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Review Article

Hydrogen as Fuel for ICEs: State of Art and Technological Challenges

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The climate change, as the main consequence of the polluting emissions due to anthropic activities, is nowadays a well-known threat to human health as well as for the environment safety. Several plans and strategies have been announced by the governments to detach from fossil fuel-based energy and gradually reduce the carbon footprint of their economies. In this scenario, the road transport sector is turning out to be the main "technological gym" to test and improve new powertrain solutions to achieve as soon as possible the goal of the net-zero carbon emissions, at least in the tank-to-wheel (TTW) context. In view of this, the hydrogen as fuel is gaining ever more attention from the scientific community, both as the middle-term solution to achieve the abovementioned goal and as a prominent future energy carrier. In this review, the performance and main characteristics of the hydrogen-internal combustion engines (H2-ICEs) are discussed based on the most recent studies available in literature. A comprehensive overview of various topics is offered, from the production stage to the combustion anomalies, mixture formation strategies, and the challenge of reducing the nitrogen oxides (NOx) emissions under high load conditions.

1. Introduction

The increasing energy demand expected in coming years and decades [1], combined with the strong dependence of the current society on fossil fuels [2], is pushing the various governments to the search for new and alternative solutions capable to guarantee a "soft" transition towards more sustainable energy production models. This modal shift mainly passes through investments for the production of energy coming from renewable sources [3]. Anyway, even though most of the countries agree on the need for a rapid common action to mitigate and limit the temperature increase below 1.5°C by 2030 with reference to the preindustrial period, there are several obstacles that could make this and future goals difficult to achieve. Some of these relate to the economic acceleration and associated greenhouse gas (GHG) emissions expected from China and India [4], not to mention the relatively recent conflict between Russia and Ukraine which has prompted many European countries back to partially relying on energy production derived from fossil coal [5] to compensate the gas shortages due to the stop of supplies from Russia. Few examples show how fragile is the current energy production model. Not only that, the road transport sector, perhaps one of the most susceptible sectors to the "world events," still relies mainly on fossil fuels. In view of this, the development of alternative energy carriers and innovative powertrains for automotive applications has become fundamental, and among the range of possible solutions, hydrogen is gaining a renewed interest. H2 is a very simple and abundant element and thanks to its properties, it has been used in various industrial processes in the last century. Moreover, the improvement of production technologies, as well as the growing interest for its application in the road transport sector, is leading to a rapid increase in its global production [6]. In 2020, the European Commission announced the hydrogen strategy and in 2022, the REPowerEU plan, both aimed to boost the EU production of energy from renewable sources, including green and low-carbon hydrogen to reduce its dependence on imported fossil fuels [7, 8]. Similarly, the U.S. department of energy released the National Clean Hydrogen Strategy and Roadmap in 2023 [9]. These legislative measures include electrifying industries and mobility, supporting the decarbonisation of those sectors hard to detach from fossil fuels with hydrogen, such as heavy-duty road transport, maritime, and aerospace fields. Furthermore, these plans are also aimed to set favourable conditions for the development of internal market for hydrogen using the already existing gas infrastructure (e.g., the creation of the ENNOH, European Network of Network Operators for Hydrogen). However, there are several hurdles to be addressed before hydrogen actually becomes a cost-efficient solution, from the huge investments needed [10] to the improvement of production technologies needed to make the hydrogen a cost-effectiveness competitor [11]. Hence, the abovementioned premises are not intended to provide or justify future predictions about the role that the hydrogen may or may not play to tackle down some of the challenges related to the climate change. The author endeavours only to provide an overview on technical pros and cons of the hydrogen's life cycle assessment (LCA), from the production stage to its use in internal combustion engines with a comparison with the main alternatives that are under investigation, namely, the fuel cell vehicles (FCEVs) or those that have already gained space for large-scale production and on road applications, i.e., battery electric vehicles (BEVs).

2. Why H2-ICEs?

There are several studies providing different opinions on the validity of using hydrogen as energy carrier and its future as viable solution for on road transport, as well as the many comparisons in the literature with FCEVs and BEVs. However, as a matter of fact, the BEV is the only technology which already found space for in series production and on road application. Nevertheless, there are different aspects to be taken into account, ranging from the greenhouse gas emission along all the LCA, costs of production, limits of application, and last but not least, the costumer's acceptance. For example, both FCEVs and H2-ICEs use hydrogen as fuel but the working principles are completely different. A fuel cell follows the electrolysis technique, thus it consists of anode and cathode where chemical reactions take place and generate direct current (DC) and water as a byproduct, while inside an ICE, the hydrogen works like conventional fuels, thus leading to a combustion process inside the combustion chamber so as to transfer work to the engine crankshaft. FCs are characterized by a high efficiency as well as being pollution free; in addition, they can be divided into two main types as follows: the proton exchange membrane fuel cell (PEMFC) and solid oxide fuel cell (SOFC). However, the latter is poorly suitable for automotive application due to the high light-off temperature and the presence of ceramic materials which fragility makes it unsuitable for the transport sector. Moreover, the main "Achille heels" for the FCs lie in the high production cost [12] and the need of extremely pure hydrogen to avoid the sulphur-poisoning effect which causes the deactivation of bound sites with consequent irreversible loss of catalytic activity towards the hydrogen oxidation reaction [13]. Moving to the BEVs, these operate depending only on the battery pack capacity, thus avoiding all the issues related to the hydrogen storage system (which is one of the main technological challenges for both FCEVs and vehicles equipped with a H2-ICE), but on the other hand, limiting the range to typical values around 200–400 km, depending on the passenger car model [14]. BEVs, as well as the FCEVs, can be considered zero emission vehicles in a tank-to-wheel (TTW) context and they have advantages such as the production costs that are partially offset by the absence of complicated mechanical modules needed for the conventional vehicles and improved driving performances. Anyway, the key concerns on BEVs come to surface when talking about heavy-duty vehicles, where the limited range and extended time required for recharging strongly hinder its applicability for this sector. In addition, another aspect that should be taken into account is also the one about the LCA of battery packs which need the so called "rare-earth" materials whose concerns for supply are already rising with the increasing risk of having fluctuating price trends in the future, with consequent repercussions on costumers [15]. Coming to the last option examined, there are several combustion strategies suitable for H2-ICEs. The mainstream solution involves the retrofit of spark plug-ignited engines. In such engines, the hydrogen can be injected into the intake manifold (PFI) or directly inside the combustion chamber (DI). Further technical detail and challenges will be discussed in the dedicated chapters of this review, but the abovementioned premise highlights the first advantage of the latter solution; the possibility to retrofit old engines and thus to use an already well-known technology. Then, focusing on the propulsion systems which involve the use of hydrogen as fuel, several studies have been performed to compare the LCA to study the influence of current technologies on the environment in terms of emissions. In [16], the authors have considered the best scenario by comparing alternative powertrain solutions fuelled with green hydrogen and using a CNG vehicle as benchmark. Thus, taking into account the global warming impact (GWP), acidification impact potential (AP), and cumulative nonrenewable energy demand (CED), it was observed that the use of green hydrogen always entails improved performance in terms of environment impact and energy demand compared to the conventional fuel. The situation was found to be different for the FCEVs that turned out to be the worst option with regards to the AP. Indeed, for the latter, the infrastructure (i.e., the vehicle manufacturing) is very "expensive" due to the need of processing rare materials, the presence of the battery pack (although smaller in size than a BEV), and other electrical components which are present to a lesser extent in vehicles equipped with H2-ICEs. Figure 1 shows the LCA comparison among main powertrain alternative solutions.

