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Optimization approach for the design of a sustainable hydrogen supply chain through steam methane reforming

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Hydrogen is emerging as a key energy carrier in the global transition to more sustainable energy systems. There are various technologies for its production, each with its own advantages and disadvantages in terms of efficiency, costs, and environmental impact. More than 97% of the hydrogen produced globally is mainly obtained through steam methane reforming (SMR) of natural gas, which is the most widely used and cheapest method of production. Although the conventional SMR process is energy-intensive and results in significant carbon dioxide emissions, there are opportunities to enhance its sustainability by integrating Carbon Capture, Utilization, and Storage (CCUS) technologies. However, a crucial factor for its large-scale deployment is the development of an efficient and sustainable hydrogen supply chain (HSC) design, where all processes along the chain are optimized while simultaneously satisfying environmental, economic, and social criteria. The present study proposes an optimization model for designing a HSC, focusing on the development of hydrogen refueling stations simultaneously considering all aspects of sustainability. It is formulated in terms of Mixed-Integer Linear Programming (MILP) and includes the necessary parameters, decision variables, and environmental, economic, and social performance of the HSC, along with an objective function and constraints. The model is designed to be proved on a real case study on the territory of the Republic of Bulgaria with its 27 administrative regions.

Keywords: hydrogen production; steam methane reforming; supply chain design; multi-objective optimization; economic, environmental and social criteria

INTRODUCTION

Hydrogen is emerging as a key energy carrier in the global transition to more sustainable energy systems [1]. It offers the potential for decarbonizing various sectors, including transportation, industry, and electricity generation, especially when produced from renewable sources [2].

In the context of growing concerns about climate change and the need to reduce greenhouse gas emissions, sustainable hydrogen production is becoming increasingly important [3].

Global hydrogen consumption is growing rapidly, reaching 95 Mt in 2022, and is expected to reach 180 Mt by 2030. The greenhouse gas emissions generated from the production of 95 Mt of hydrogen in 2022 are over 1,291 Mt CO_2-eq [4].

There are various hydrogen production technologies, each with its own advantages and disadvantages in terms of efficiency, costs, and environmental impact [2]. Today, roughly 95% of all hydrogen is produced from fossil fuels through the steam methane reforming (SMR) of natural gas [4].

Regardless of the production method used, an efficient and sustainable hydrogen supply chain design is crucial for its large-scale deployment [2]. A well-optimized supply chain should encompass all transportation, and distribution, while considering stages of hydrogen production, storage, all aspects of sustainability, including economic, environmental, and social factors.

The lack of developed infrastructure is often identified as a major barrier to the development of the hydrogen economy. In this context, the optimal design of a sustainable supply chain for hydrogen production via SMR represents an important research challenge.

The aim of this study is to develop a framework for modeling and optimizing a sustainable hydrogen supply chain (HSC) that enables informed decision-making regarding its structure and operation.

By including various storage and transportation options, this study aims to propose an optimization

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approach that ensures environmentally and economically efficient hydrogen production and delivery from methane, while achieving social satisfaction for the participants in the supply chain.

The proposed approach will contribute to overcoming the existing challenges and supporting the realization of hydrogen's full potential as a clean energy carrier.

LITERATURE REVIEW

In recent years, the concept of using hydrogen as a potential clean energy carrier has gained significant interest due to the need to achieve energy sustainability and flexibility in production systems [5]. In addition to its role in decarbonizing the transportation and residential sectors, hydrogen enables the efficient production of so-called e-fuels (electrofuels), such as ammonia, methanol, and synthesis gas, when combined with CCUS technology and renewable sources [6].

However, despite the efforts of numerous researchers in developing methods for hydrogen production from renewable sources, the production of "green" hydrogen remains significantly more expensive compared to technologies that use fossil fuels [7].

Among the various production methods, the production of so-called "grey" hydrogen from natural gas through SMR is the most widespread and well-established technology for hydrogen extraction. Global natural gas consumption for this purpose accounts for 6% [8].

