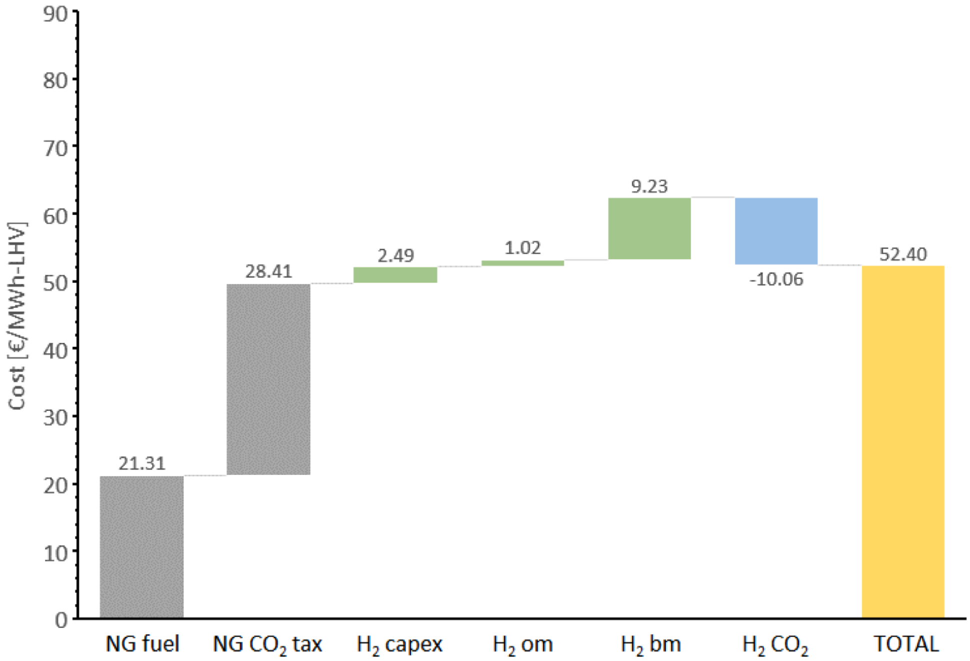
Concentration of HyBECCS [%]	Biomethane to produce HyBECCS [TWh]	Natural gas [TWh]	CO_2 emitted [Mt]
20 %	1.420	13.02	2.089
50 %	4.693	10.76	1.162

Table 6
Absolute performance in biomethane/natural gas blends to meet a 14 TWh-HHV demand with the same emissions as the HyBECCS/natural gas blends specified in Table 4.

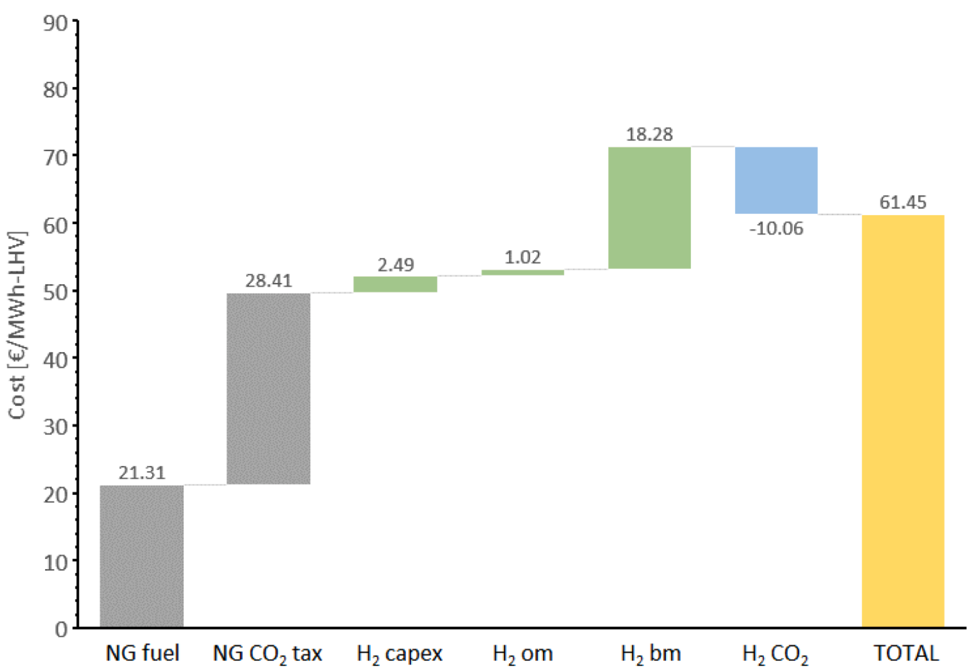
Concentration of biomethane [%]	Biomethane in blend [TWh]	Natural gas [TWh]	CO_2 emitted [Mt]
16.14 %	2.259	11.74	2.089
53.4 %	7.467	6.533	1.162