Obviously, the above graph considers the best possible scenario where the hydrogen fuel comes from renewable energy sources and the operating points of the alternative powertrains are not demanding in terms of drive cycle (i.e., characterized by low NOx emissions). Therefore, depending on the production methods, the previous considerations con

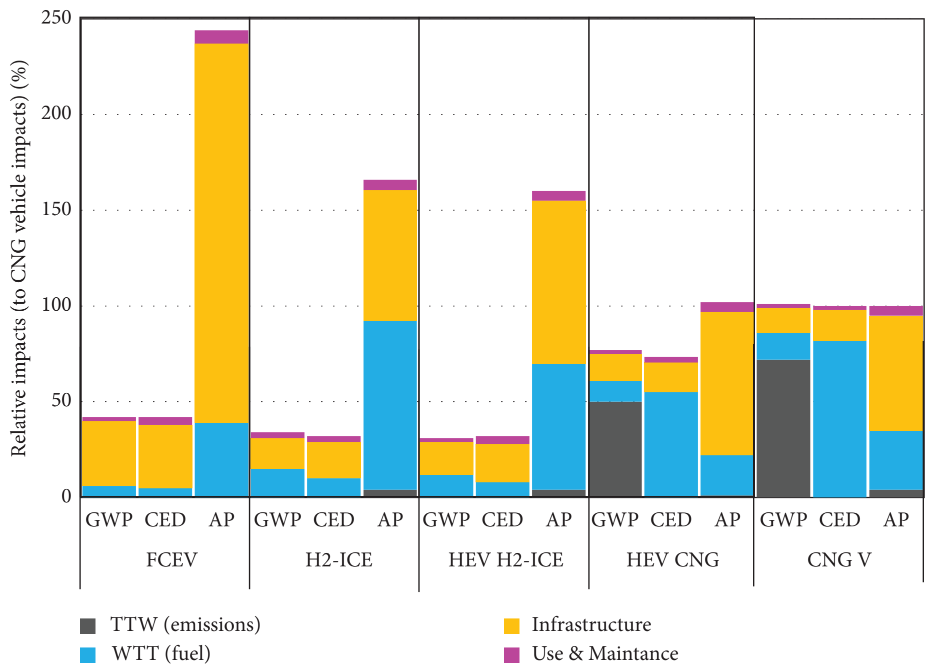

Figure 1: Relative environmental impacts of the three pure-hydrogen vehicle systems and the two CNG systems. Adapted from [16].

lead to a completely different situation in terms of impact on the environment. Figure 2 [17] shows the CO_2 equivalent emissions due to the production of grey, blue, and green hydrogen, thus highlighting the sensitivity of the available studies to the "boundary conditions" and then the need for more investments required to shift the production patterns to the cleanest option as soon as possible.

In the end, when talking about H2-ICEs, there are also different technological challenges to deal with (i.e., the mixture formation, abnormal combustions, boosted intake pressure, and NOx emissions) that need to be solved before talking of a possible in series production. Therefore, it is clear that a single silver bullet does not exist, including the H2-ICEs, but each technology is characterized by different pros and cons which more or less justify its use in a certain sector. Tables 1–3 provide a literature survey on main topics treated in this review.

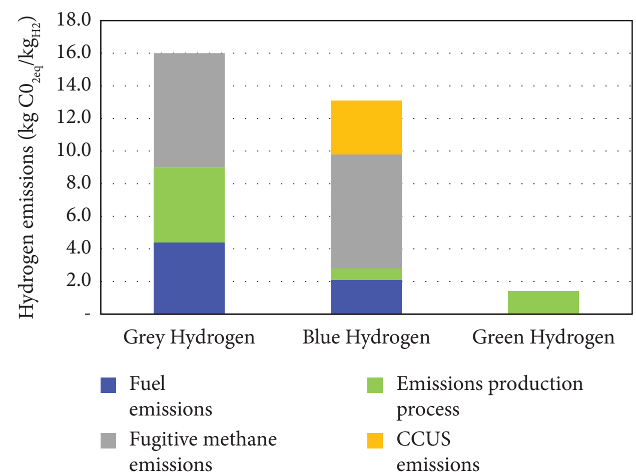

Figure 2: Hydrogen emissions for various production methods. Adapted from [17].

3. Hydrogen Production, Transport, and Storage

3.1. Hydrogen: Production Processes. One of the main reasons for which hydrogen gained the current renewed interest is its possibility of being produced through various processes, from the large-scale diffused steam methane-reforming (SMR) process to the laboratory-scale electrolysis technique. There are different "types" of hydrogen depending on the energy source used to produce it, i.e., grey, blue, and green hydrogen [39], where the colour provides an overall idea about the environmental sustainability degree of the different methods (Figure 3, top). The "grey" refers to hydrogen produced using fossil-based energy sources. The blue hydrogen differs for the addition of carbon dioxide capture step and then returning a (almost) net-zero carbon emissions. Lastly, the green hydrogen is produced from renewable energy sources, mainly through the electrolysis process. Unfortunately, the latter only accounts for a minimum share of the total amount of hydrogen produced (i.e., <5%) [41] and the situation does not improve even when looking at the cost competitiveness [40], where the green hydrogen can reach peaks of price more than doubled if compared to the grey and blue counterparts (Figure 3, bottom).

Table 1: Literature survey on hydrogen production, transport, and storage.

References	Year	Title	Type of study and parameter	Key findings
[18]	2023	Technoeconomic Assessment of Various Configurations Photovoltaic Systems for Energy and Hydrogen Production	Economic benefits evaluation (i) PV system efficiency (ii) Hydrogen cost	Improvement of the PV system to produce hydrogen at competitive costs (2.1 cent/kWh and $2.53/kg-H2)
[19]	2022	Simultaneous analysis of hydrogen productivity and thermal efficiency of hydrogen production process using steam reforming via integrated process design and 3D CFD modelling	Experimental/numerical investigation (i) SMR thermal efficiency	H2 production and efficiency optimization for finding the best working condition via simulation in a SMR process
[20]	2023	Comparison of derivative-free optimization: energy optimization of steam methane reforming process	Numerical investigation (i) SMR thermal efficiency	Improvement of thermal efficiency through the use of DFO algorithms
[21]	2022	Impact of hydrogen on natural gas compositions to meet engine gas quality requirements	Gas grid evaluation (i) Wobbe index (ii) Hydrogen share	Maximum share of hydrogen (13.4%) that can be added to the current gas network to fulfil the EU regulations
[22]	2023	Modeling of catalyst poisoning during hydrogen production via methane steam and dry reforming	Numerical investigation (i) Catalyst poisoning (SMR)	Heterogeneous fixed-bed reactor model showed high sensitivity to sulphur poisoning, also with a very low concentration of H2S
[23]	2016	Solar water splitting by photovoltaic-electrolysis with a solar-to-hydrogen efficiency over 30%	Experimental investigation (i) Solar-to-hydrogen (STH) efficiency	Solar-to-hydrogen efficiency of about 30% reached on the laboratory scale

Table 2: Literature survey on hydrogen as fuel for ICEs.