The successful implementation of this technology also faces several challenges, with the biggest being the lack of suitable infrastructure.

One way to achieve high efficiency and sustainability in this type of production, as well as to address the aforementioned issues arising during their implementation, is the application of an approach for the optimal design of the hydrogen supply chain, while simultaneously considering environmental, economic, social, and other aspects. The optimal hydrogen supply chain must consider the number, location, and capacity of both production facilities and storage capacities, as well as transportation methods. Additionally, demand, temporal, and spatial factors should be taken into account from the early stages of HSC design.

Mathematical programming is a broadly used approach for optimal design and exploitation of hydrogen supply chains.

A large number of studies propose single-objective and multi-objective optimization models for the design and operation of regional HSC under deterministic and stochastic conditions, formulated in terms of MILP programming.

Erdoğan & Güler [9] propose a multi-objective optimization approach to simultaneously minimize costs, carbon emissions, and risk in the operation of an HSC for hydrogen production through SMR, combined with water electrolysis. Li *et al.* [3] develop a multi-period MILP model for designing a regional HSC, which takes into account the availability of primary energy sources (e.g., natural gas, coal, biomass, and renewable energy), production technologies, transportation methods, and types of storage.

Although economic and environmental factors are the most common objectives in the design and planning of HSC infrastructure, recent studies increasingly consider factors related to risk assessment in the operation of HSCs. Robles *et al.* [10] propose a stochastic, spatially-based, multi-objective, multi-period MILP model to minimize daily costs, global warming potential, and risk when determining the optimal number, type, capacity, and location of production and storage facilities, as well as transportation units, in addition to the hydrogen flow between different locations.

On the other hand, social responsibility is one of the key factors for assessing the willingness of end-users to switch to hydrogen fuel cell vehicles.

An example of such a study is the one by Fazli-Khalaf *et al.* [11], who, in developing their HSC models, include reliability and social responsibility alongside environmental and economic criteria.

Their study focuses on people and their quality of life, taking into account social responsibility, such as job creation and timely satisfaction of customer needs. The authors develop a fuzzy probabilistic flexible programming model to enhance the flexibility of the hydrogen network under mixed uncertainties and to maximize the reliability and sustainability of the hydrogen supply chain in Iran.

Based on the literature review of recent years on approaches for the optimal design of hydrogen supply chains produced through steam methane reforming, it can be concluded that the majority of studies are dedicated to the design and planning of internal hydrogen supply chain networks for the transportation sector. The developed models are either deterministic or stochastic and are defined in terms of MILP. Approaches that consider only the economic performance of the examined HSC are widespread. Another, smaller group of approaches, alongside economic aspects, simultaneously considers environmental factors related to the assessment of greenhouse gas emissions released into the atmosphere as a result of hydrogen production via SMR, as well as the transportation of raw materials and products. Some authors have also included additional optimization criteria in their studies, related to the assessment of the risk of the technologies used, achieving the highest quality of the produced product, and others. There are few optimization approaches in the available literature for designing a sustainable HSC through the application of steam methane reforming (SMR) that, alongside economic and environmental aspects, also consider social factors.

This gives us the basis to propose a multi-objective optimization framework for designing a sustainable hydrogen supply chain that simultaneously considers the economic, environmental, and social performance models of the chain, with a focus on building infrastructure for hydrogen fuel stations.

HYDROGEN PRODUCTION THROUGH STEAM REFORMING OF METHANE

Steam methane reforming (SMR), commonly known as steam reforming, is a standard industrial method for producing hydrogen. More than 97% of the hydrogen produced globally is primarily obtained through SMR [12]. SMR is mainly applied in the chemical and oil industries, and currently, it is the most widespread and cost-effective method for hydrogen production.

The production of hydrogen through SMR undergoes the following main stages: (a) synthesis gas production, (b) conversion of carbon monoxide to hydrogen (water-gas shift reaction), and (c) purification [13] (Figure 1).

