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Contribution of upcycling surplus hydrogen to design a sustainable supply chain: The case study of Northern Spain

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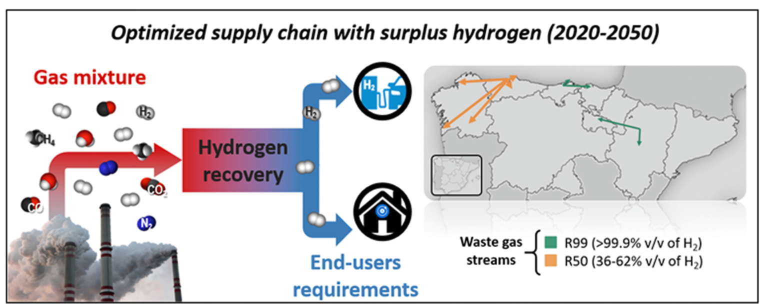
HIGHLIGHTS

- Upcycling of surplus hydrogen streams.
- Integration of recovered hydrogen into a hydrogen supply chain.
- Techno-economic feasibility of hydrogen upcycling using optimization-modelling.
- Surplus hydrogen has a pivotal role in initiating the shift to a hydrogen economy.

GRAPHICAL ABSTRACT



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ABSTRACT

To further advance a world powered by hydrogen, it is essential to take advantage of the environmental benefits of using surplus industrial hydrogen to energy conversion. In this paper, the integration of this renewable source in a hydrogen supply chain has been analysed with the following considerations, (1) the techno-economic modeling is applied over the 2020–2050 period, at a regional scale comprising the north of Spain, covering the main sources of surplus hydrogen in the region, (2) the supply chain feeds fuel cell devices powering stationary and mobile applications and, thereby stablishing the quality standards for the upcycled hydrogen and, (3) a mixed-integer programming model (MILP) is formulated to predict the optimal integration of surplus hydrogen. The advantages of this research are twofold, (i) on the one hand, it provides the methodology for the optimal use of surplus hydrogen gases promoting the shift to a Circular Economy and, (ii) on the other hand, it contributes to the penetration of renewable energies in the form of low cost fuel cell devices to power stationary and mobile applications. The results show that the combination of all the infrastructure elements into the mathematical formulation yields optimal solutions with a plan for the gradual infrastructure investments over time required for the transition towards a sustainable future energy mix that includes hydrogen. Thus, this work contributes to improving the environmental and economic sustainability of hydrogen supply chains of upcycling industrial surplus hydrogen.

1. Introduction

It has been reported that besides its prominent role in hydrogen-to-chemical processes, hydrogen-based energy storage systems could play in the future a key role as a bridge between intermittent electricity provided by alternative sources and the common fossil fuel-based

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Nomenclature

MILP	Mixed-Integer Linear Programming
GHG	Greenhouse Gas
PEM	Polymer Electrolyte Membrane
ISO	International Organization for Standardization
HFCV	Hydrogen Fuel Cell Vehicle
HSC	Hydrogen Supply Chains
SMR	Steam Methane Reforming
CCS	Carbon Capture and Storage
NPV	Net Present Value
COG	Coke Oven Gas
BOF	Basic Oxygen Furnace
BTX	Benzene, Toluene and Xylenes
INE	Spanish Statistical Office
PSA	Pressure Swing Adsorption
MEM	Membrane Technology
CH2	Gas Hydrogen
LH2	Liquid Hydrogen
JuMP	Julia for Mathematical Optimization
O&M	Operating and Maintenance

energy system. The versatility and unique properties of hydrogen open the way to accomplish this goal. Hydrogen is an odorless, tasteless and colorless gas that, despite its lower volumetric energy density (0.0108 MJ/L) compared to hydrocarbons, it has the largest energy content by weight (143 MJ/kg) [1–3].

Hydrogen can be obtained from a number of primary or secondary energy sources, depending on regional availability, such us natural gas, coal, wind, solar, biomass, nuclear, and electricity using electrolyzers [4]. Hydrogen production from carbon-lean and carbon-free energy sources could be the long-term aim of the hydrogen utopia [5].

The promotion of sustainable mobility has significantly increased demand for green hydrogen as an attractive alternative to non-renewable energy. In Spain, the transportation sector contributes 25% to the total greenhouse gases emissions, followed by the residential and commercial sectors contributing 15%. With regard to GHG diffuse emissions transportation accounts for 50% [6]. These figures clearly reveal the importance of a shift to a hydrogen economy in both sectors; within this goal, hydrogen technologies must overcome efficiency, cost, and safety challenges [7].

