Review

Review and evaluation of hydrogen production options for better environment

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ABSTRACT

In this study, different hydrogen production sources and systems and some hydrogen storage options are comparatively investigated in detail. Economic, environmental, social, and technical performance and reliability of the selected options are compared in detail. Biomass, geothermal, hydro, nuclear, solar, and wind are the selected hydrogen production sources; biological, thermal, photonic, and electrical are the selected hydrogen production methods; and chemical hydrides, compressed gas, cryogenic liquid, metal hydrides, and nanomaterials are the selected hydrogen storage systems. In addition, some case studies and basic research needs to enhance the performance of hydrogen energy systems and to tackle the major challenges of the hydrogen economy are provided. The results show that solar has the highest environmental performance (8/10) and the total average ranking (7.40/10), nuclear has the lowest environmental performance (3/10), and geothermal has the lowest total average ranking (4/10/10) among selected hydrogen production sources. Hydrogen production systems' comparison indicates that photonic options have the highest environmental performance ranking (8/10), thermal options have the lowest environmental performance ranking (5/10), electrical options have the highest average ranking (7.60/10), and biological options have the lowest average ranking (4.80/10).

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Contents

1. Introduction

Hydrogen has the potential to provide economically feasible, financially promising, and socially advantageous, and energetically efficient solutions to issues related to the ever-increasing global energy demand, including global warming (Dutta, 2014). In addition, the recent studies show that it will be inevitable to initiate and accelerate the energy transition from traditional energy systems to innovative and sustainable alternatives (Shafiei et al., 2017).

Hydrogen is the fundamental pillar of the energy transition critically needed to combat global warming and other issues related to traditional energy systems (Zhang et al., 2016). As can be seen

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from Fig. 1, hydrogen systems could potentially have eight significant roles during this energy transition (Dincer and Acar, 2016):

- Large-scale renewable energy integration to the existing energy infrastructure
- Accessible, reliable, safe, clean and affordable energy to all sectors and regions
- Highly resilient energy systems
- Integration to multigeneration systems to offer many valuable products with minimal losses
- Cleaner transportation via fuel cells and hydrogen-fueled internal combustion engines
- Cleaner energy source to the industry, residential applications, buildings, etc.
- Cleaner heating, cooling, drying, and power to all end users of the energy sector
- Cleaner industrial feedstock

Hydrogen has the potential to provide clean, efficient, reliable, and affordable solutions in all these application fields with significant social benefits (Kalinci et al., 2015). In the literature, it is reported that hydrogen can enable wide use and full market penetration of renewable energy sources (Singh et al., 2016). On the end-user (service) side hydrogen is a critical complement to electricity to store intermittent renewables which is a great step towards greenization of the energy systems (Cipriani et al., 2014).

In the literature, it is reported that hydrogen could meet 18% of the final energy demand, reduce 6 Gt of CO_2 emissions annually, and create 30 million new jobs by 2050 (Uyar and Besikci, 2017). The literature also shows that hydrogen could power over 400 million cars, 15–20 million trucks, and around 5 million buses in 2050, which make up about 20–25% of the transportation industry (Mostafaeipour et al., 2016). Along with its significant economic and environmental benefits, hydrogen energy systems are expected to operate at higher efficiencies in the future (Nakamura et al., 2015). The literature shows that hydrogen is seen as the key to sustainable growth and solution to global warming issues (González et al., 2015).

Hydrogen has another advantage which is the ability to be built on the existing natural gas infrastructure in buildings (Gong et al., 2016). The literature shows that hydrogen could possibly meet about 10% of the global building heating demand by 2050 (Valente et al., 2018). Another advantage of hydrogen is the efficient use of medium and high-temperature heat processes in the industry (Maroufmashat et al., 2016). In these processes, electricity is not a very effective solution so hydrogen could be the perfect solution (Sgobbi et al., 2016). Furthermore, hydrogen production methods are getting more efficient, affordable, and environmentally friendly and less fossil fuel dependent as the innovative hydrogen production pathways and systems are developed (Salvi and Subramanian, 2015). In addition to being a sustainable energy storage medium and heating/cooling source, hydrogen is a valuable industrial chemical feedstock used mainly in methanol, ammonia, and steel production processes (Acar and Dincer, 2014).

Accomplishing a fully developed hydrogen economy requires well-established hydrogen energy systems from better resources, production methods, end use options (3S approach) including the storage and distribution of hydrogen (Dincer and Acar, 2015a). A fully developed hydrogen economy is essential to combat global warming and critical for a sustainable future with significant environmental, economic, and societal benefits (Göllei et al., 2016). Hydrogen does not only eliminate greenhouse gas emissions, but it can also eliminate other emissions which damage the environment and cause global warming (Sarrias-Mena et al., 2015). These emissions are sulfur oxides, nitrogen oxides, and particulates causing smog (Owierkowicz and Malinowska, 2017). Hydrogen can also minimize noise pollution by replacing fossil fuel combustion processes in the transportation sector, in buildings, and in the industry (Marchenko and Solomin, 2015). Furthermore, energy security can be accomplished with hydrogen economy by taking advantage of domestic, locally available, reliable, and secure energy and material sources for hydrogen production, distribution, storage, and end-use (De Santoli et al., 2017).

In order to accomplish a fully developed hydrogen economy and to make hydrogen a critical component for the energy market, significant research and investment are required on hydrogen production systems (Chintala and Subramanian, 2015). The goal is to make hydrogen production systems more efficient, affordable, reliable, safe, and ready for different types of end-user needs such as small/large scale, portable/stationary, etc. As the current hydrogen production systems use already available technologies, the need is to set up a well-developed hydrogen infrastructure (Joshi et al., 2016). This can be achieved by increasing hydrogen production systems' capacities and subsequently reducing costs which would lead to wide acceptance by the public, industry, and the governments.

In the transportation sector, hydrogen-fueled internal combustion engines and fuel cells can possibly be used together with battery electric vehicles to reach the true greenization of all land, air, and water transportation (Maleki et al., 2016). Fuel cell vehicles are also reported to have some advantages over battery electric vehicles such as lower investment costs and quicker fueling and refueling. In addition to transportation, significant quantities of hydrogen are utilized as feedstock during methanol refining and production, ammonia synthesis, and iron and steel industry. Producing hydrogen from clean and sustainable sources could have a direct impact on the greenization of these industries as well (Mehrpooya et al., 2017). Hydrogen can also provide heating, cooling, and power to buildings by using the existing natural gas infrastructure. The literature estimates that in the near future, there might be entire cities converting to solely hydrogen based heating and cooling (Gao et al., 2017). Last but not least, hydrogen can also become a crucial renewable energy storage medium and if necessary can be used for clean electricity production. With hydrogen, it is possible to store and transport renewable energies effectively over long distances and time periods (Beheshti et al., 2016). For that reason, hydrogen is a fundamental component during the transition to 100% renewable energy systems to eliminate global warming.

In this study, a thorough evaluation of the long-term potential of hydrogen production systems to combat global warming is presented along with a roadmap to tackle the significant challenges and threats to a well-developed hydrogen economy. Hydrogen production systems have the potential to greenize all end users of
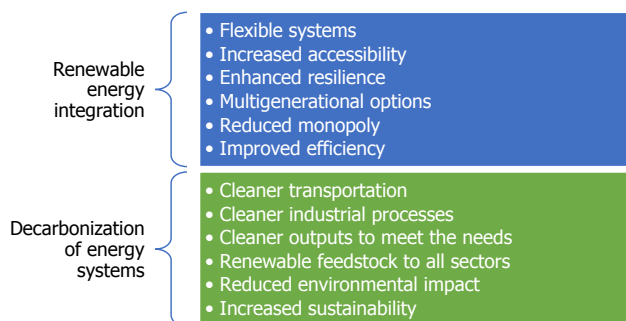

Renewable energy integration
- Flexible systems
- Increased accessibility
- Enhanced resilience
- Multigenerational options
- Reduced monopoly
- Improved efficiency

Decarbonization of energy systems
- Cleaner transportation
- Cleaner industrial processes
- Cleaner outputs to meet the needs
- Renewable feedstock to all sectors
- Reduced environmental impact
- Increased sustainability

Fig. 1. Hydrogen's critical roles during the energy transition to combat global warming.

the energy sector such as transportation, industry, buildings and so on and hydrogen can make well-developed renewable energy systems a practical option for all end users of the energy sector. For this reason, this study intends to use the 3S (Source-System-Service) approach to hydrogen production systems and aims to discuss the sustainability of novel hydrogen production, storage, distribution, and end use options. In the open literature, there is a limited number of studies comparatively assessing different hydrogen production sources and systems and hydrogen storage options by using different technical, social, environmental, and economic criteria. This study is to first to provide a broader assessment perspective to all three aspects of hydrogen options: (i) hydrogen production sources, (ii) hydrogen production systems, and (iii) hydrogen storage systems by using 17 different perspective criteria to comparatively assess their economic, environmental, social, and technical performance as well as reliability. Another critical goal of this study is to provide a guide to the industry, academia, and the governments to combat global warming with the help of hydrogen production systems. Therefore, in the end, key research needs and directions and transition steps toward a fully developed hydrogen economy is provided.