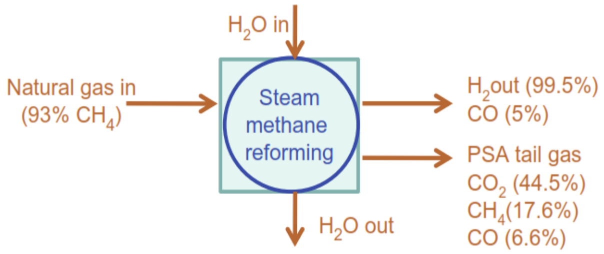

Figure 1. Block diagram of the steam methane reforming process [14]. In the first stage of the process, a catalytic endothermic reaction (Eq. 1) takes place between methane (natural gas) and steam at a high temperature (the steam methane reforming process). As a result of this reaction, synthesis gas is produced, which contains 85% of carbon monoxide, hydrogen, and small amounts of residual methane and carbon dioxide and water [15].

$$CH_4 + H_2O \xrightarrow[15-20bar]{800°C} CO + 3H_2,$$

$$\Delta H = +206KJ \quad (1)$$

$$CO + H_2O \xrightarrow[15-18bar]{400°C} CO_2 + H_2, \Delta H = -42KJ \quad (2)$$

The steam methane reforming process is conducted at temperatures ranging from 700 to 1000°C and pressures of 15-20 bar in the presence of steam, during which hydrocarbons are converted [16]. After the exothermic water-gas shift reaction (Eq. 2), carbon monoxide is converted into carbon dioxide and hydrogen. The process takes place at a temperature of 400°C and a pressure of 15-18 bar.

The governing reaction of the steam methane reforming process is as follows:

$$CH_4 + H_2O \rightarrow 4H_2(g) + CO_2, \Delta H = 164KJ \quad (3)$$

In the purification stage, the pressure swing adsorption process is the predominant method [12], where the reactive gas mixture containing methane and hot steam is fed into the catalytic reactor. Since the reaction is endothermic, the combustion of methane with air in the reactor's furnace provides the required reaction heat. The energy efficiency of these processes is 64% [17].

OPTIMIZATION APPROACH FOR DESIGNING A SUSTAINABLE SUPPLY CHAIN FOR HYDROGEN PRODUCTION OBTAINED THROUGH THE STEAM METHANE REFORMING PROCESS

General formulation of the optimization problem

Steam reforming is typically applied to natural gas, but it can also be successfully adapted for processing other hydrocarbons such as propane, gasoline, or ethanol, providing flexibility in raw material selection.

The availability and economic viability of natural gas determine the competitiveness of steam reforming for hydrogen production. The hydrogen produced by this method is available at an affordable price of 1,2-1,5$\$/kg_{-H_2}$.

It is evident, however, that carbon dioxide is an inevitable by-product. The primary pollution with carbon dioxide comes from the energy used to heat the reaction mixtures, which amounts to approximately 10 kg CO_2 of kg H_2. This contributes to generation of greenhouse gas emissions, making the method less environmentally friendly. Carbon dioxide is a valuable raw material for a number of industries, but the process of its utilization increases the cost of H_2 to 1,5–1,8$\$/kg_{-H_2}$ [18].

Before hydrogen fuel can be successfully introduced to the market, however, there are several challenges to overcome, such as the high production cost and the lack of a developed infrastructure for hydrogen fuel transportation [19]. One way to overcome these challenges is by applying an approach for the optimal design of sustainable HSCs. Key aspects in designing such chains include the simultaneous consideration of the three aspects of sustainability – environmental, economic, and social (Figure 2) [20].