References	Year	Title	Type of study and parameter	Key findings
[24]	2023	Conversion of a small size passenger car to hydrogen fueling: 0D/1D simulation of port vs. direct injection and boosting requirements	Numerical investigation (i) Engine performance (ii) Injection pressure effect	Increased pressure of injection improves the efficiency, the PFI layout requires much higher intake pressure boost than DI
[25]	2020	Lean combustion analysis using a corona discharge igniter in an optical engine fueled with methane and a hydrogen-methane blend	Experimental investigation (i) Blend composition (ii) Spark plug design	Hydrogen addition considerably stabilizes the combustion, thus extending the lean limit
[26]	2016	Analysis of combustion of methane and hydrogen–methane blends in small DI SI (direct injection spark ignition) engine using advanced diagnostics	Experimental investigation (i) Engine performance (ii) Flame area	The 2D imaging showed how the hydrogen addition makes the flame front faster of more homogeneous
[27]	2017	Effects of hydrogen direct injection strategy on characteristics of lean-burn hydrogen–gasoline engines	Experimental investigation (i) Thermal efficiency (ii) Lean burn strategies	Hydrogen shortened the flame development, increasing the thermal efficiency and mean effective pressure
[28]	2015	Influence of spark timing on the performance and emission characteristics of gasoline–hydrogen-blended high-speed spark-ignition engine	Experimental investigation (i) Ignition timing influence (ii) NOx and CO emissions	Spark timing turned out to be a key factor for the thermal efficiency, shifting the combustion towards the TDC
[29]	2021	Effect of negative valve overlap on combustion and emissions of CNG-fuelled HCCI engine with hydrogen addition	Experimental investigation (i) Valve timing effect (ii) NOx emissions	Symmetric NVO strategy can achieve the highest EGR rate easing the HRR and reducing the PRR, which improves the rough running of HCCI engine, preventing the occurrence of knocking
[30]	2022	Effects of gasoline and hydrogen blends on exhaust gas emissions and fuel consumption from gasoline internal combustion engines	Experimental investigation (i) Engine temperature influence (ii) Fuel consumption	Hydrogen improves the combustion quality by reducing the unburnt fractions, thus the possibility of explosive phenomena into the exhaust line
[31]	2019	Computational investigation of diesel injection strategies in hydrogen-diesel dual fuel engine	Numerical investigation (i) Spray model	The advance of diesel pilot injection results in a reduction of HC and CO and increase of NOx
[32]	2023	Hydrogen-diesel dual-fuel direct-injection (H2DDI) combustion under compression-ignition engine conditions	Experimental investigation (i) Spray model (ii) Hydrogen jet and diesel spray interaction	The H2 jet ignites after a period from the interaction with pilot fuel. The duration depends on the pilot SOI, the onset of jet interaction, and the charge temperature

Table 3: Literature survey on challenges of H2-ICEs.

References	Year	Title	Type of study and parameter	Key findings
[33]	2020	A numerical investigation on De-NOx technology and abnormal combustion control for a hydrogen engine with EGR system	Numerical investigation (i) EGR ratio	The EGR can reduce the in-cylinder hot spots, thereby suppressing the occurrence of preignition of the hydrogen engine
[34]	2023	Extending the knock limits of hydrogen DI ICE using water injection	Experimental/numerical investigation (i) Water injection (ii) Advanced ST CR variation	Water impacts autoignition via three potential routes: charge cooling effect, thermophysical effect, and a kinetic effect. The impact of charge cooling was dominant
[35]	2023	Comprehensive analysis on the effect of lube oil on particle emissions through gas exhaust measurement and chemical characterization of condensed exhaust from a DI SI engine fueled with hydrogen	Experimental investigation (i) Effect of lube oil (ii) Particles size and number	The presence of PAH, alkyl-PAHs, oxy-PAHs, and mineral oil in the exhausts suggests that the lubricating oil passed into the combustion chamber and degraded at high temperature
[36]	2017	Study on the NOx emissions mechanism of an HICE under high load	Numerical investigation (i) NOx formation routes	The thermal NO played a key role in the NO emission, contributing more than 75% of the total NO emission under high load
[37]	2020	Experimental study on ammonia/hydrogen/air combustion in spark ignition engine conditions	Experimental ammonia-hydrogen blend composition influence	A moderate amount of hydrogen can make the H2-NH3 mixture suitable for ICE operations
[38]	2021	Study on the mechanism of the ignition process of ammonia/hydrogen mixture under high-pressure direct-injection engine conditions	Numerical investigation (i) High-pressure DI influence (ii) Radical formation process	The addition of hydrogen can provide the concentration of H-free radical at the early stage of combustion, promoting the rapid generation of OH-free radical and thus accelerating the consumption rate of NH3

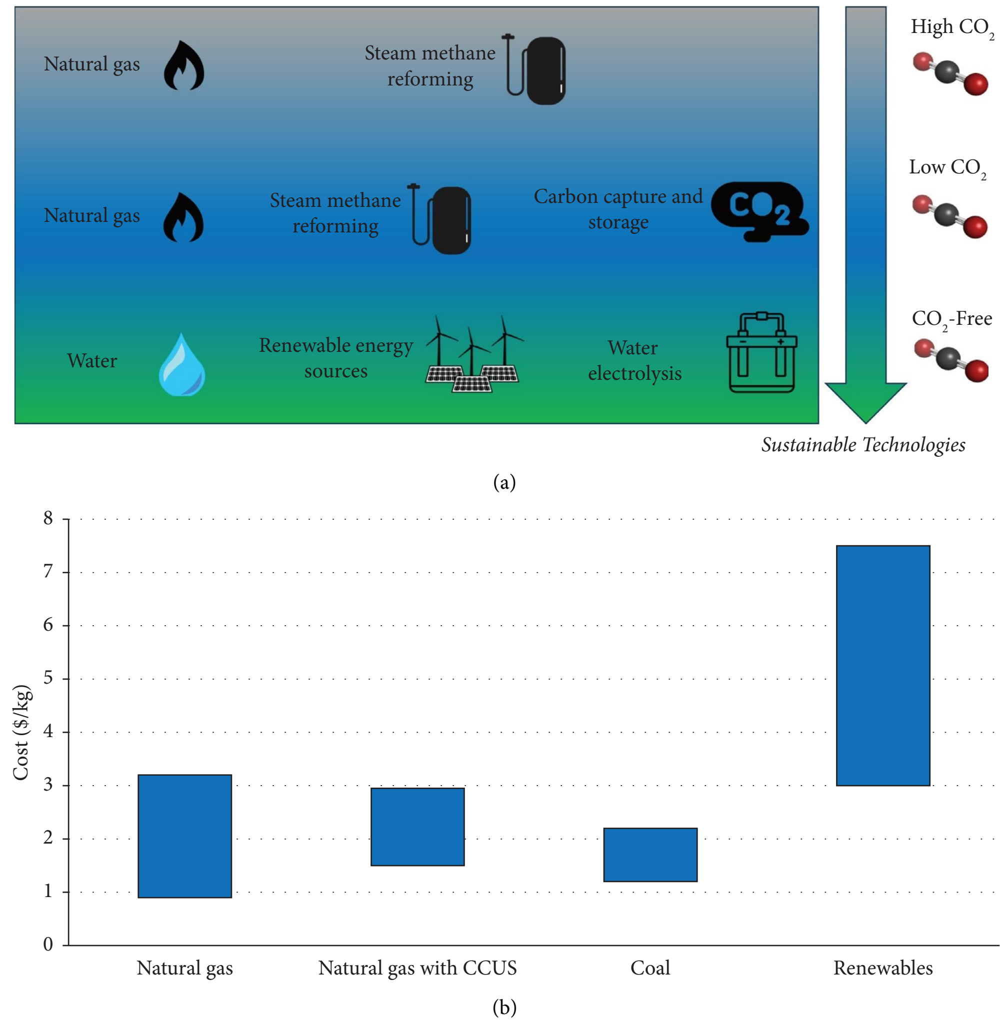

Figure 3: Hydrogen production pathways (a) and hydrogen production cost $/kg by energy source type (b). Adapted from [40].