Fig. 11 shows information analogous to Fig. 10, but using a natural gas price (excluding CO_2 tax) of 40 €/MWh-HHV, in line with current gas prices [70]. Again, natural gas exhibits the largest share in the final cost (62.51 €/MWh-LHV), due to the strong offset of the biomethane cost by carbon credits (a revenue). As a reference, the cost of 100 % natural gas, including CO_2 taxes, is 81.35 €/MWhLHV, higher than both blends. In this case, the share of natural gas costs ranges from 95.89 % when using landfill biogas to 84.2 % when using biogas from OFMSW. These results demonstrate that the cost of the proposed blends is dominated by natural gas, whether landfill or even OFMSW are used as the biogas substrate. The CO_2 tax plays a key role in this result, as it increases the base cost of natural gas while reducing the cost of hydrogen. In the absence of this tax, the natural gas cost previous to Ukraine crisis was similar to that of hydrogen from OFMSW (21.31 €/MWh-LHV versus 21.79 €/MWh-LHV), exceeding by 2.68 times the cost of hydrogen from landfill gas at current natural gas prices (34.1 €/MWh-LHV versus 12.74 €/MWh-LHV).

In order to assess the competitiveness of the HyBECCS blends respect to biomethane and pure natural gas, a sensitivity study has been carried out. Fig. 12 illustrates the results of obtaining HyBECCS from the organic fraction of municipal solid waste, while Fig. 13 focuses on the same process but specifically from landfill sources. Figs. 12 and 13 analyze the sensitivity of the levelized cost of the proposed solution (50 % of HyBECCS /50 % of natural gas) to changes in natural gas prices compared to the blend of biomethane/natural gas with the required concentration (53.4 % biomethane) to achieve the same decarbonization level as the HyBECCS/natural gas blend. Current natural gas prices (including CO_2 tax) are included for comparison. As expected, the costs of the blend are lower when using landfill as substrate, with both blends exhibiting the same slope (0.8524 for the blend with hydrogen and 0.5547 for the blend with biomethane). As the blend with hydrogen requires more natural gas, it is more sensitive to its price. When OFMSW is used, the breakeven point is approximately 16.5 €/MWh for the natural gas price, with the blends having lower costs than natural gas for prices higher than this and vice-versa. The biomethane/natural gas blend has lower costs than the HyBECCS/natural gas blend for natural gas prices higher than 16.5 €/MWh, with the hydrogen blend having lower costs if the price of natural gas is lower than this value. It is worth mentioning that the biomethane blend uses more resources than the HyBECCS blend, thereby preventing other users from employing biomethane instead of natural gas. If this avoidance saving (considering the lower prices of biomethane compared to natural gas with the CO_2 tax) for these users is added as a cost to the biomethane blend, the dashed red



(a)