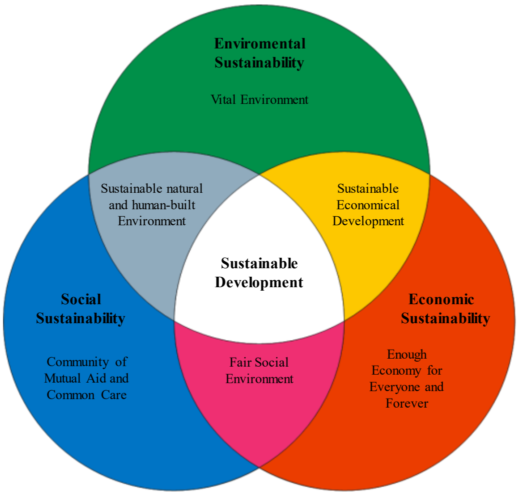

Figure 2. Concept of sustainable HSC management for hydrogen production and distribution [20].

This research aims to propose an optimization framework for designing a sustainable HSC, taking into account the three aspects of sustainability, with a focus on the construction of fueling stations. This issue is relevant as many countries are preparing to introduce hydrogen fuel for transportation purposes, as well as planning and building the necessary infrastructure for this. Initially, we consider the existing enterprises, such as chemical plants and fertilizer factories involved in hydrogen production from natural gas, as producers. The idea is that once the infrastructure for hydrogen fueling stations is built and hydrogen fuel end-users are formed, investors will then engage in the construction of facilities for producing carbon-neutral hydrogen.

Superstructure of sustainable HSC through SMR process

For the purposes of our study, we consider a superstructure of an HSC produced through SMR, which includes the following key elements:

1. Hydrogen production in chemical plants and fertilizer factories. The following processes are included:

- Steam methane reforming (SMR). This is the main process where methane reacts with steam at high temperatures (700–1000°C) using a nickel catalyst to produce synthesis gas (H_2, CO, CO_2).
- Carbon monoxide conversion (Water-Gas Shift) – an additional reaction to increase hydrogen yield by converting CO with steam into CO_2 and H_2.
- Hydrogen purification – through Pressure Swing Adsorption or membrane technologies for separating pure hydrogen.

2. *Hydrogen storage and transport*

- Short-term storage – using compressed hydrogen tanks.
- Distribution – transportation via cryogenic tankers.
- Local distribution stations – hydrogen refueling stations for industrial, transportation, and residential needs.

3. *Consumption and applications*

- Transport sector – hydrogen fuel cells for vehicles.

Figure 3 illustrates the superstructure of the considered HSC.

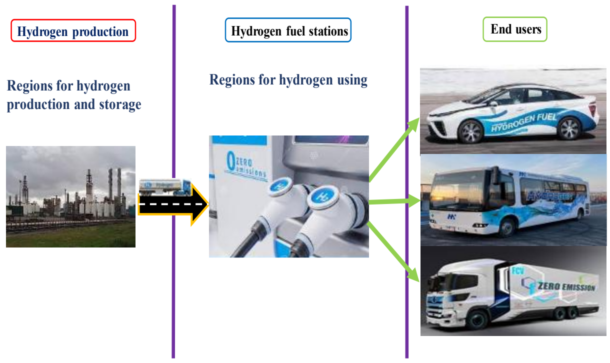

Figure 3. Superstructure of the HSC for hydrogen production *via* steam methane reforming with a focus on building hydrogen refueling stations.

Optimization framework of the approach

Optimal design of the HSC through SMR requires the definition of a multi-objective mathematical model that takes into account economic, environmental, and social factors. This model includes the following components:

Necessary data

To create the optimal model for designing a sustainable HSC through SMR, the following data is required:

1. *Economic Data*: Cost of Natural Gas; Cost of Water; Capital cost of SMR plants; Operational cost of SMR plants; Hydrogen storage cost; Transportation cost *via* trucks.

2. *Technical Data*: SMR facilities efficiency; Hydrogen yield per unit of methane; Production capacity of the SMR facilities; Hydrogen storage facilities; Transportation distance and mode.

3. Environmental Data: greenhouse gas emissions generated during hydrogen production; greenhouse gas emissions generated during hydrogen transportation; greenhouse gas emissions generated during hydrogen storage; Regulatory emission limits.