2. 3S approach to hydrogen

In this section, a 3S approach introduced by Dincer and Acar (2015a) is applied to hydrogen energy systems. For a truly sustainable approach to combat global warming, hydrogen energy systems must be considered from source including all energy and material sources to a system including hydrogen production, storage, and distribution systems and finally to service including different end-use options such as fuel cells, internal combustion engines, and so on (Walker et al., 2016). This approach is depicted in Fig. 2.

By using the approach provided in Fig. 2, the potential of hydrogen energy systems to combat global warming is evaluated and thoroughly discussed. In this study, hydrogen energy systems are considered as the key to sustainability and hydrogen is also considered a versatile fuel with practical advantages for a broad variety of end-use applications such as transportation, industrial processes, residential applications, buildings, etc. When considering hydrogen in future energy systems, a realistic approach is taken into account. The approach used in this study does not consider fictional solutions in imaginary case studies. Instead, this study only considers the existing systems both in large and small scales, their scalability, potentials, strengths, weaknesses, research challenges and needs, and possible future directions.

In this study, the aim is to identify and discuss hydrogen's role in energy systems to combat global warming. For this reason, a

roadmap is presented to achieve fully developed and deployed hydrogen energy systems. In addition, within the scope of the 3S approach; the necessary short-, medium-, and long-term milestones are discussed and investment requirements for wide-deployment of hydrogen energy systems are identified for hydrogen production, storage, delivery, and end use systems for a sustainable future.

Eliminating the negative impacts of global warming is essential for a sustainable future (Creutzig, 2016). In hydrogen energy systems, the first point to look at should be the source selection (Sharma and Ghoshal, 2015). In order to be truly sustainable and environmentally benign, hydrogen should be produced from clean, abundant, reliable, and affordable energy and material sources (Hosseini et al., 2015). In cases where the source is not continuous, a well-developed storage system must be integrated into the hydrogen production network (Sethia and Sayari, 2016). Renewables seem to be the most advantageous sources for hydrogen production systems (Shaner et al., 2016). However, most of the renewables are not available in large scale energy systems due to cost issues, therefore, more traditional systems (i.e., fossil fuel powered processes) can be used for hydrogen production with proper carbon capture and emissions control during the initial part of the transition to a well-developed hydrogen economy (Gradisher et al., 2015). In the course of this period, advancements in material sciences and technology are expected to lower the cost, enhance the efficiency and enable the possibility of large-scale operations for renewable-based hydrogen energy systems (Nastasi and Basso, 2016). When affordable, reliable, clean, and efficient renewable-based hydrogen systems are available on larger scales, there will be a faster transition from fossil fuels to renewables for better sustainability (Shafiei et al., 2015).

Selecting the most suitable energy and material sources is essential in hydrogen energy systems (Yilmaz et al., 2016). However, if not accompanied by the proper system(s), it might not be possible to get the most optimum outcome from the selected source(s) (Bundhoo and Mohee, 2016). In hydrogen energy systems, the "system" step should be efficient, clean, reliable, safe, available in small and large scales, and applicable to portable and stationary systems (Asghar et al., 2015). The "system" step can be divided into three categories for hydrogen energy: hydrogen production systems, hydrogen storage systems, and hydrogen delivery (distribution) systems (Sinigaglia et al., 2017). Hydrogen production systems can be classified by the use of primary energy type; biological, electrical, electrochemical, electrothermal, photochemical, photoelectrochemical, photonic, thermal, and thermochemical are some of the most commonly used alternatives in the literature (Dincer and Acar, 2015b). It should be noted that there could be additional novel options in the future depending on the advancements in material science and technology (Choi et al., 2015). Hydrogen delivery systems connect end users and hydrogen production systems (Demir and Dincer, 2018). The most commonly used hydrogen delivery options are storage tanks, pipelines, and vessels (Singh et al., 2015). The expectation from these systems can be listed as the ability to support varying demand, applicability to both small and large scales, reliability, safety, and zero or minimal losses (Ogden et al., 2018). In hydrogen storage systems, compressed gas, cryogenic liquid, and metal hydrides are the most commonly used options (Aneke and Wang, 2016). In addition, depending on the advancements in materials science and technology, novel hydrogen storage options can be introduced in the future (Liu et al., 2016). The key expectations from hydrogen storage systems can be listed as (Chanchetti et al., 2016):

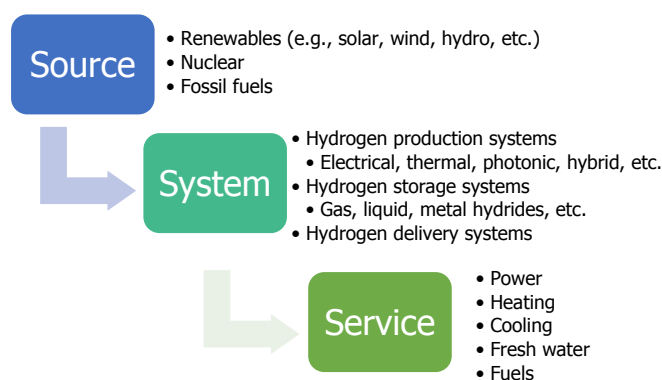- high gravimetric and volumetric energy density
- high power output

Fig. 2. 3S approach to hydrogen energy systems.

- low energy and power cost
- zero or minimal emissions and waste
- safe operation
- accessibility
- ease of use
- efficient operation
- low-performance degradation
- long lifetime
- minimal or zero loss during charging/storage/discharging

Hydrogen has a very wide range of applications that could be benefited by different end-user types which are considered as the service step of the hydrogen energy systems. The service step of hydrogen energy systems can be grouped based on the end user types such as an energy source for transportation (Ahmed et al., 2016), industry (Otto et al., 2017), residential applications (Maleki et al., 2017), power generation (Prananto et al., 2017), and heating (Dodds et al., 2015) and cooling (Khani et al., 2016) purposes. In addition, hydrogen can be used as a material feedstock in industrial processes, for instance in ammonia (Bicer et al., 2016) and methanol production processes (Bellotti et al., 2017) and in the steel industry (Rao et al., 2019).

Currently, the transportation industry relies almost completely on fossil fuels, and as a result, the transportation sector contributes to more than 20% of the global CO_2 emissions (Van Fan et al., 2018). If it is not greenized, the transportation sector's share on global CO_2 emissions is expected to increase in the future with the increase in global population and mobility (Creutzig et al., 2015). Hydrogen produced, stored, and distributed by following the 3S approach in a sustainable manner could significantly lower the emissions of the transportation sector (Siyal et al., 2015). The expectations from hydrogen energy systems in the transportation sector can be listed as a long driving range, flexible operation, and high performance (Moliner et al., 2016). Hydrogen can be used in fuel cells and internal combustion engines to meet the energy demands of the transportation industry in a sustainable manner (Maniatopoulos et al., 2015).