In recent years, the situation has been rapidly evolving, and several studies have analysed how the current policies and future plans of investment will probably play a key role in making the hydrogen a sustainable energy carrier both from the technological and economic perspectives. Accordingly to these studies, hydrogen is already experiencing a significant reduction of cost compared to few years ago [18–42] and an important improvement in production efficiency [43]. Thus, the hydrogen can be a unique opportunity to accelerate the transition towards a low-carbon economy model, and the European Union (EU), in line with its efforts to move away from carbon-based fuels, could act as watershed. As far as the production methods, there are different opinions and studies that shed light on the necessity of higher investments for increasing the share of green hydrogen. Indeed, based on current literature, it appears clear that in the middle term, the fossil fuels will be the main energy source for the hydrogen production, then it is proper to provide a more detailed analysis of the processes involved and their efficiencies. Based on the CO2 capture and thermal efficiency, in [44], the authors assessed six sorption-enhanced SMR configurations. The article shows how the air-gas mixture to the calciner returns the highest efficiency (~77%), lowest cost (~£1.90 H2/kg), and a CO_2 capture efficiency (i.e., around of 60%), while on the other hand, the replacement of natural gas with biomass could effectively increase the latter to ~86% with a lifted cost of 2.16 $ H2/kg. Using oxy-natural gas or chemical looping to heat the calciner would result in an almost complete CO2 capture, even to a carbon-negative production by adding biomass (nevertheless, with highest costs). Given the importance of the calciner, several studies turned their attention to building SMR reactor models [19] used to simultaneously simulate the H2 production and while evaluating the thermal efficiency and their dependencies on the syngas composition and operating temperatures. A further key aspect in the production of hydrogen is its purity degree. In that sense, the potential presence of elements such as hydrocarbons, chloride, and sulphate could prevent its usability for the purpose for which it is intended [20, 21] (i.e., domestic heating applications or as fuel for both fuel cells and ICEs). More in detail, taking into account the SMR process, the presence of hydrogen sulphide can negatively affect the reactor by lowering its efficiency or leading to a partial deactivation of the same. Fabrik et al. in [22] developed a simplified fixed-bed reactor model and simulated the influence of the hydrogen sulphide on the nickel-alumina layer of the catalyst. The investigation, validated against experimental data present in literature, revealed that even a relative low concentration of H2S (~25 ppm) can influence the production process, leading to a relevant pressure drop, issue that can be avoided with a combination of lower feed flow rate and higher temperatures. Anyway, looking to the future, the likely spread of renewable energy sources will consequently bring to a less predictable energy production model. The intermittent nature of renewable energy sources forces to waste a part of what is produced or cover the shortages with fossil energy sources. To this end, the hydrogen production would lead to an enhanced flexibility to manage these peaks of production storing the exceeding energy [45]. As far as other pathways, one of the most promising techniques is the production of hydrogen via solar-powered electrolysis which could become commercially competitive in the near future. However, the current efficiency of these systems is still too low to make them attractive, where small-scale demonstrations showed high efficiency with peaks of around 30% [23], but with unsustainable costs for large-scale production.

3.2. Hydrogen: Transport and Storage. In view of the production processes described, albeit in a very simplified manner, it is also worth briefly discussing the main transport and storage methods of hydrogen. Currently, the hydrogen is produced and used on a local scale. The perspective that this energy carrier becomes more widespread involves the need for an efficient and cost-effectiveness distribution network. To this end, several investigations reveal how the number of hydrogen refuelling stations (HRSs) is increasing and between the 2015 and 2022, the total number of HRSs is almost quadrupled [46], rising from 214 to over 800 operating stations worldwide. Hydrogen is commonly delivered in the gas form as it requires relatively low pressures of compression (20–30 bar), thus making it suitable for pipeline transport. Not only that, the already existing global gas network has a considerable transport and storage capacity, and this is one of the main reasons why researchers are studying the feasibility of injecting hydrogen directly into existing infrastructures [47]. This approach would drastically reduce the startup costs while obtaining benefits from reduced GHG emissions, especially if the added hydrogen comes from renewable sources. On the other hand, the downsides are the many modifications needed to build checkpoints for controlling that the hydrogen concentration does not increase over the safety level. In addition, sampling points for domestic applications must be carefully controlled and standardised [48] due to the different properties of the hydrogen compared to the methane. In any case, regardless of the abovementioned considerations, nowadays, no experimental test has yet been performed (at least not on a large-scale scenario); then, any step towards the implementation of the hydrogen in the already existing infrastructures would first require extensive studies, monitoring, and tests on the feasibility and safety. In absence of pipelines and if it is needed a high volume, the hydrogen can be liquefied. Gaseous hydrogen gets liquefied by cooling it to below 20K; the problem in this case is that the process takes about 30% of the energy content of the hydrogen. Another key concern is also the issue related to the evaporation of liquefied hydrogen that leads to the "boil off" phenomena. For the abovementioned reasons, this method is poorly adopted, and its use is limited to the aerospace field for the production of fuel for space rockets [49].

Regarding the hydrogen storage, there are two main methods; the first is physical based and involves the use of high-pressure tanks for the gaseous state and cryogenic vessels for the liquid counterpart. The other techniques exploit the complex property of certain materials to store hydrogen, thus with the adsorption on the surface of solids or within solids with the absorption (Figure 4).

The most widespread solution used for automotive applications is the physical storage of gaseous hydrogen with "simple" compressed tanks or using cryogenic techniques so as to increase the density of the fuel. The latter types of tanks are capable of 350–700 bar, but compressed tanks are usually preferred to their cryogenic counterparts due to the high demand for technologically advanced cryogenic systems and related energy consumption. Looking at the structure of these devices, the external shell is commonly made of composites or carbon fiber while the inner line is instead made of high-molecular weight polymer to avoid hydrogen leaks and work as the hydrogen permeation barrier. The ends of the outer shell are normally reinforced using metal dome protections and inside the tank, a sensor for monitoring the temperature is normally provided. In Figure 5, a schematic representation of a compressed hydrogen fuel tank is presented [51].

4. H2 as Fuel for ICEs

4.1. Fundamentals. In this section, the author provides an overview of the main topics that have been treated concerning the use of hydrogen as fuel for the internal combustion engines. To this end, before proceeding with the description of the findings on the use of H2 in ICEs, it is useful to recall its physical properties (Table 4).