4. Demand & Location Data: Geographical locations of demand centers; Projected hydrogen demand over time; Availability of natural gas sources and infrastructure; Distance between production sites, storage, and hydrogen fuel stations.

5. Social and Economic Data: Labor costs related with employes hired in hydrogen production, storage, and transportation sites.

6. Regulatory Data: Government policies: Regulations, subsidies, or incentives for hydrogen production, transportation, and use.

Goal of optimization

The goal of solving the optimization problem is to determine the optimal operating conditions, as follows:

1. Structure of the Hydrogen Supply Chain.
2. Capacity for hydrogen produced through SMR process, considering the available renewable energy sources and technological efficiency, to meet the hydrogen demand.
3. Number, location, and capacity of the fueling stations that need to be built.
4. Performance of the fueling stations.
5. Number, location, and capacity of the hydrogen storage facilities that need to be built.
6. Design of the hydrogen transportation network (trucks, etc.), including the capacity, route selection, and distribution methods.
7. Number of jobs created as a result of the operation of all HSC elements.
9. Operational schedules for hydrogen production, storage, and transportation, ensuring system flexibility to handle varying demand patterns, renewable energy availability, and operational downtime.

The solution to this optimization problem provides a design that ensures not only the technical feasibility of the process but also its environmental, economic, and social sustainability, which is crucial for the future of the hydrogen economy.

Decision variables

For the purpose of defining the optimization model of the HSC, the following decision variables need to be introduced:

1. Continuous variable for the amount of hydrogen produced in the SMR facility.
2. Continuous variable for the amount of hydrogen transported from the SMR facility to the storage location.
3. Continuous variable for the amount of hydrogen transported from the storage location to the hydrogen fueling station.
4. Continuous variable for the amount of hydrogen stored in the respective facilities.
5. Continuous variable for the capacity of hydrogen production at SMR facilities.
6. Binary variable for establishing SMR facilities at a particular location.
7. Binary variable for selecting storage facilities.
8. Binary variable for transportation mode selection (truck).
9. The number of jobs that will be created for the building of facilities for production and operation of the hydrogen supply chain.

Mathematical model for the optimal design of a sustainable HSC through SMR process

The HSC is planned over a long-term horizon H of 10 years. This total period is segmented into a series of discrete time intervals t, each of which is further subdivided into multiple equal subintervals of duration ∇t.

Throughout the planning horizon, hydrogen consumption is expected to vary according to forecasted values. The optimal configuration of the sustainable HSC is mathematically represented as follows:

➢ *Mathematical model of environmental performance of the HSC -* $mTEI_t$, $[\mathrm{kg}_{\mathrm{CO_2eq}}/\mathrm{y}]$

The environmental criterion will include the environmental impact during the operation of the HSC through the greenhouse gas emissions generated at each time interval $t \in T$. These emissions are equal to the cumulative impact of each stage of the hydrogen production lifecycle through this process. Greenhouse gas emissions are typically determined as follows for each time interval $t \in T$:

$$mTEI_t = mELH_t + mEHS_t + mELD_t + mETT_t + mESW_t + mEAPP_t, \forall t \quad (4)$$

where:

$mTEI_t$ is the total environmental impact of the operation of the HSC over its entire lifecycle, $[\mathrm{kg}_{\mathrm{CO_2eq}}/\mathrm{y}]$;

$mELH_t$ are the total greenhouse gas emissions from the hydrogen production process through SMR, $[\mathrm{kg}_{\mathrm{CO_2eq}}/\mathrm{y}]$;

$mEHS_t$ are the total greenhouse gas emissions associated with the compression and storage of the produced hydrogen, $[\mathrm{kg_{CO_2eq}}/\mathrm{y}]$;

$mELD_t$ are the total greenhouse gas emissions from the production of diesel fuel associated with the transportation of hydrogen to the fuel stations, $[\mathrm{kg_{CO_2eq}}/\mathrm{y}]$;

$mETT_t$ is the environmental impact associated with the transportation of hydrogen to the fuel stations, $[\mathrm{kg_{CO_2eq}}/\mathrm{y}]$;

$mESW_t$ are the greenhouse gas emissions released during the utilization of waste (CO_2 as a by-product and components of natural gas such as N_2, H_2S, etc.), generated from hydrogen production for each time interval $t \in T$, $[\mathrm{kg_{CO_2eq}}/\mathrm{y}]$;

$mEAPP_t$ are the greenhouse gas emissions from the use of hydrogen as a fuel $[\mathrm{kg_{CO_2eq}}/\mathrm{y}]$.