The industry is the second largest energy consumer, following the power sector. Currently, the industry contributes to more than 30% of the global final energy consumption (Fais et al., 2016). Due to the heavy reliance on fossil fuels, the industrial energy consumption contributes to around 25% of the global CO_2 emissions (Schandl et al., 2016). The five industries with the highest energy consumption from the highest to lowest consumption are iron and steel (28% of the industrial energy consumption), chemicals, petrochemicals, and refining (14% of the industrial energy consumption), cement (8% of the industrial energy consumption), aluminum (5% of the industrial energy consumption), and pulp and paper (5% of the industrial energy consumption) industries (Sorrell, 2015). A major reason behind this high energy demand of these industries is their high-grade heat requirement (Schulze et al., 2016). In these industries, hydrogen could be a lot more affordable, clean, and efficient energy option than using fossil fuels or electricity to generate high-grade heat (Bakenne et al., 2016).

The energy demand of both residential and commercial buildings are very high due to their heating, cooling, and power requirements (Reinhart and Davila, 2016). Some major energy consuming processes in buildings are space heating and cooling, hot water, lighting, appliances, and so on (Yarbrough et al., 2015). The energy demand of buildings is a lot higher than the transportation sector and almost as high as the energy demand of the industry in total (Arteconi et al., 2017). This high energy demand is mainly met by fossil fuels which is a reason behind the negative impact of buildings on global warming (Delmastro et al., 2017). To tackle this issue, buildings must be greenized with highly efficient

energy systems (Frayssinet et al., 2018). Greenization of buildings can be accomplished via waste heat recovery (i.e., district heating/cooling combined with traditional power plants), heat pumps, and transitioning to hydrogen energy (Wong et al., 2015). Hydrogen has the advantage to be easily integrated into the existing natural gas network with minimal modification (Alanne and Cao, 2017). Hydrogen can effectively provide heating, cooling, power, drying, and fresh water to the buildings in a safe, reliable, and affordable manner (Nastasi and Di Matteo, 2017).

In addition to their significant energy consumption, chemical and petrochemical industries are using large quantities of hydrogen as an industrial feedstock (Ball and Weeda, 2015). Among these industries, ammonia production for urea and other fertilizers is the largest consumer of hydrogen making about 51% of the total industrial hydrogen demand (Kuntke et al., 2017). Second highest hydrogen consumer as industrial feedstock is the refining industry where hydrogen is used for hydrocracking and hydrotreating such as desulfurization which makes around 31% of the total industrial hydrogen demand (Speight, 2016). When combined together, ammonia and refining industries consume more about 82% of the total industrial hydrogen demand (Setoyama et al., 2017). The remaining consumers of hydrogen as an industrial feedstock are methanol production, fuel processing, glass production, and so on (Hanley et al., 2018). With the global population increase and rising standards of living the demand for hydrogen as industrial feedstock is expected to keep increasing as well (Simon et al., 2015). Therefore, it is essential for the industry that the hydrogen supply comes from clean sources to reduce the negative impact of the industry on global warming (Liu et al., 2015). Further greenization of the industry could be achieved by substituting carbon with hydrogen as a reductant in steel production (Karakaya et al., 2018). In addition, hydrogen could be used to capture the industrial byproduct CO_2 into commercially viable products such as methanol (Liu and Liu, 2015). Hydrogen's role as an enabler of carbon recycling in the industry is shown in Fig. 3.

3. Review methodology of hydrogen options

In this study, recent data from the literature on the environmental, economic, social, and technical performance of hydrogen production sources and systems as well as hydrogen storage
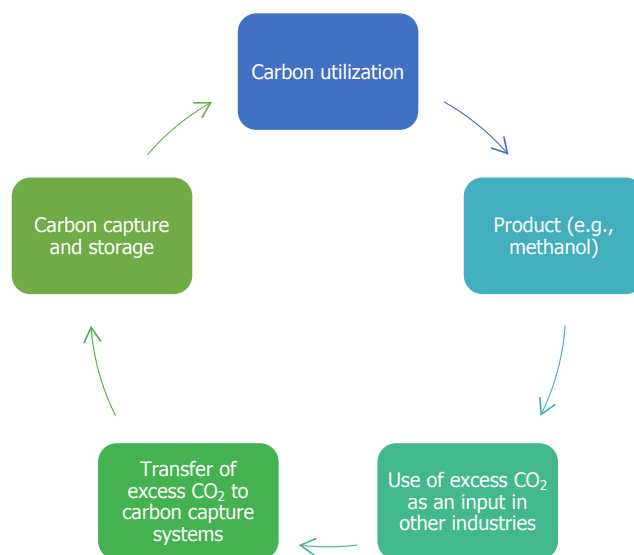

Fig. 3. Hydrogen's role as an enabler of carbon recycling in the industry.

options are reviewed and comparatively evaluated. Here, the studies are selected firstly based on their publication year: more specifically, the studies published before 2015 are not taken into review process as the aim is to evaluate the most recent data in the literature. ScienceDirect database is used during the review process and the content of publications are categorized based on the following criteria:

- Studies focusing on hydrogen production
  o Studies evaluating hydrogen production sources
  o Studies evaluating hydrogen production systems
- Studies focusing on hydrogen storage systems

These studies are further classified and selected based on their evaluation criteria. In this study, publications focusing on one or more of the following criteria when evaluating hydrogen production and storage options are selected for sustainability analysis. These criteria are:

- Economic performance
  o Initial cost (USD/kg H_2)
  o Running cost (USD/kg H_2)
- Environmental performance
  o GHG emissions (kg CO_2 eq./kg H_2)
  o Land use (m^2 land requirement/kg H_2)
  o Water discharge quality
  o Solid waste generation (kg solid waste/kg H_2)
- Social performance
  o Impact on public health
  o Employment opportunities
  o Training opportunities
  o Public acceptance
- Technical performance
  o Energy efficiency (%)
  o Exergy efficiency (%)
  o Process control
  o Raw material input (kg raw material/kg H_2)
- Reliability
  o Dependence on imported resources
  o Predictability
  o Scalability

Detailed information on the selected criteria and how they affect the overall sustainability analysis of the selected hydrogen options are given in the next section. Here, the most recent data found in the literature are provided along with their scope and key findings (Table 1). When selecting the data from the literature, first "hydrogen production" keyword is used in ScienceDirect and all review articles are retrieved, resulting 39,081 published articles. After filtering the studies published before 2015, the remaining studies (14,253) are narrowed down by using classification criteria as "energy" and "sustainability". For comparison purposes, the studies which comparatively assess at least four different hydrogen production sources or systems by using at least four different criteria are selected. In addition, the studies which do not explain the evaluation procedure or cite the sources of any previously published results are eliminated. The literature review process has led to four sources which are presented in Table 1 based on their selected hydrogen production sources and systems and performance criteria.

Table 2 shows the selected hydrogen storage studies along with their scope and performance evaluation criteria. Here, "hydrogen storage" keyword is used in ScienceDirect and the results are restricted to review articles, and 18,157 results are retrieved. Next, the articles published before 2015 are eliminated and as a result, the number of articles is lowered to 7510. In the next step, "energy" and "sustainability" keywords are used to further narrow down the results and the remaining articles are scanned through to select the studies with useful data on at least five different hydrogen storage options which are comparatively assessed by using at least five different performance criteria. Furthermore, the studies which do not describe the assessment methodology clearly or cite the sources of any previously published results are not taken into account. As a result of this process, six literature sources are selected which are listed in Table 2 based on their evaluated hydrogen storage systems and performance criteria.

4. Sustainability analysis of hydrogen production options

In this study, economic, social, environmental, and technical performance and reliability of hydrogen production sources and systems and hydrogen storage options are comparatively assessed. The selected options are shown in Fig. 4.

In economic performance evaluation criteria, initial and running costs are taken into account. The initial cost is the capital cost requirement which is comparatively assessed in the literature based on USD/kg hydrogen. The running cost includes operation and maintenance costs which are also given in terms of USD/kg hydrogen. The overall economic performance ranking is calculated by taking the average of initial and running cost rankings.