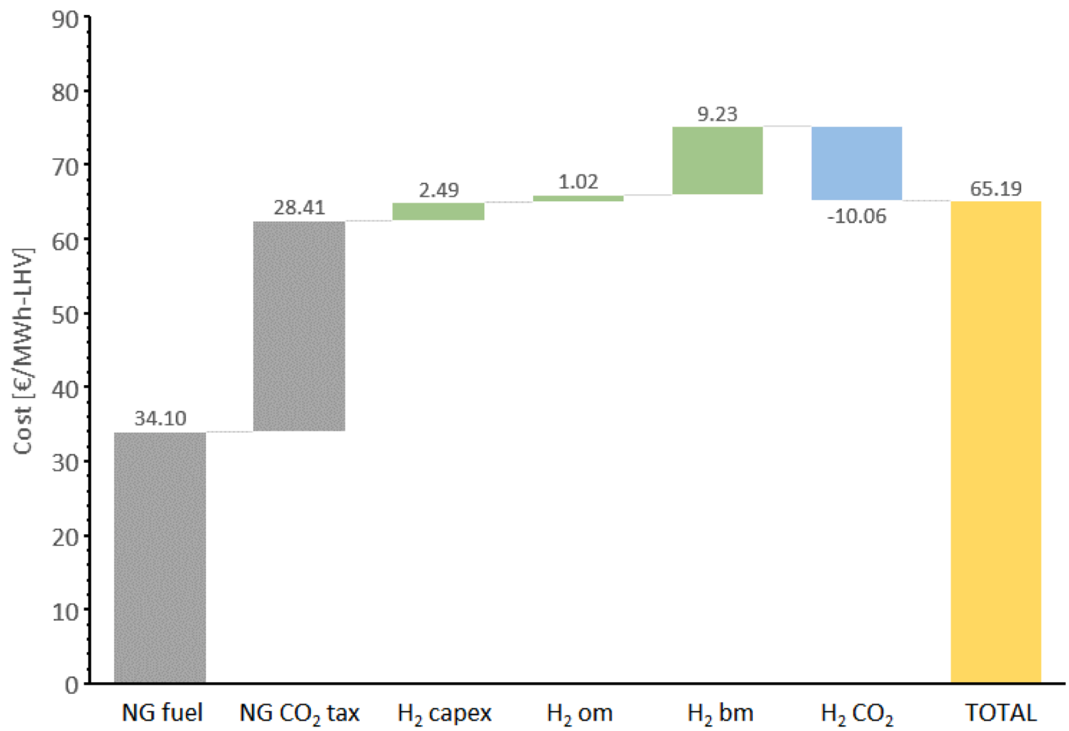


(b)