➢ *Mathematical model of the economic performance of HSC - $mTHC_t$* $[\$/\mathrm{y}]$

The annual operating costs of hydrogen production through SMR include: capital costs for the construction of hydrogen production facilities through methane steam reforming; costs for purchasing the necessary raw materials for this production—water and electricity; production costs; costs for waste disposal from hydrogen production; costs for compressing and storing the produced hydrogen; transportation costs for raw materials and final products; maintenance costs for the built facilities; government incentives, etc.

The production costs take into account both the fixed annual operating costs, which are given as a percentage of the corresponding total capital investment, as well as the net variable cost, which is proportional to the processing amount.

The economic criterion represents the operating costs of the enterprise, which include the total investment costs for production facilities and the functioning of the HSC.

This price is expressed through the dependency for each time interval $t \in T$:

$$mTHC_t = mTIC_t + mTPC_t + mTPW_t + mTHH_t + mTHE_t + mTHS_t + mTMO_t + mTTC_t + mTTAXB_t + mTHSC_t + mTOEC_t - mT1L_t - mTA_t - mTH_t, \forall t \quad (5)$$

where:

$mTHC_t$ are the total annual costs of the HSC, [$/y];

$mTIC_t$ are the total investment costs for the production capacity of the HSC relative to the period of operation and the buyout of the installation per year, $[\$/\mathrm{y}]$;

$mTPC_t$ are the costs associated with hydrogen production, $[\$/\mathrm{y}]$;

$mTPW_t$ are the costs for waste disposal from hydrogen production (CO_2 as a by-product and components of natural gas such as N_2, H_2S, etc.), [$/y];

$mTHH_t$ are costs for raw material purchase – water, $[\$/\mathrm{y}]$;

$mTHE_t$ are costs for raw material purchase – electricity, $[\$/\mathrm{y}]$;

$mTHS_t$ are costs associated with compressing and storing the produced hydrogen, $[\$/\mathrm{y}]$;

$mTMO_t$ are costs for maintenance and operation of the hydrogen fueling stations, $[\$/\mathrm{y}]$;

$mTTC_t$ are the total transportation costs of HSC, $[\$/\mathrm{y}]$;

$mTTAXB_t$ is the carbon tax, levied based on the total amount of CO_2 generated during the operation of HSC, $[\$/\mathrm{y}]$;

$mTHSC_t$ are the total investment costs for the commercial capacity of the HSC relative to the period of operation and the purchase of the hydrogen station, $[\$/\mathrm{y}]$;

$mTOEC_t$ are the operational costs for the end hydrogen retailers, $[\$/\mathrm{y}]$;

$mT1L_t$ are government incentives for hydrogen consumption, $[\$/\mathrm{y}]$;

mTA_t is the total value of by-products (CO_2) $[\$/\mathrm{y}]$;

mTH_t is the revenue from the sale of produced hydrogen from all installed refueling stations, $[\$/\mathrm{y}]$;

➢ *Mathematical model of social performance of the HSC - $mJobs_t$* $[\$/\mathrm{y}]$

The model for the social assessment of the operation of the HSC determines the expected total number of jobs created as a result of the actions of all system components during its operation:

$$mJobs_t = mNJ1_t + mLT_tNJ12_t + mNJ2_t + mLT_tNJ21_t, \ \forall t \quad (6)$$

where:

the terms of equation (6) are determined according to the relationships at each time interval $t \in T$, $[\$/\mathrm{y}]$:

$mNJ1_t$ are the costs for creating jobs related to the installation of hydrogen production facilities, $[\$/\mathrm{y}]$;

mLT_tNJ12_t are the costs for creating jobs related to the operation of hydrogen production facilities, $[\$/\mathrm{y}]$;

$mNJ2_t$ are the costs for creating jobs related to the installation of hydrogen refueling stations, $[\$/\mathrm{y}]$;

mLT_tNJ21_t are the costs for creating jobs related to the operation of hydrogen refueling stations, [$/ y].