The high value of autoignition temperature, combined with the low stoichiometric fuel-air ratio, makes the hydrogen more suitable for operation in spark ignition engines (SI) than the compression ignition (CI) counterpart despite several studies have been performed in this regard. The flammability limits immediately suggest the possibility of using the hydrogen as fuel for lean or ultralean conditions, thus minimizing the only harmful emissions of NOx since it is carbon free. On the other hand, despite the almost tripled lower heating value (LHV) compared to CNG and gasoline, the very low-density results challenging for the design of storage systems and turbochargers capable of providing enough air flow rate at low speeds with high loads or during ultralean operations, thus limiting the output power discharged to the crankshaft. Moreover, the low ignition energy predisposes the hydrogen to preignition events, resulting

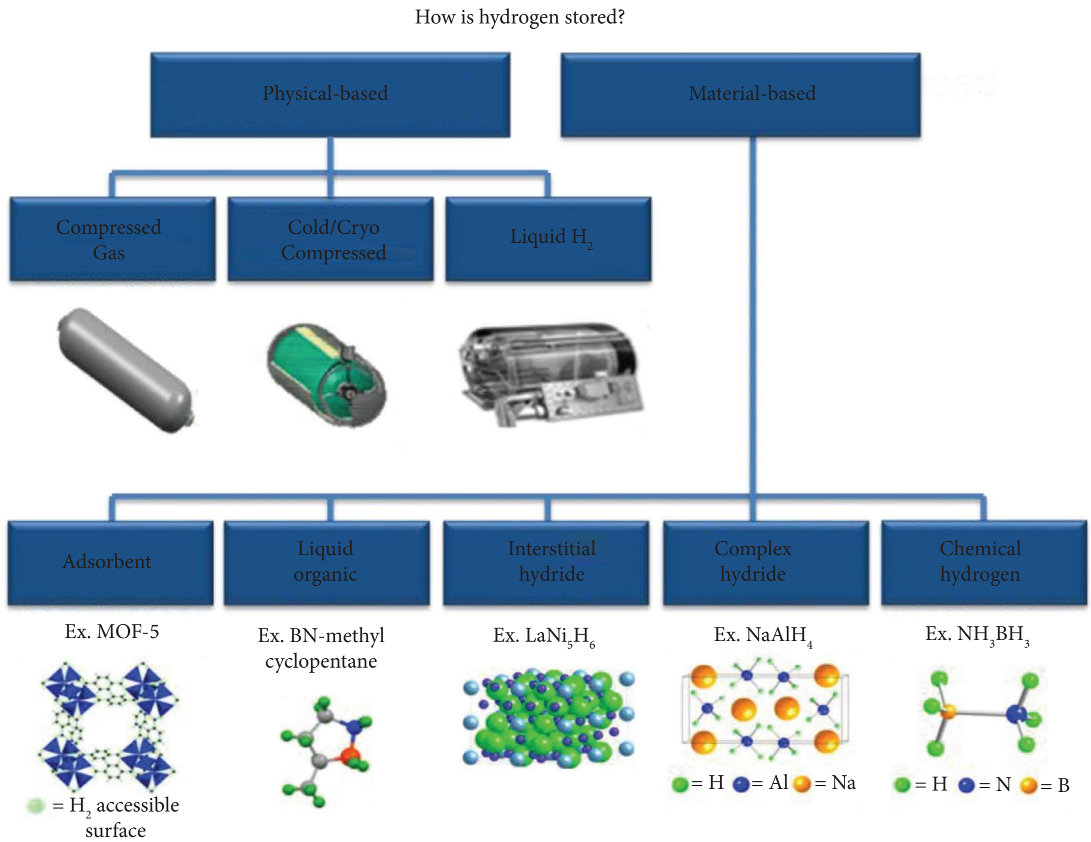

Figure 4: Hydrogen storage methods [50].

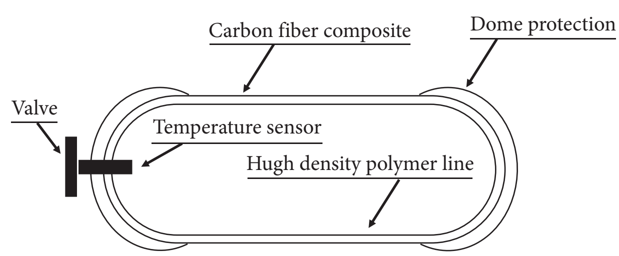

Figure 5: A schematic of a composite overwrapped pressure vessel.

Table 4: Hydrogen, CNG, gasoline, diesel, and ammonia fuel properties at 298K and 1 atm.

Property	Hydrogen	CNG	Gasoline	Diesel	Ammonia
Density (kg/m^3)	0.08	0.70–0.72	720–760	820–845	0.73
Flammability limits (volume % in air)	4–75	4.3–15	1.4–7.6	0.6–5.6	15–28
Flammability limits (ø)	0.1–7.1	0.4–1.6	0.7–4	—	—
Autoignition temperature in air (K)	858	723	550	527	914
Minimum ignition energy (mJ)	0.02	0.28	0.24	0.25	8
Flame velocity (m/s)	1.85	0.38	0.37–0.43	0.2–0.8	0.015
Adiabatic flame temperature (K)	2480	2214	2580	2600	2070
Quenching distance (mm)	0.64	2.1	~2	~2	—
Stoichiometric fuel/air mass ratio	0.029	0.069	0.068	0.068	0.165
Lower heating value (MJ/kg)	119.7	45.8	44.79	42.78	18.8

from the presence of hot spots (e.g., surface of the spark plug electrodes, exhaust valves, and carbon deposits). In addition, another aspect to consider is related to the reduced quenching distance which is addressed to be the main cause of the back-fire phenomenon, i.e., when the intake valves are still open and there is an uncontrolled propagation of the flame to the intake manifold. The latter can be limited with an appropriate strategy of mixture formation coupled with optimized cam valve timing or completely avoided employing the DI layout. Finally, it is noteworthy to note the high value of flame velocity of the hydrogen. The latter is one of the main reasons why several studies have investigated the use of dual-fuel combustion strategies to explore the influence of hydrogen addition to "balance" or to improve those "shortcomings" that confined the individual fuels to limited running conditions [52], mainly methane for the low flame speed, or to reduce the pollutant emissions [53] when talking about gasoline and diesel.

4.2. H2-ICEs Combustion Strategies

4.2.1. Pure H2 Spark Ignition. The mainstream application of the hydrogen in internal combustion engines (ICEs) is with spark ignited engines. Depending on the position of the injector, it is possible to distinguish between port fuel injection (PFI) and direct injection (DI) layout. The PFI is normally preferred thanks to the long time for mixture preparation as well as for the subsequent limited presence of air-fuel rich areas, thus reducing the NOx formation and knock tendency. On the other hand, due to the low density of the hydrogen, when injected into the intake manifold, it expands, partially replacing the air. Therefore, fresh air must be forced into the cylinder to compensate the reduction of volumetric efficiency. This task requires the use of a turbocharger unit capable to guarantee very high boost levels over the entire engine load map. This challenge can be partially overcome employing the DI system but with the drawbacks related to rising issues regarding the combustion anomalies, except the backfire phenomenon which regards only the PFI layout. Currently, several studies paid attention on both solutions and following benefits and challenges. Irimescu et al. in [24] performed a numeric analysis with a 0D/1D model previously calibrated based on a small size spark ignition. The model involved the use of dedicated flame laminar speed pattern derived from experimental evidence while the knock tendency was taken into account with a simplified approach (i.e., the knock induction integral method). The goal of the work was to compare the output performance among the use of conventional gasoline and H2 with both injection layouts. It was observed (Figure 6) that the use of hydrogen returned a comparable value of IMEP over all the engine full-load map when operating in the DI mode with a maximum gain of about 6% in terms of efficiency while the H2-PFI resulted in a loss of performance when the engine was in a full load at low speeds.