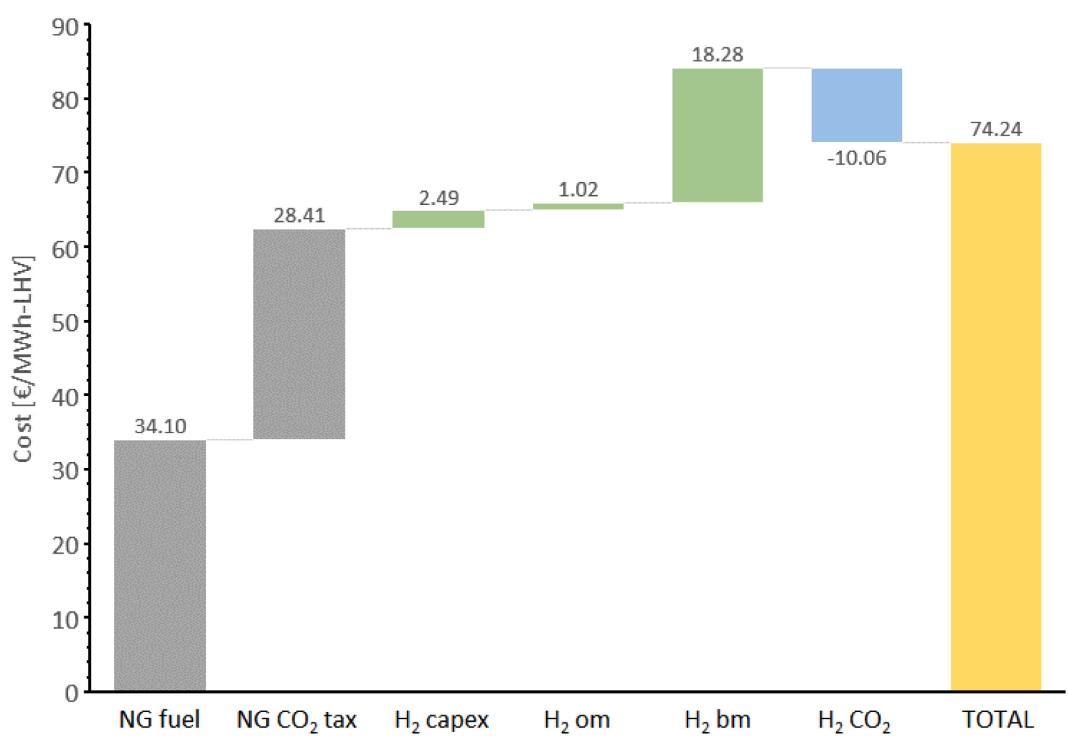
Fig. 10. Cost breakdown of 50/50 HyBECCS/natural gas blend to cover the thermal demand of the tile sector in Spain (14 TWh-HHV). Natural gas price (without CO_2 tax): 25 €/MWh-HHV. The biomethane required comes from landfills (a) or OFMSW (b).

line is obtained, which nearly aligns with the biohydrogen blend curve (blue line). The differences arise from the efficiency of the SMR+CCS capture process and the minimal impact of CAPEX and maintenance costs. The use of landfill as substrate produces breakeven points with lower natural gas prices, with the HyBECCS blend always costing less than natural gas within the analyzed price range. For natural gas prices higher than 5 €/MWh, the biomethane blend has lower costs than with hydrogen. Again, if the cost for the biomethane blend is adjusted for the extra biomethane consumption, similar costs to the HyBECCS blend are obtained.

Figs. 12 and 13 indicate that if landfill biogas is used as a substrate, the biohydrogen blend is always competitive against natural gas at the expected costs. Given that the biomethane demand is less than 54 % of its potential for this substrate, prioritizing its use in the tile sector in the



(a)



(b)

Fig. 11. Cost breakdown of 50/50 HyBECCS/natural gas blends to cover the thermal demand of the tile sector in Spain (14 TWh-HHV). Natural gas price (without CO_2 tax): 40 €/MWh-HHV. The biomethane required comes from landfills (a) or OFMSW (b).