The environmental performance comparison is conducted based on GHG emissions, land use, water discharge quality, and solid waste generation. GHG emissions are given in terms of kg CO_2 eq./kg hydrogen production and land use is the amount of land area required to produce or store hydrogen (m^2 land requirement/kg hydrogen). Water discharge quality is ranked by experts in the literature within a range of 0–10.0 means low quality which indicates the selected option has polluted water discharge or the discharge water is too hot or too cold which disturbs the ecosystem. On the other hand, 10 means the best option with clean water discharge at closer temperatures to the environmental state. Solid waste generation is given in terms of kg solid waste generated/kg hydrogen production. The overall environmental performance ranking is calculated by taking the average of GHG emissions, land use, water discharge quality, and solid waste generation rankings. The literature data used in this study are taken from the references mentioned in Tables 1 and 2.

Impact on public health, employment and training opportunities and public acceptance are the social performance criteria. These social performance criteria are ranked by experts in the literature. The social performance criteria rankings are also provided in the 0–10 range where 0 indicates a high negative impact on public health, or no employment or training opportunities or no public acceptance. On the contrary, 10 means no negative impact on public health, or vast employment or training opportunities or full public acceptance. The overall social performance ranking is calculated by taking the average of impact on public health, employment opportunities, training opportunities, and public acceptance rankings.

In technical performance criteria, energy and exergy efficiencies, process control, and raw material input data are taken into account by using the numerical results and expert performance evaluations provided in the literature. Energy efficiency rankings are between 0 and 10 and the evaluation is done based on the following equation:

Table 1
Selected data sources from the recent literature used in the sustainability analysis of hydrogen production options in this study.

Source	Hydrogen Production Sources	Hydrogen Production Systems	Performance Criteria	Notes
Hosseini and Wahid (2016)	Wind Solar Hydro Geothermal Nuclear Natural gas Biomass Coal Oil	Electrolysis Reforming Gasification Partial oxidation Fermentation Pyrolysis Thermochemical cycles	Cost (initial and running) Efficiency (energy and exergy) Maturity Predictability Scalability Dependence on imported sources Raw material input Process control Public acceptance	Overview of the state-of-the-art hydrogen production technologies using renewable and sustainable sources
Dincer and Acar (2017)	Coal Natural gas Solar Wind Hydro Geothermal Biomass Nuclear	Thermolysis Thermochemical Photocatalysis Photoelectrochemical Biophotolysis Photofermentation Artificial photosynthesis	Emissions Efficiencies (energy and exergy) Cost (initial and running) Renewability (raw material use) Staff (employment/training opportunities) Scalability Safety Support (public acceptance)	Comparative assessment of selected hydrogen production sources and systems from the 18S point of view
Nikolaidis and Poullikkas (2017)	Nuclear Solar Wind Biomass	Electrolysis Thermochemical Pyrolysis Gasification Photolysis Dark fermentation Photofermentation Thermolysis Photoelectrolysis	Production rate Capacity factor (%) Cost (initial and running) Efficiency	Technical and economic evaluation of different hydrogen production sources and methods
Acar et al. (2018)	Fossil fuels Wind Solar Nuclear	Electrolysis Thermochemical cycles Photoelectrochemical cells	Initial and running costs GHG emissions Land use Water discharge quality Solid waste generation Impact on public health Employment and training opportunities Public acceptance Energy and exergy efficiencies Process control Raw material input Dependence on imported resources Predictability Scalability	Comparative investigation of the sustainability of hydrogen production systems

$$Energy\ efficiency\ ranking = \frac{Energy\ content\ of\ the\ desired\ product}{Energy\ content\ of\ the\ input} \times 10 \tag{1}$$

Similarly, exergy efficiency rankings are between 0 and 10 and the calculations are done as shown in the equation below:

$$Exergy\ efficiency\ ranking = \frac{Exergy\ content\ of\ the\ desired\ product}{Exergy\ content\ of\ the\ input} \times 10 \tag{2}$$

Process control rankings of the experts use the 0–10 scale as well, 0 meaning hardest process control and 10 indicating easiest process control. Raw material input includes any material requirement of the selected option from clean water to catalysts, biomass etc. The unit of raw material input is kg raw material input/

kg hydrogen production. The overall technical performance ranking is calculated by taking the average of energy efficiency, exergy efficiency, process control, and raw material input rankings.

Dependence on imported resources, predictability, and scalability are the availability and reliability criteria. These selected performance indicators are ranked by experts in the literature. Similarly, rankings are also provided in 0–10 range where 0 indicates sole dependence on imported resources or no predictability or no scaling options. On the other hand, 10 means no dependence on imported resources or extensive predictability or numerous available scaling options. The overall availability and reliability ranking is calculated by taking the average of dependence on imported resources, predictability, and scalability rankings.

In this study, the data from the literature are taken as a basis. Water discharge quality, impact on public health, employment and training opportunities, public acceptance, and process control, dependence on imported resources, predictability, and scalability are already ranked within 0–10 scale by experts in the literature. The data for the remaining categories are taken from the literature

Table 2
Selected data sources from the recent literature used in the sustainability analysis of hydrogen storage options in this study.

Source	Hydrogen Storage Systems	Performance Criteria
Niaz et al. (2015)	Compressed gas	Initial and running costs
	Cryogenic liquid	Area requirement (land use)
	Chemical hydrides	Waste generation
	Metal hydrides	Employment and training opportunities
	Nanomaterials	Energy and exergy efficiencies
		Raw material input
		Predictability
Zhang et al. (2015)	Compressed gas	Employment and training opportunities
	Cryogenic liquid	Emissions
	Chemical hydrides	Safety
	Metal hydrides	Scalability
	Nanomaterials	Acceptability
Zhang et al. (2016)	Chemical hydrides	Emissions
	Metal hydrides	Employment and training opportunities
	Compressed gas	Safety
	Cryogenic liquid	Initial and running cost
	Nanomaterials	Predictability
Reuß et al. (2017)	Chemical hydrides	Emissions
	Metal hydrides	Water use and discharge
	Compressed gas	Safety (health impact)
	Cryogenic liquid	Acceptability
	Nanomaterials	Process control
		Imported resources
		Scalability
Ren et al. (2017)	Chemical hydrides	Resource requirements
	Metal hydrides	Area requirement
	Nanomaterials	Employment and training opportunities
	Compressed gas	Acceptability
	Cryogenic liquid	Energy and exergy efficiencies
Nagpal and Kakkar (2018)	Chemical hydrides	Imported resources
	Metal hydrides	Acceptability
	Nanomaterials	Control
	Compressed gas	Area requirement
	Cryogenic liquid	Safety
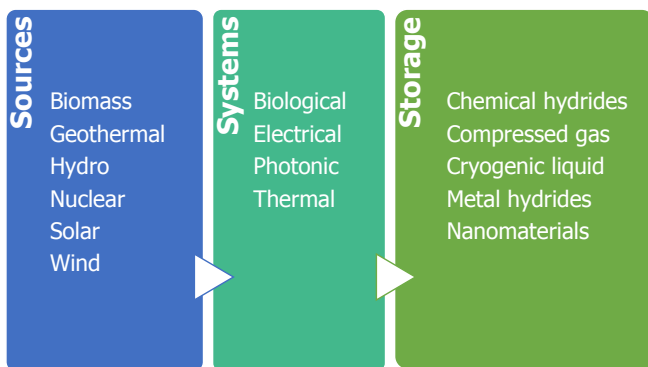
Fig. 4. Selected hydrogen production sources and systems and storage options.

and ranked and normalized to 0–10 scale. For categories that are aimed to be minimized such as initial and running costs, GHG emissions, land use, solid waste generation, and raw material input:

$$Ranking = \frac{maximum - data_{selected\ option}}{maximum - minimum} \times 10 \qquad (3)$$

Here, *maximum* and *minimum* indicate the highest and lowest initial and running costs, GHG emissions, land use, solid waste generation, and raw material input among selected hydrogen production sources and systems and storage options. The data$_{selected\ option}$ stands for the initial or running cost, GHG emissions, land use, solid waste generation, or raw material input of the selected hydrogen production source or system or storage option. The

procedure of normalizing energy and exergy efficiency rankings are given in Equations (1) and (2), respectively. The primary aim of the sustainability assessment procedure explained in this section is to provide a consistent and clear ranking methodology for the readers which would effectively rank all selected options within 0–10 scale where 0 indicates the least desired point and 10 indicates the ideal scenario in terms of sustainability.