The intake pressure increase needed with the PFI system was more than 30% compared to the gasoline baseline, while the DI only required an increase of 8%. Nevertheless, the DI system has its disadvantages; in fact, the injector characteristics are very demanding, with a required pressure ranging from 50 to 100 bar to cover all the operating conditions, not to mention the drawback in terms of nonretrofit (i.e., due to the very low density of hydrogen, these types of injectors usually are very voluminous, thus requiring the redesign of engine head).

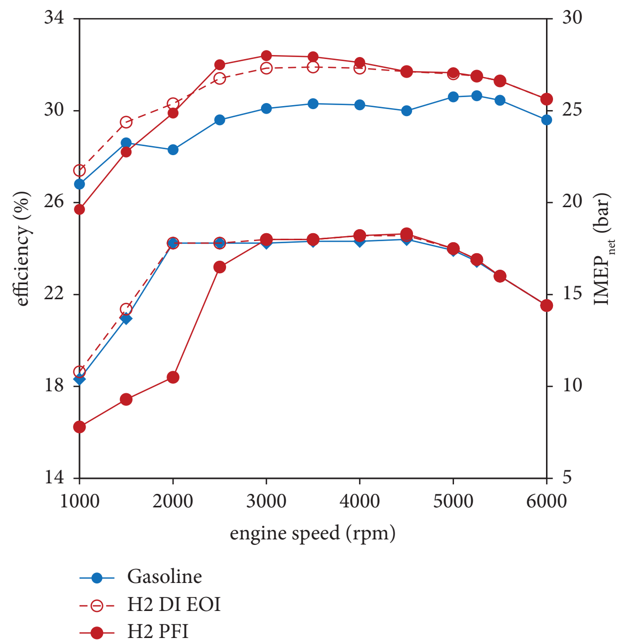

Figure 6: Full-load brake efficiency and IMEP throughout the engine speed range in PFI and DI configuration with 2.5 mm injector nozzle and 5 bar injection pressure. Adapted from [24].

4.2.2. Dual-Fuel Combustion with Spark Ignition. Among the different fuels used for the automotive field, methane is definitely the leading candidate for enriched operations with hydrogen [54]. Its low cost, low carbon content, and easy application in conventional SI engines make it very attractive as a cost-competitive solution. Nevertheless, despite the excellent knock resistance and proper fuel-air ratio for extended combustion characteristics for lean operations, the main "weakness" of the methane is its low flame speed. The latter limits operations under lean-operating conditions due to the considerable increase in combustion duration, thus resulting in a decrease of output power of approximately 5–10% [52]. The abovementioned premises prompted to study the influence of hydrogen addition on flame speed characteristics (Figure 7).

The literature extensively covers this topic with several studies carried out. For example, many studies have focused on the influence of hydrogen addition in the early stages of the combustion process, which is addressed to be one of the main contributors to cyclic variation. In [25], the authors examined the first phase of the combustion process, evaluating the effect of a conventional spark plug and a corona discharge igniter on the flame development under lean operations in an optically accessible engine. The study underlined the overall advantages of hydrogen addition to the methane, showing a reduced cycle-to-cycle variability with limited losses of IMEP at high loads. Moreover, the corona ignition system made it possible to further lower the lean limit of the mixture, showing a high-probability presence of a stable and fully developed flame kernel from the first stages of the combustion process following the ignition event. A comparison among standard gasoline fuel,

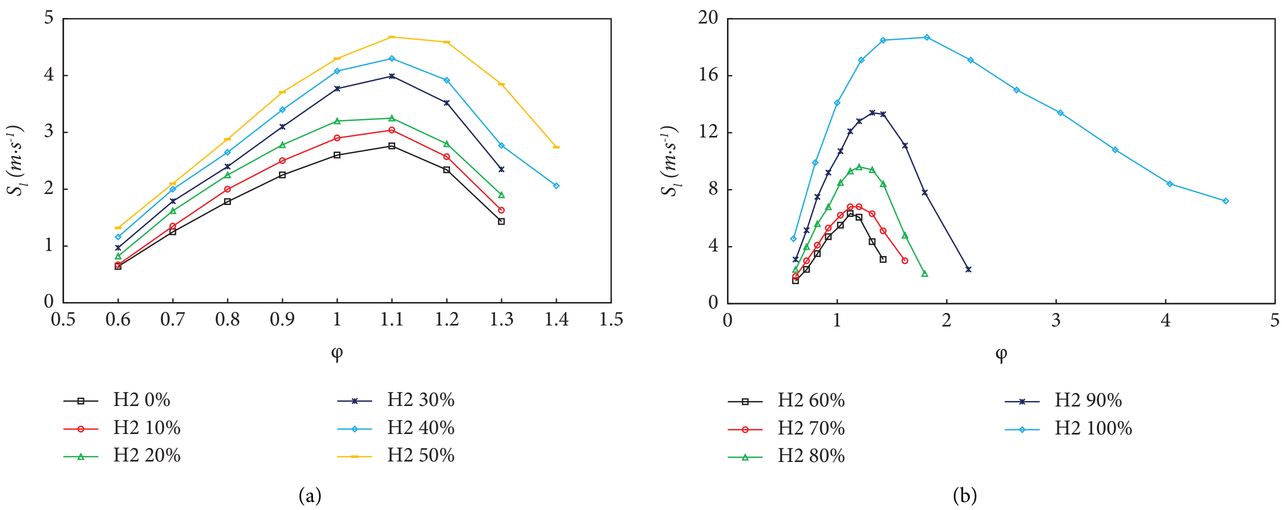

Figure 7: Flame velocity for hydrogen-methane-air mixtures on the equivalence ratio φ under various hydrogen concentrations: hydrogen concentrations 0–50% vol. (a) and hydrogen concentrations 60–100% vol. (b). Adapted from [54].

methane, and methane-hydrogen blends (i.e., M20H2 and M40H2 with a hydrogen content by volume of 20 and 40%, respectively) has been carried out in [26]. The authors used a transparent single-cylinder small size engine to characterize the fuel jet morphology. The chemiluminescence of the imaging data highlighted how the hydrogen strongly influences the reactivity of the mixture, allowing for a more homogeneous propagation of the flame front in all directions, therefore, improving the thermal efficiency. In addition, the high diffusivity of the hydrogen reflected in an enhanced distribution of the fresh charge, thus in a reduction of pollutant emissions (Figure 8), compared to pure methane and gasoline operations; in fact, the latter were both characterized by the presence of rich and lean regions that reflected in an irregular flame front progression.