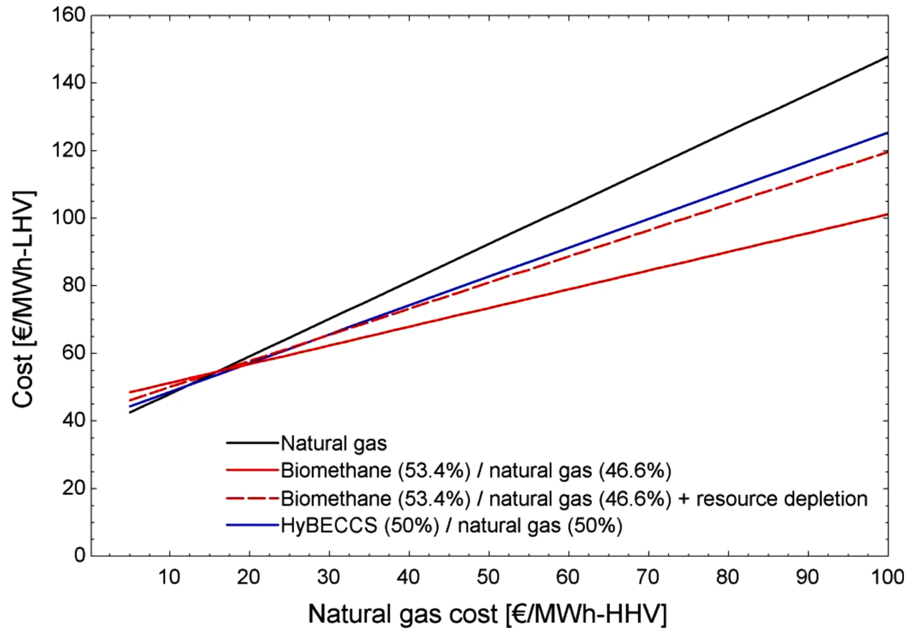


Fig. 12. Cost per thermal energy of different alternatives (both blends exhibit the same level of decarbonization) depending on natural gas cost. Biomethane is assumed to be produced from OFMSW.

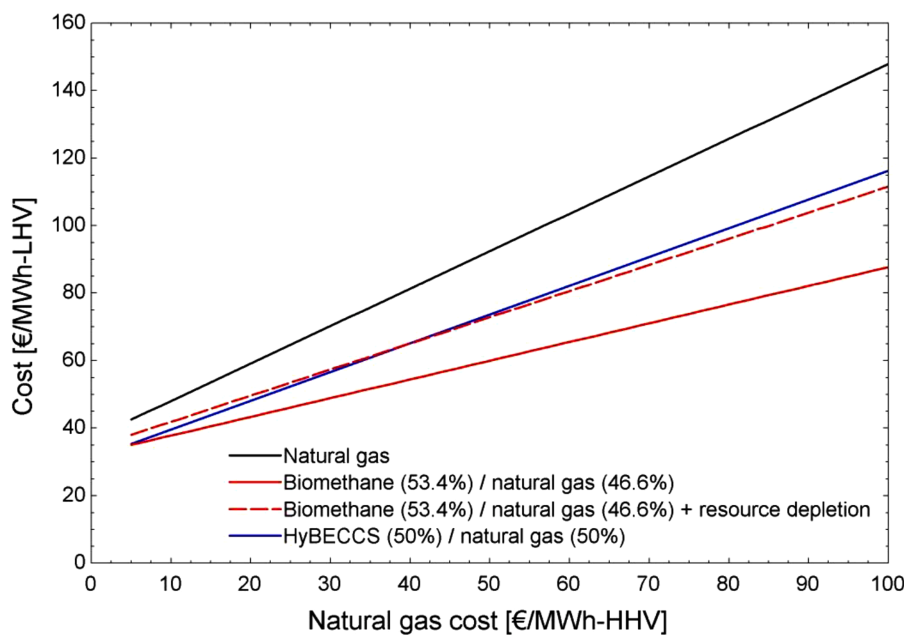


Fig. 13. Cost per thermal energy of different alternatives (both blends exhibit the same level of decarbonization) depending on natural gas cost. Biomethane is assumed to be produced from landfill.

near-term is recommended. Meanwhile, promoting biogas production in municipal solid waste treatment plants is essential to replace the finite resource from landfills. Using OFMSW, this solution is competitive for gas prices above 33 €/MWh-HHV, which is comparable to current natural gas prices [70]. While the biomethane blend is easier to implement and more cost-effective than the HyBECCS blend for expected natural gas prices, both options nearly reach cost parity when considering the lower yield of directly using biomethane.

4. Conclusions

Spain faces a significant challenge in decarbonizing its energy sector by 2030 and 2050, aligning with the country's ambitious climate targets. To achieve these goals, innovative solutions are crucial, especially in hard-to-abate sectors. Biohydrogen with negative emissions emerges as a promising candidate in this endeavor, offering a pathway towards substantial emissions reductions. This hydrogen could become a key player in the decarbonization of hard-to-abate sectors, thanks to the negative emissions generated when CO_2 is captured in Steam Methane Reforming (SMR) with CCS units using biomethane instead of natural gas.