5. Results and discussion

In this section, economic, environmental, social, and technical performance and reliability of hydrogen production sources and systems, storage and end-use technologies are comparatively assessed based on the recent findings in the literature. In economic performance, initial (capital) cost and running (operating and maintenance) cost criteria are taken into account. GHG emissions, land use, water discharge quality, and solid waste generation are the environmental performance criteria. The indicators of social performance are the impact on public health, employment opportunities, training opportunities, and public acceptance. Energy and exergy efficiencies, process control, and raw material input requirements are considered in the technical performance evaluation. And last, dependence on imported resources, predictability, and scalability are the reliability criteria. The selected options are assigned scores between 0 and 10, based on their performance. In all cases, 0 means poor performance such as high costs, high emissions, and land use, and more damage to the environment, high negative impact on the society or less social benefits, low efficiencies, high material requirements, high dependence on imported sources, less predictability, and lack of scalability. On the other hand, 10 means ideal performance such as low costs, low

emissions, and land use, and less damage to the environment, low negative impact on the society or more social benefits, high efficiencies, less material requirements, less dependence on imported sources, high predictability, and scalability.

In the first step, the economic, environmental, social, and technical performance of energy sources for hydrogen production is comparatively assessed. Selected energy sources for hydrogen are biomass, geothermal, hydro, nuclear, solar, and wind. The performance data of the selected sources are gathered from the sources mentioned in Table 1. The environmental performance ranking results of the selected hydrogen production sources are given in Fig. 5 and the overall performance results are presented in Fig. 6. The environmental performance ranking results show that in terms of GHG emissions, the wind is the most favorable source and biomass has the highest emissions. In terms of land use, solar requires the least area and nuclear has the maximum land area requirements. Solar and wind are the most favorable in the water discharge quality category while geothermal and nuclear seem to have the lowest water discharge qualities. In terms of solid waste generation, nuclear has the highest solid waste and solar has the lowest. The results show that solar has the highest average performance (7.40/10) as hydrogen production source, followed by hydro and wind (6.00/10), biomass (5.80/10), nuclear (4.60/10), and geothermal (4.60/10). Solar based hydrogen production has significant advantages since all solar-based processes are taken into account such as photoelectrochemical cells, solar thermochemical cycles, photobiological processes, and so on. Most of these processes obviously have a very low negative impact on the land, air, and water sources, highly reliable, and have high social performances, such as the low negative impact on the public health. In order to make solar based hydrogen production processes competitive with traditional systems, their economic performance must be enhanced by lowering initial and maintenance costs. Both geothermal and hydro have some risks associated to impact on the natural habitat and water discharge quality, and these issues reduce their performance score. Nuclear has a solid waste risk and public acceptance concerns and if these issues are resolved along with lowering the costs, it can be a promising source of hydrogen production for a sustainable future.

Fig. 5 shows that in terms of GHG emissions, among the proposed options, the wind option seems to be the most environmentally benign and the efforts should focus on to lower the GHG

emissions of biomass. In terms of land use, solar seems to be the best option and nuclear seems to require the largest area due to extensive safety and control requirements such as cooling and solid waste and pressure control. Waste discharge quality rankings show that solar and wind have the least negative impact on clean water reserves as opposed to geothermal and nuclear. Geothermal option's negative impact is due to the risk of polluting the underground water sources. Nuclear, on the other hand, has a low ranking because of the temperature of the wastewater coming from cooling towers directly sent to lakes and rivers which harms the water ecosystem. Solid waste generation category shows that solar is the most environmentally benign option while nuclear, due to the radioactivity of its solid waste, has the lowest ranking. When all environmental performance criteria are taken into account, solar seems to be the most environmentally benign option and nuclear has the lowest environmental performance ranking. Overall, hydro has the highest performance in the economic evaluation category since it is a well-developed and scaled-up technology. The wind has the lowest economic performance as hydrogen production source. From the social and technical dimensions, the geothermal option has the lowest rankings. both solar and wind have high social performance rankings since they end up with the lowest negative impact on public health, and they are further considered to have major new job opportunities.

Biological, electrical, photonic, and thermal hydrogen production options are comparatively investigated to present the recent status of the "System" step of the hydrogen energy systems. In Table 3, some advantages and challenges of the selected systems are presented. The environmental performance ranking results of the selected hydrogen production systems are given in Fig. 7. The environmental performance ranking results show that in terms of GHG emissions, photonic systems are the most favorable options. In terms of land use, photonic and electrical hydrogen production systems are advantageous. Photonic systems are the most favorable in the water discharge quality category. And the overall technical, economic, environmental, and social performances and the reliability of the selected systems are presented in Fig. 8. The performance data of the selected hydrogen production systems are gathered from the sources mentioned in Table 1. The results show that electrical hydrogen production systems have the highest performance (7.60/10), followed by thermal-based systems (6.60/10), photonic systems (5.40/10), and biological processes (4.80/60). In
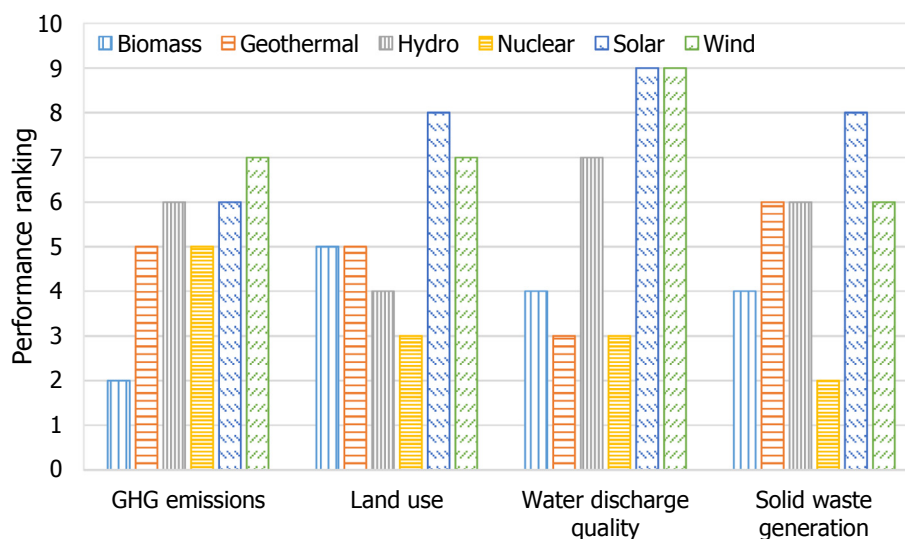

Fig. 5. Environmental performance ranking results of the selected hydrogen production sources.
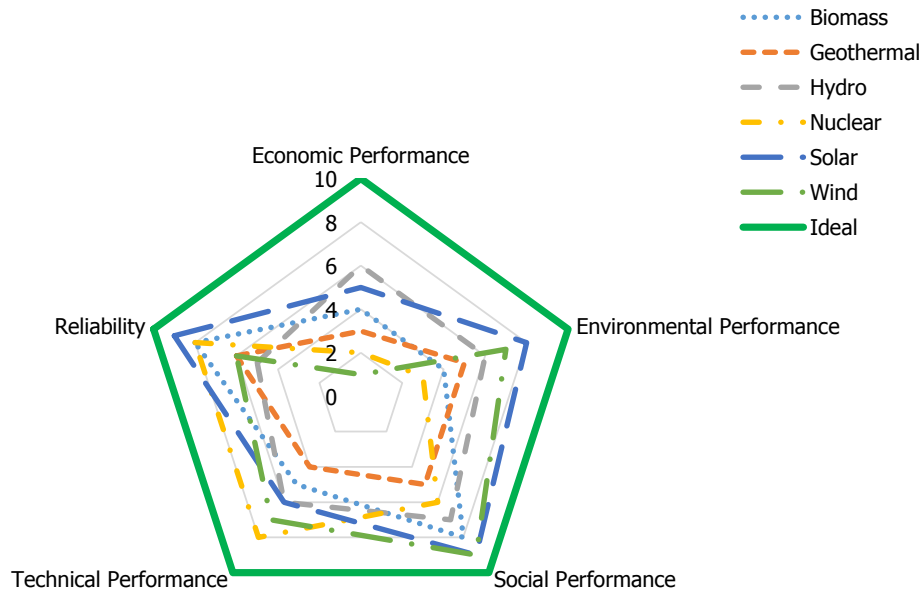
Fig. 6. Performance comparison results of selected energy sources for hydrogen production.