However, there are also a large number of studies in the literature that have thoroughly investigated the benefits of adding hydrogen to both gasoline and diesel. In these studies, the main aim was not only the performances but also the carbon particles emissions reduction and potential fuel savings. In [27], the authors demonstrated how a small amount of hydrogen added to gasoline can bring the engine to stable operation under lean-burn conditions (AFRrel~1 0.5) with the COV reduced to below 1%. More in detail, keeping the hydrogen content to 10% in volume fraction, it was possible to increase the mean effective pressure and thermal efficiency by 10% and 4.0–4.5%, respectively. Other studies evaluated the effect of the typical engine combustion-controlling factors such as spark ignition timing and valve timing [28, 29] on the extension of the optimal operating range of hydrogen-gasoline blends. The spark timing plays a key role in advancing the combustion process close to the TDC and then improving the thermal efficiency and fuel consumption, while most of the investigations on the valve timing clearly showed the benefits of anticipating the intake valve opening (IVO) for medium- and high-load conditions in order to increase the volumetric efficiency, while at low load, this effect results quite limited. Anyway, another aspect to be taken into account is the possible presence of unburnt fractions of the fresh charge into the exhaust line that can lead to the occurrence of explosive phenomena. In fact, in its pure state, the hydrogen has a lower limit of explosion (LEL) of 4% by volume compared to the 1.2% of gasoline. Therefore, it is of paramount importance to use the best mixture formation strategy to ensure a clean combustion, and this is also another reason for which the PFI layout is normally preferred to the DI [30]. Then, it is noteworthy to underline the development of increasingly accurate simulation software which is able to return results faithful to experimental tests, thus representing valid tools to speed up the research and saving costs compared to the expensive test campaigns necessary to gather data. In that sense, it is possible to distinguish two main categories of works; the first in which the authors investigated the effect of adding hydrogen through the use of whole engine simulation models, comprehensive of the entire intake and exhaust line, developed in 0D/1D environment [55] or those which involve the use of computational fluid dynamic (CFD) codes mainly aimed to understanding the phenomena underlying the pollutant emissions mechanism [56] or the combustion chamber geometry influence on the mixture formation process [57].

4.2.3. Dual-Fuel Combustion with Diesel Pilot Ignition. Due to the high autoignition temperature of hydrogen, its implementation in compression ignition (CI) engines results quite challenging; thus, the addition of diesel turns out to be a key factor thanks to its high reactivity. Although most of the studies carried out highlight the benefits related to the reduction of carbon emissions, the increase in NOx emissions at medium-high load conditions still remains the main challenge to overcome. In view of this, several studies focused on the interaction between the diesel jet and the premixed charge of air hydrogen (PCCI). They demonstrated how the injection timing plays a key role in reducing

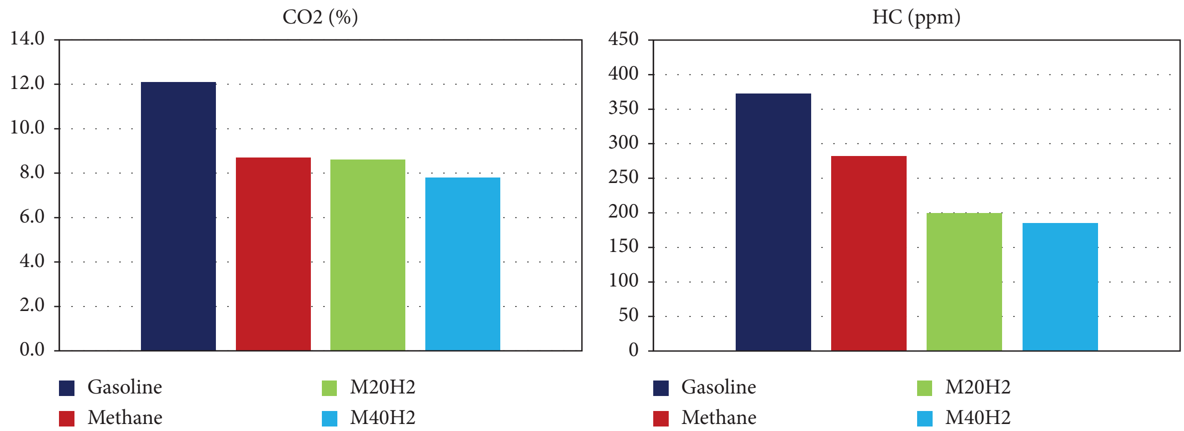

Figure 8: CO_2 and HC emissions for all the tested fuels at 2000 rpm full load. Adapted from [25].

emissions of HC and CO when anticipating the SOI thanks to the lower temperature inside the combustion chamber but on the other hand, increasing the peak pressure rise rate and thus the NOx [31]. Another approach is given by the use of two injectors, both inside the combustion chamber, so as to have a hydrogen-diesel dual-fuel direct injection (H2DDI). In this case, the focus fall on the interactions between the two jets, with detailed analysis on the factors that lead to the combustion of hydrogen and related chemical interactions [32]. Anyway, the latter is a novel approach which availability in literature is still limited.

Moving to more valuable considerations about the performance given by the use of the dual-fuel combustion with the PCCI technique, Ali et al. in [58] investigated the combustion performance of a common rail diesel engine modified to run with methane and hydrogen mixture under different load conditions and with different compression ratios. The paper compared the results with the engine fuelled with diesel, pure methane, and hydrogen-methane mixtures with a content of the first of 5 and 10% in volume fraction. For all the abovementioned conditions, the diesel was used as pilot fuel and the engine crankshaft velocity was kept at 1750 rpm. The study showed an overall enhancement of the BTE (Figure 9) and a reduction of the brake-specific energy consumption (BSEC) of the dual-fuel mixture on the pure methane-fuelling mode over all load conditions.

On the other hand, as a consequence of the lower density, it was found to be difficult to ignite the fuel-air mixture at low loads with subsequent repercussions on the combustion quality and thus a dramatical drop of around 40% of the performance compared to the pure diesel-fuelling mode. The situation results to be partially improved by increasing the load; therefore, with an appropriate spark timing, it was possible to concentrate the pressure peak close to the top dead centre (TDC) and then increasing the thermal efficiency and reducing the above gap to 24%. Moreover, it was observed how the combustion noise was reduced during dual-fuel operations, especially with high CR and high load conditions.

5. Challenges of H2-ICEs

5.1. Combustion Anomalies. In this section, more attention is given to the application of hydrogen as the sole fuel in internal combustion engines, focusing on the main criticisms that, as matter of fact, characterize and represent the main challenges with which to deal when talking about H2-ICEs, the combustion anomalies. These are caused by the same properties that make the hydrogen such an attractive fuel for ICEs. In particular, as already anticipated in the "Fundamentals" section, the wide flammability limits associated with the low energy requirements can result in the rising of undesired phenomena such backfire, knock, and preignition. These combustion anomalies are further discussed in the following.

5.1.1. Preignition. The preignition is the phenomenon whereby the mixture ignites when valves are closed before spark plug ignition; this leads to an increase in pressure during the compression stroke with a consequent loss of performance (Figure 10). The causes underlying the preignition cannot be addressed to a single source and many studies offer different conclusions. Due to its stochastic nature, the investigations already present in literature of the abovementioned phenomenon usually led to different findings; from the spark plug and exhaust valves-related hot spots to the hot oil particles from previous combustion cycles [33–59]. The dependence of this combustion anomaly on the minimum autoignition energy makes it to rise in frequency when the hydrogen-air mixture is close to the stoichiometric condition. Furthermore, also, operating conditions at high speeds and high loads are prone to the occurrence of the preignition due to the increase of the pressure and gas temperatures.