At the same time, hydrogen losses in industrial waste gas streams have been estimated to be 10 billion Nm^3 per year in Europe [8]. Despite this figure being largely based on statistical assumptions, and not on a site-by-site assessment, this surplus hydrogen volume is quite significant. This available “surplus hydrogen” is often recovered as fuel burnt for heat and power production, although cheaper energy sources could be used instead. Within a more sustainable framework, this surplus hydrogen could be recovered as feedstock for the manufacture of commodity chemicals such as ammonia or methanol, or even be used as fuel for both transportation and stationary applications [9].

Polymer Electrolyte Membrane (PEM) fuel cells are electrochemical devices that could be fed with hydrogen to generate clean energy where water and heat are products. In this case, the hydrogen fed must meet a quality standard that requires its purification from multicomponent gas mixtures as per end-users requirements [10–12]. In compliance with the International Standard ISO 14687, hydrogen gas should have a purity of at least 99.97% (minimum mole fraction) for road vehicle PEM fuel cells, and of at least 99.9% for stationary appliances. Furthermore, the maximum mole fraction of total non-hydrogen gases may not exceed 300 μmol/mol for automotive fuel cells and 0.1% for stationary fuel cells.

Industrial waste streams with hydrogen content higher than 50% are considered to be potential promising sources for hydrogen recovery through the use of separation techniques. It has been estimated that the price of recovered hydrogen could be 1.5–2 times lower than the price of hydrogen from natural gas reforming [13,14]. These figures highlight the potential and attractiveness of using these hydrogen-rich waste streams as source for hydrogen. However, the final price and opportunity of recovering wasted hydrogen streams is highly dependent on the implementation of cost-effective separation technologies, where membrane separation systems are well positioned [15].

Although in recent years, the prospects of a shift to a hydrogen economy have created great interest in the scientific community and social stakeholders, the success relies on the availability of the necessary infrastructures [16]. In the specific case of the mobility sector, the main obstacle hindering vehicles manufacturers and consumers from embracing hydrogen fuel cell vehicles (HFCVs) is mostly the lack of a hydrogen infrastructure [17]. A number of works focused on the use of decision-support tools for the design and operation of hydrogen supply chains (HSC), have been reported addressing questions such as the design of the hydrogen fuel infrastructure applied at the country, region and city levels with Almansoori and Shah leading the way [18]. Some studies include the selection of the production technology (primary and secondary energy sources) and hydrogen transport forms (pipeline, truck and on-site schemes) through each node of the supply chain [19]. Also, most of these studies analyze future hydrogen network in terms of capital and operating expenditure of the infrastructure focusing on the transportation sector [20–23]. However, Europe’s future plants expect an increased hydrogen demand in both road vehicle transportation and residential/commercial sectors [24]. Recent evidence suggests that steam methane reforming (SMR) with carbon capture and storage (CCS) is expected to be the most economically and environmentally attractive technology for producing hydrogen while renewable source infrastructures like wind and solar farms continue developing [25–27]. Other studies have been focused on the distribution network for hydrogen describing what is the optimal delivery form inside the chain [17,28,29]. The assessment of environmental, economic and risk aspects by using multi-objective optimization-based approaches has been also reported [16,20,30–37]. This approach is ideal for optimal decisions when two or more conflicting objectives exist. Furthermore, advanced research has been assessed on the environmental impacts of a broad variety in hydrogen production technologies by recent researchers [38–40]. In economic terms, the final decision will define the time when stakeholders shall make their investments in developing the hydrogen infrastructure regarding payback and profit. Finally, economies of scale need to be taken into account to compare the advantages of centralized versus distributed production, as well as the impact in the transportation costs. Interesting studies have been conducted establishing efficient investment strategies over a specific timeframe by using multi-period optimization models. Some optimization models have also considered demand uncertainty by using stochastic modeling approaches [41–45].

The latest studies have included the production of biohydrogen from solid waste streams such as biomass into the hydrogen network showing significant decreases in producing costs and CO_2 emissions [46,47]. Meanwhile, among the list of hydrogen waste gas streams, some studies have concentrated on the management, optimization, and utilization of steel-work off gases in integrated iron and steel plants [48–51]. However, little work has focused on the optimization of various by-product gases in the HSC. To the best of our knowledge, reported optimization models for HSCs do not consider the competitiveness of upcycling hydrogen-rich waste gas sources for its reuse in both transportation and residential sectors.

Hence, the novelty of this study is a methodology for analysing the techno-economic feasibility of a HSC with contribution of upcycled

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