Table 3
Main advantages and challenges of the selected hydrogen production systems.

| System	Advantages	Challenges
Biological	Potentially large resources	Slow hydrogen production, large area requirements, research needed to find the most suitable biological organism for optimum production and large-scale production
Electrical	Commercial availability, proven technology, well-understood process, modularity, highly pure production, applicable to small and large scales as well as stationary and portable needs	Competition with green or renewable electricity
Photonic	Environmentally benign process, abundant source, less damage to health, good efficiencies, applicable in small and large scales and can be portable or stationary	High cost, groundbreaking research needed to find novel materials for the effective and affordable caption of solar energy
Thermal	Low cost, appropriate for large-scale production, less damage to the environment, can take advantage of the industrial waste heat	Complex process, research is needed to develop corrosion and heat-resistant materials
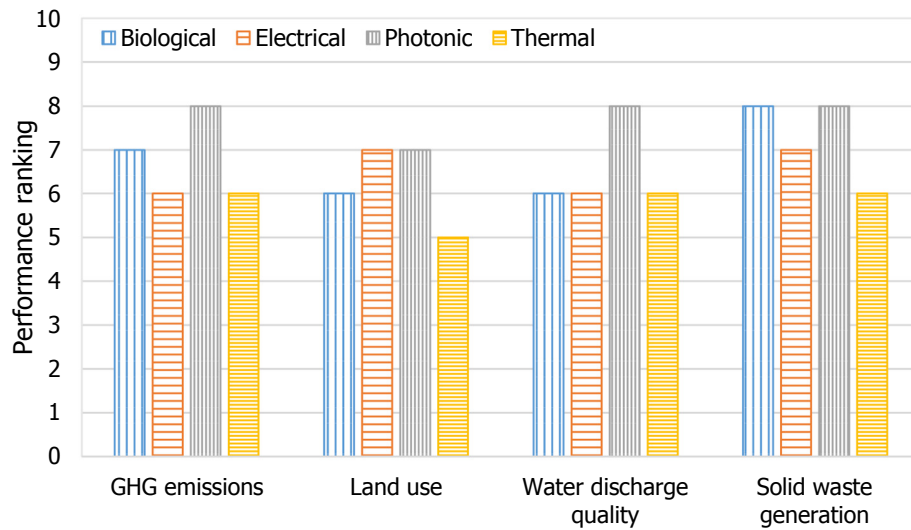
Fig. 7. Environmental performance ranking results of the selected hydrogen production systems.

addition to electrical hydrogen production systems, the economic performance evaluations of all selected systems may significantly be enhanced by lowering the capital cost of renewable-based hydrogen production systems. Since most of the selected systems can be integrated with renewable and clean sources, their environmental performance appears to be satisfactory. The biological
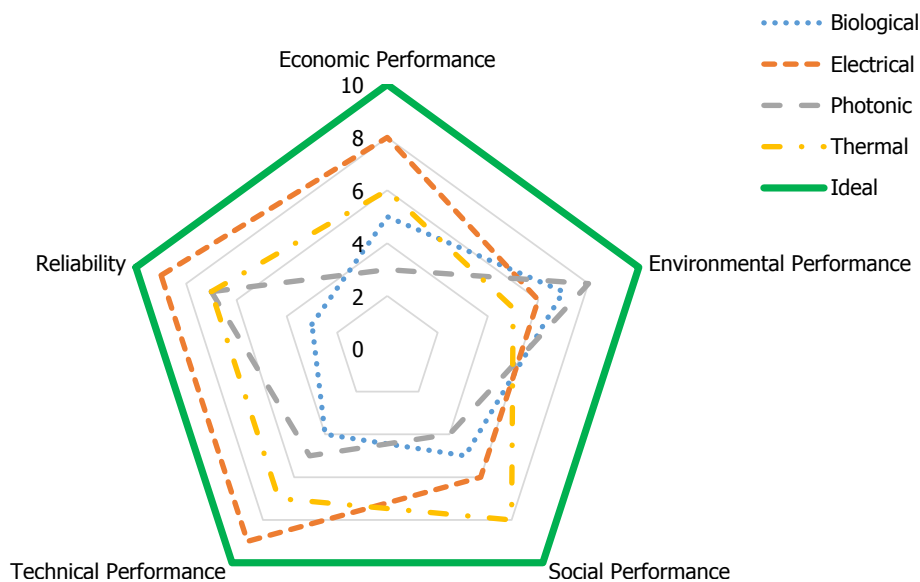
Fig. 8. Performance comparison results of selected hydrogen production systems.

hydrogen production systems also have the reliability issue since controlling and predicting the behavior of biological organisms may become quite challenging.

Fig. 7 shows that in terms of environmental performance criteria, including GHG emissions, land use, water discharge quality, and solid waste generation, thermal hydrogen production options are seen to be most harmful. However, the thermal hydrogen production option has the highest social performance. In all environmental performance categories, photonic (solar based) hydrogen production options have the highest performance rankings, therefore, are considered to be the most environmentally benign options. On the other hand, photonic hydrogen production has the lowest economic rankings since this technology is comparatively new and more technological advancements are needed to lower its initial cost. In terms of economic, technical, and reliability performances; electrical based hydrogen production options are the most promising due to well-developed technology and the long history of water electrolysis. The biological hydrogen production options are seen as the least preferred options in terms of technical performance and reliability because microorganisms are seen to be harder to control, regulate, and optimize. Therefore, it is harder to estimate/quantify the production rate and scale up hydrogen production via biological options.

In Table 4, the advantages and challenges of the selected hydrogen storage systems are listed and compared which are chemical hydrides, compressed gas, cryogenic liquid, metal hydrides, and nanomaterials. The environmental performance ranking results of the selected hydrogen storage systems are given

in Fig. 9 where the environmental benefits of nanomaterials can clearly be seen. In Fig. 10, comparative assessment results of economic, environmental, social, and technical performance and reliability of selected storage systems are presented. These results are gathered from the recent literature provided in Table 2 which show that among the selected storage options, nanomaterials have the highest score with 8.40/10 average, followed by chemical and metal hydrides, with average scores of 6.80/10 and 6.60/10, respectively. Cryogenic storage options have the lowest average score (3.40/10), followed by compressed gas (6.00/10). The reason behind the poor performance of cryogenic liquids are the extreme temperature requirements and associated safety risks which increase their costs as well. In addition, the loss of hydrogen is very high in this option, especially compared to the other selected options. In order to minimize such losses, the cryogenic systems must be highly insulated which also increase the complexity and cost of the storage option. Therefore, cryogenic storage option has the lowest performance rankings in all categories: economic, environmental, social, technical, and reliability. Similar to the cryogenic option, the compressed gas has some operational requirements due to high operating pressures which may bring a safety issue that effects system cost and social performance as well. Overall, nanomaterials appear to be a promising option for future hydrogen energy systems. This is because nanomaterials emit the least amount of harmful solid, liquid, or gaseous waste; have the highest efficiencies with minimal losses; therefore high reliability; and possible job creations.