In this research, blends of natural gas with both HyBECCS or biomethane have been evaluated for one of Spain's hard-to-abate sectors, namely the tile sector. The results demonstrate that HyBECCS blends allow for saving more than 37 % of biomethane to achieve the same emissions reduction. A 50 % blend of HyBECCS and natural gas would require 4.7 TWh-HHV of biomethane for the entire tile sector, which is only 53 % of the potential from landfill sources or 59 % of the potential from the organic fraction of municipal solid waste. This indicates that there is sufficient potential available.

Due to the lower heating value of hydrogen per volume unit compared to methane (approximately 1/3), the use of a 50/50 blend of HyBECCS compared to a 53.4/46.6 blend of biomethane incurs a penalty when natural gas prices exceed 16.5 €/MWh for biomethane sourced from organic fractions of municipal solid waste, or 5 €/MWh for biomethane from landfills (with an 80 €/t of CO_2 tax). Despite this penalty, such blends of HyBECCS remained competitive with pure natural gas at prices exceeding 16.5 €/MWh when using biomethane from organic fractions of municipal solid waste, or at any natural gas price when using biomethane from landfills. This results in over a 53.4 % reduction in CO_2 emissions and more than a 37 % saving of biomethane compared to a blend of biomethane/natural gas with the same CO_2 reduction. If the extra resource consumption of biomethane blends by other users is factored into their costs, near parity is achieved between these blends and biohydrogen blends, with both being competitive compared to natural gas at its expected price.

A key feature of the proposed biohydrogen approach is the negative emissions from carbon capture and storage, quantified at −8.64 kg CO_2/kg H_2. This value can increase to −26.5 kg CO_2/kg H_2 if manure is used as a biomethane substrate, though certifying such emissions might delay its application. Results show that accounting only for the CO_2 captured at the SMR facility is sufficient to achieve a significant reduction in emissions. The existence of potential CO_2 storage sites near the tile cluster enhances the feasibility of this proposal. However, regulatory adjustments are needed to recognize carbon credits for negative emissions.

The importance of these findings lies in the potential of biohydrogen with negative emissions to significantly reduce carbon emissions in the industrial sector, particularly in areas where traditional natural gas usage poses environmental challenges. By optimizing the blend of HyBECCS and biomethane, industries such as tile manufacturing can achieve substantial emissions reductions while maximizing the use of renewable and low-carbon energy sources. This not only aligns with sustainability goals but also enhances the economic viability of transitioning to cleaner energy alternatives.

The model presented in this work relies heavily on the successful implementation of CCS in existing SMR facilities. According to the IEA in [30], "there is significant potential to expand CCS retrofitting to reduce emissions". They also highlight that this CCS application is relatively low-cost and that many existing facilities are located in coastal industrial areas, creating opportunities to share CO_2 transportation and storage infrastructure with other industrial establishments.

The study has some limitations. First, there is a discrepancy between Spain's updated NIECP target for biomethane deployment (20 TWh by 2030) and sectoral estimates, which may hinder its full implementation. Second, while CO_2 transport networks are progressing in the EU, development in Spain remains stalled, particularly affecting the potential for large-scale CCUS projects. Third, social opposition to CO_2 injection into underground storage could further delay these initiatives. Finally, although negative emissions are recognized, regulatory challenges persist in monetizing them, limiting their integration into formal carbon markets. Future work should focus on addressing these regulatory and infrastructure challenges to enhance the viability of biohydrogen with CCUS in hard-to-abate sectors. Despite these limitations, the current study demonstrates the significant potential of HyBECCS to reduce emissions in Spain's tile industry, offering a promising pathway for decarbonization.

CRediT authorship contribution statement

Luis Yagüe: Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **José Ignacio Linares:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Eva Arenas:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization. **José Carlos Romero:** Writing – review & editing, Supervision, Methodology, Conceptualization.

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.

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Data availability

Data will be made available on request.

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