Constraints

For the design of HSC and the creation of a model using MILP, certain constraints need to be considered. These constraints vary in nature. They are the following:

- Production constraints related to the hydrogen produced through steam methane reforming; potential production from renewable energy sources; demand in different sectors such as industry, transportation, and residential needs; losses during compression and transportation.
- Constraints regarding the maximum capacity of steam methane reforming as well as the capacity of storage facilities.
- Environmental constraints regarding the allowable levels of carbon dioxide emissions according to adopted regulatory requirements.
- Transportation constraints regarding the total transport capacity, capacity of the transport mode, and the number of transport units.
- Budgetary constraints regarding the total investment and operational costs of the HSC.
- Constraints regarding the achievement of demand satisfaction and others.

Objective function

To solve the optimization problem for the optimal design of HSC using SMR, an objective function has been defined, which includes the previously defined optimization criteria regarding the environmental, economic, and social performance of HSC (equations 4, 5, and 6). It aims simultaneously to:

1. *Minimizing the environmental impact of the HSC:* The reduction of the environmental impact of the considered HSC can be achieved by minimizing: greenhouse gas emissions related to hydrogen production; its storage; the production of diesel fuel for the vehicles used for transportation; the transport of raw materials and products; the utilization of waste carbon dioxide; as well as the use of hydrogen as a fuel.

2. *Minimizing the economic impact of the HSC:* The minimization of total costs across all stages of HSC, including minimizing: the total investment costs for building facilities for hydrogen production through steam methane reforming; costs associated with hydrogen production; costs for disposing of waste from hydrogen production; costs for purchasing raw materials for hydrogen production (water, electricity); costs related to compressing and storing the produced hydrogen; operational costs; the carbon tax imposed on the generated greenhouse gas emissions during the operation of the HSC.

3. *Achieving social satisfaction for those employed in all elements of the HSC:* Maximizing the social and economic benefits of the HSC can be achieved by optimizing the costs of creating jobs associated with: the building of hydrogen production facilities; the operation of hydrogen production facilities; the building of hydrogen refueling stations; the operation of hydrogen refueling stations.

The overall optimization criterion is formulated as follows:

$$COST = \sum_{t\epsilon T} LT_t(mTEI_t + mTHC_t) \qquad (7)$$

$$Find: X_t[Decision\ variables]$$

$$MINIMIZE\{COST(X_t)\} \rightarrow (Eq.7) s.t.: \{Constraints\}$$

where:

LT_t are the durations of time intervals $t \in T$, [year].

The proposed model is formulated in terms of mixed-integer linear programming (MILP) and includes the necessary data, parameters, environmental, economic, and social models for the representation of the HSC, the objective function, and the constraints. The model is created with the aim of being further tested on a real-case study on the territory of Bulgaria with its 27 administrative regions. The optimization model will be defined and solved based on the environmental and economic optimization criteria, while the social criterion will be defined as a constraint using the commercial optimization software GAMS.

CONCLUSIONS

The study proposes a mathematical approach for the optimal design of a sustainable HSC, produced through steam methane reforming, with a focus on the construction of hydrogen fueling stations. The proposed model is defined in terms of mixed-integer linear programming (MILP) and includes the necessary parameters, decision variables, environmental, economic, and social models for the representation of the HSC, as well as the objective function and constraints. In the future, its application is planned on a real case study from Bulgaria to support investment decisions, reduce costs, and minimize the carbon footprint, which is crucial for the development of the hydrogen economy.

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