In this section, the comparative performance evaluation results

Table 4
Main advantages and challenges of the selected hydrogen storage systems.

| Storage option	Advantages	Challenges
Chemical hydride	Well understood, reversible reactions and compact design	Waste generation and logistics, infrastructural change needs
Compressed gas	Existing and well-developed technology, availability and low cost	Very high pressures needed increasing safety issues and cost, lower storage densities compared to traditional fuels
Cryogenic liquid	Existing and well-understood technology, higher densities compared to compressed gas	Very low temperatures needed increasing cost, loss of stored hydrogen, energy intensity of the process, lower densities compared to traditional fuels
Metal hydride	Safe and modular operation with wide applicability, relatively high densities	Recycling of the storage material and waste issues
Nanomaterials	High energy densities	At early research and development phase, costs yet to be decreased
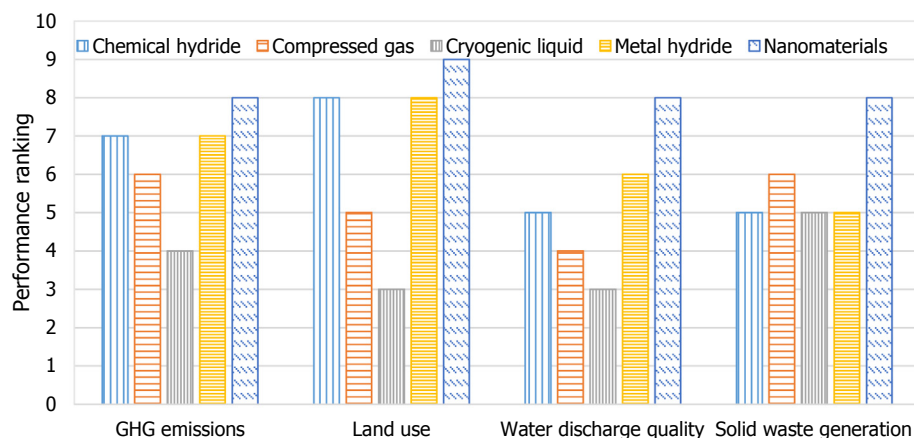
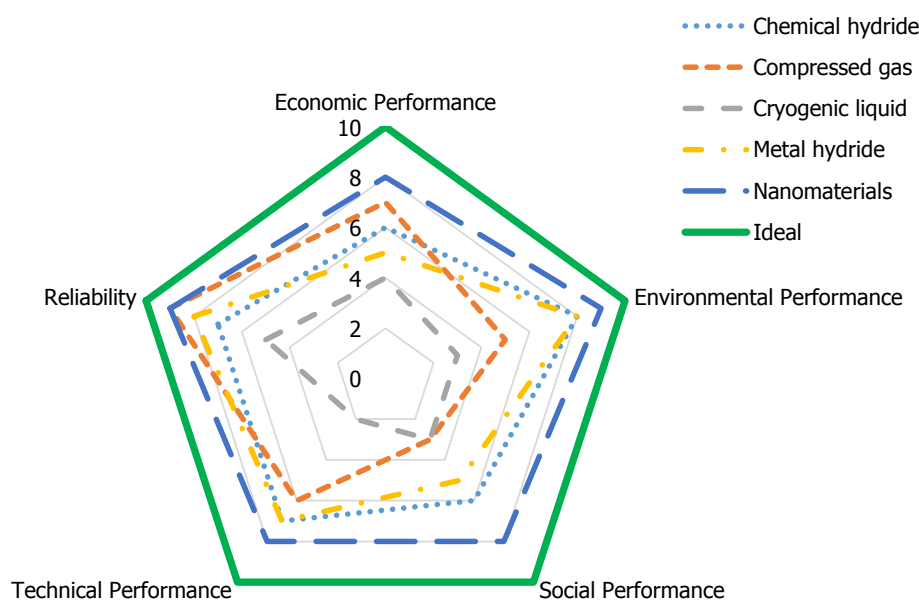Fig. 9. Environmental performance ranking results of the selected hydrogen storage systems.

Fig. 10. Performance comparison results of selected hydrogen storage systems.

of hydrogen production systems and sources and storage systems are presented and discussed. Here, the aim is to give a broad perspective on the current status and anticipated future research directions on hydrogen energy systems. By investigating the economic, environmental, social, technical performance and reliability of the components of hydrogen energy systems, the goal is to accelerate the transition to hydrogen economy to combat global warming. When produced, stored, and used in a sustainable manner, hydrogen could potentially greenize the industrial processes, residential applications, and transportation sectors.

6. Future Directions

Developing, constructing, and widely developing hydrogen energy systems to meet the energy needs require significant investments and innovative approaches to current energy systems along the entire hydrogen chain. A well-developed hydrogen chain has to meet the energy demands in all scales (e.g., small and large scales) and for all types of needs (e.g., stationary and portable). In addition, the 3S approach must be followed to develop this

hydrogen chain including production, distribution, storage, retail, fueling infrastructure, and all end-use applications. The key research needs of the elements of the entire hydrogen energy chain are presented in Fig. 11, respectively.

Hydrogen production requires a significant amount of research to greenize existing hydrogen production systems. Currently, hydrogen production systems are heavily dependent on fossil fuels and the critical need is to switch to renewables to make hydrogen supply truly "carbon free". Most of the renewable hydrogen production systems have low efficiencies, high investment costs, or they are only available in small scale. Therefore, the essential need is to develop hydrogen production systems that are efficient, cost-effective, and clean which can work off-grid in remote areas as well. In hydrogen production, use of non-renewable material and energy sources must be eliminated or minimized. In addition, novel hydrogen production systems must not have very high and low temperature or pressure requirements for safety and economic reasons.

For wide use and high market penetration of hydrogen energy systems, hydrogen storage systems and hydrogen transportation,
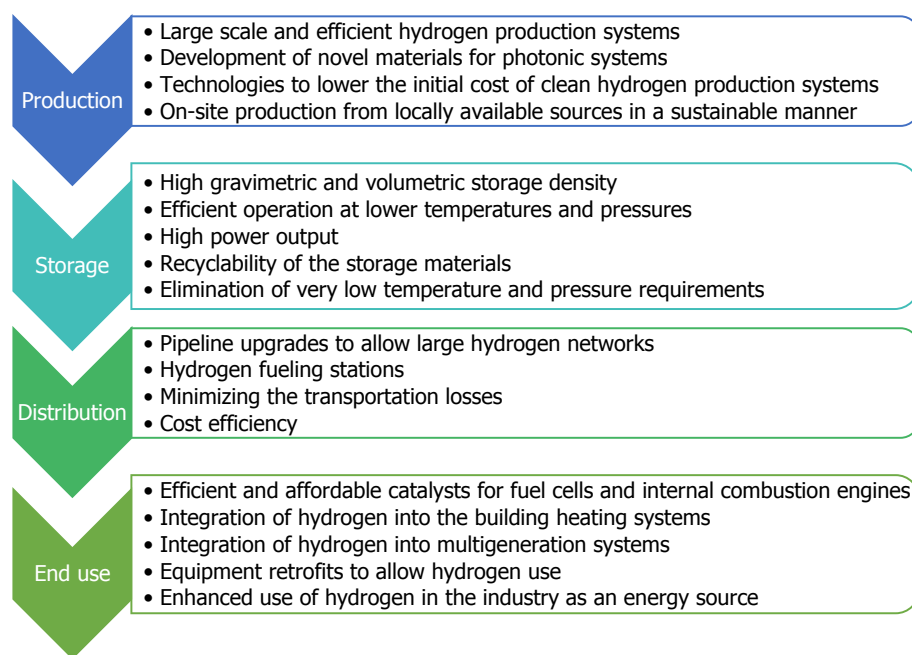
Fig. 11. Key research needs of the elements of the entire hydrogen energy chain.

and distribution network should be strengthened. The research needs include enhancing the gravimetric and volumetric densities of hydrogen storage systems, developing affordable storage options at medium temperature and pressure ranges, reducing the charging and discharging times of storage systems, and identifying the optimum strategies for stationary, portable, small, and large-scale applications. These items are related to sustainable hydrogen storage. In addition to these needs, recyclability and reuse of hydrogen storage mediums must be enhanced by increasing the lifetime of these materials. This can be done by finding novel materials which do not have corrosivity and performance degradation issues and can store hydrogen in small spaces and lighter systems. It should be noted that these materials are also expected to become cost competitive with the existing elements in the traditional energy systems. Hydrogen distribution is essential for broad range utilization of hydrogen which requires research and development to modify the existing natural gas pipelines for long distance hydrogen distribution with minimal losses. Currently, hydrogen is either transported as compressed gas or cryogenic liquid which are both expensive and not very efficient. Any technological advancements to enhance the performance of hydrogen storage systems would impact the hydrogen distribution network performance in a positive manner as well. And last but not least, a well-developed distribution network with easily accessible fueling stations would greatly help the wide utilization of hydrogen by all types of end users. This requires significant cost reduction in hydrogen storage, transportation, and distribution networks which could lead the customers to prefer hydrogen energy systems in buildings and industry as well as in the transportation systems.

The research activities on hydrogen energy systems could eventually become a fundamental component of the transition from fossil fuels to clean energy systems to combat global warming. Hydrogen energy systems are required to have higher efficiencies and lower costs at larger scales which can be achieved via continuous improvements in materials science and technology. The cost of hydrogen energy systems has already been reduced significantly with the introduction of innovative renewable energy systems and

developments in some end-use applications of hydrogen such as fuel cells. Further cost reduction is needed to lower the cost of hydrogen production which could be attained by large-scale hydrogen production. The key research directions for wide deployment of hydrogen in different sectors are presented in Fig. 12.

The wide use of hydrogen in the transportation sector depends on the developments in fuel cell vehicles and hydrogen-fueled internal combustion engines. In addition, the retail price of hydrogen has to be competitive with existing transportation fuels. In order to tackle issues related to global warming, hydrogen must be cleaned, transported, and stored in a clean, efficient, safe, and affordable manner. The distribution network of hydrogen has to be well-developed, with adequate fueling stations which are easily accessible by end users. Onboard hydrogen storage systems' performance must be enhanced significantly while keeping hydrogen-fueled vehicles' cost competitive with conventional vehicles. In addition, the efficiency of each item in the hydrogen energy system should be increased while keeping the safety of hydrogen-powered vehicles a priority.

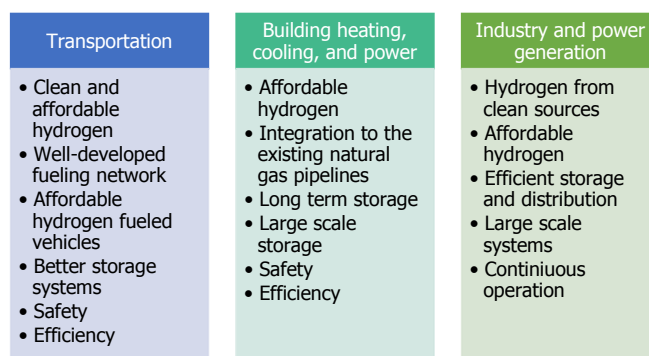The research and development requirements to make hydrogen

Fig. 12. Key research directions for wide deployment of hydrogen in different sectors.

widely used in buildings are less complex compared to the transportation industry. The first step could be integrating hydrogen into the existing natural gas network in buildings for residential needs such as cooking, hot water, and heating. The modifications required to make this switch and to eliminate or minimize hydrogen losses in pipes previously used for natural gas must be completed in the first step. Like the transportation industry, cost-competitive hydrogen is required by the buildings industry. Again, the hydrogen used in the buildings must be sustainable with affordable prices, high efficiency, low negative impact on the environment, and safety. The buildings industry require effective long-term and medium-to-large scale hydrogen storage. Safety and efficiency are also key in the buildings industry.

Hydrogen deployment in industrial processes as a fuel and as industrial feedstock could be achieved in a decentralized manner. This means, hydrogen can be integrated into the industry plant by plant and increase its market share gradually. The industrial processes and power generation sector require large amounts of hydrogen in a continuous manner so large scale and effective hydrogen storage is essential in the industry. The cost of hydrogen production is expected to be the key determiner during the deployment of hydrogen in the industry. Wide utilization of hydrogen in the industry requires hydrogen price to be either equal to or less than the cost of fossil fuels.

All of the research directions presented here aim to make hydrogen energy systems available and ready to be applied to all types of end-user needs. Transition to fully developed hydrogen energy systems is presented in Fig. 13. The first step is technology development where research is conducted to meet end-user requirements and establish a commercialization scenario. The second step is initial market penetration where commercialization of initial hydrogen energy systems starts in the transportation, buildings sector, and the industry for both small and large and stationary and portable needs. The second step is expected to include prototypes of hydrogen energy systems and the goal is to enhance their market penetration and performance. In the third step, investment is required to make hydrogen available for all end-user types and needs. In the fourth phase, a fully developed hydrogen market and infrastructure are expected where hydrogen energy systems are available in all regions for all types of energy demand. By making clean, affordable, reliable, and safe hydrogen efficiently used for all types of energy requirements from power to heating and cooling, drying to fresh water production, the dependence on fossil fuels could be eliminated or minimized. The transition from fossil fuels to renewable hydrogen could play critical roles in tackling issues related to global warming.
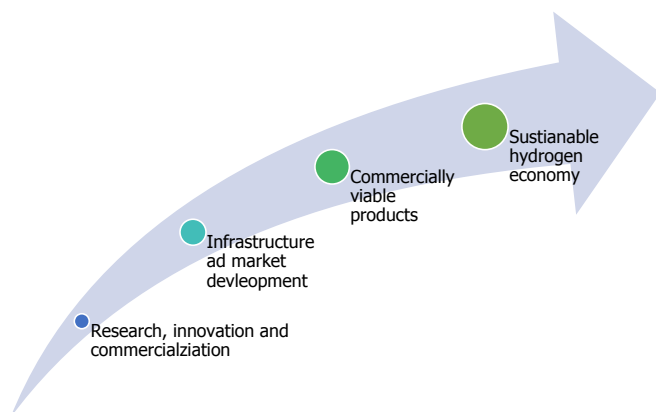

Fig. 13. Transition steps towards fully developed and the sustainable hydrogen economy.

7. Conclusions

Hydrogen energy systems are essential components of solutions towards reducing the negative consequences of global warming. For a sustainable future, hydrogen should be affordable, reliable, safe, clean, and efficient. For this reason, the aim of this study is to comparatively evaluate technical, environmental, social, and economic performance and reliability of hydrogen by using the 3S (source-system-service) approach. By selecting the most sustainable source for the most sustainable hydrogen production and storage systems and by enhancing the performance of hydrogen end-use technologies such as fuel cells and internal combustion engines, the dominant role of fossil fuels in energy systems could be eliminated. The main findings of this study can be summarized as follows:

- In terms of environmental performance, the solar option appears to be the most advantageous hydrogen production source with 8/10 ranking and the nuclear has the lowest ranking of 3/10.
- When all economic, environmental, social, technical and reliability criteria are taken into account, on average, the wind has the highest performance ranking (7.40/10) while the geothermal has the lowest (4.60/10).
- The environmental performance ranking comparison of hydrogen production systems show that the photonic systems have the highest ranking (8/10) while the thermal systems have the lowest (5/10).
- Upon considering all performance criteria overall, the electrical hydrogen production systems are the most advantageous (7.60/10), and the biological systems have the lowest ranking (4.80/10).
- Among selected hydrogen storage options, the nanomaterials have the highest ranking of 9/10 in the environmental performance category, and the cryogenic liquid has the lowest ranking (3/10)
- When all performance criteria are taken into account, the nanomaterials are the most advantageous hydrogen storage options (8.40/10) while the cryogenic liquid has the lowest average overall ranking (3.40/10)
- Solar based electrical (such as photoelectrochemical) hydrogen production accompanied by hydrogen storage with nanomaterials could be the most environmentally benign and sustainable option.

In closing, this study focuses on the sustainability aspects of hydrogen production sources and systems together with storage alternatives. In the open literature, there are some studies focusing on hydrogen production sources or hydrogen production systems or hydrogen storage options separately. Furthermore, there is a lack of studies focusing on social, technical, financial, and environmental aspects of sources and systems required for sustainable hydrogen production. By considering the technical, economic, environmental and social performance assessments of hydrogen production and storage options together with their reliability, this study is one of the first of its kind, to provide a broader sustainability investigation of hydrogen production and storage together. In future studies, hydrogen end-use options (such as different fuel cells and internal combustion engines) could be included in this 3S approach to even further enhance the sustainability analysis of hydrogen energy systems for a sustainable future.

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