Noteworthy is the repetitive aspect of the preignition for which, once occurred, if not immediately interrupted, it continues for several cycles continuously anticipating and then leading to the occurrence of the backfire phenomena. In this case, the combustion anomaly repeats cycle after cycle,

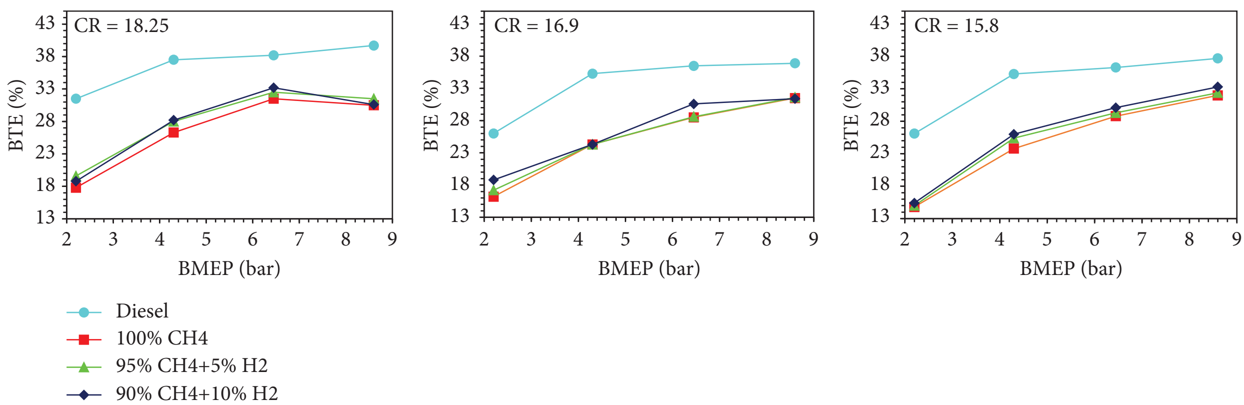

Figure 9: BTE at different loads and CRs ($n = 1750$ rpm). Adapted from [58].

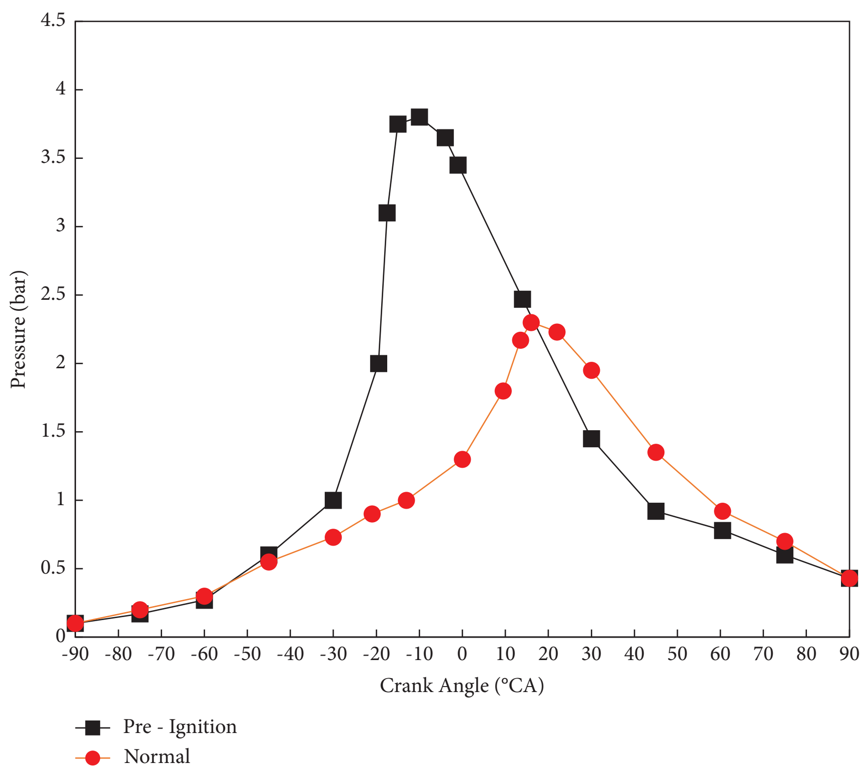

Figure 10: In-cylinder pressure comparison for a normal combustion and the one affected by preignition. Adapted from [33].

reducing the in-cylinder pressure and shifting the auto-ignition event to the initial stage of the compression stroke when the intake valves are still open, thus causing the occurrence of backfire phenomenon with a drastic increase of the pressure inside the intake manifold. Measures to avoid or limit the preignition involve the use of proper designed spark plugs and ignition systems with low residual charge. Hydrogen late injection into the combustion chamber (i.e., DI layout) is another solution to avoid the occurrence of the preignition.

5.1.2. Backfire. Backfire is the phenomenon for which the fresh charge of hydrogen-air gets ignited during the intake stroke in the combustion chamber or in the intake manifold. The backfire, as for as the preignition, owes its causes in the presence of hot spots, gas residuals from precedent cycles, or remaining charge in the ignitions system. The main difference between these two combustion anomalies is that the preignition takes place in the combustion chamber when the valves are closed, while the backfire occurs when the intake valves are still open. This results in a flame front propagation along the intake line, which is not clearly audible but can seriously damage or destroy the intake manifold. Nowadays, several strategies with hydrogen include the use of the direct injection, thus completely avoiding the occurrence of this phenomenon; therefore, the abovementioned combustion anomaly is generally limited to the port fuel injection layout. In Figure 11, it is possible to see how the backfire results into an important increase of intake manifold pressure, unlike what happens with preignition. Therefore, the fresh charge is unable to give any contribute to the engine crankshaft, thus arriving already exhausted to the combustion chamber with consequent negative indicated mean effective pressure (IMEP). A single cycle affected by this combustion anomaly can lead to the shutoff of the engine.

Different approaches to avoid the backfire phenomena have been studied [60], and most of them point the start of injection and valve timing as key factors. In this sense, for the hydrogen injection, the literature suggests using strategies for which the fuel concentration around the intake valves is reduced (i.e., homogeneous charge strategies, but in contrast with the considerations on the preignition). In addition, the intake efficiency can be improved by increasing the pressure of injection, thus shortening the duration of the injection event which results in a relevant enhancement of the power output at high speed. On the other hand, a correct intake valve timing that allows the entry of pure air into the combustion chamber, before aspirating the fuel, can cool the charge and potential hot spots. Anyway, other studies suggested ending the injection in a fixed range that is anticipated for low speeds and rich mixtures, while an opposite strategy should be approached at high speeds.

5.1.3. Knock. By knock, it is meant as the achievement of conditions, in terms of pressure and temperature, by the end gases such as them spontaneously autoignite; therefore, releasing a huge amount of heat in a short period of time, producing pressure waves that hitting the cylinder wall generate the characteristic metallic sound from which the phenomenon takes its name (Figure 12). The knock can cause engine damage as it increases the mechanical and thermal stress on the combustion chamber's components. The resistance of fuels to the knock tendency is defined through the octane rating; then, depending on the method, it will result in the research octane number (RON) or the motor octane number (MON). A cooperative fuel research (CFR) engine is used for the abovementioned methods by comparing the fuel knock resistance to that of a standardized mixture of heptane and iso-octane. Although these methods were designed for liquid fuels, with the growing interest in hydrogen as fuel for ICEs, the first investigations on its knock resistance started to appear in literature. The reported RON values range from less than 90 to more than 130 [61, 62], depending on the hydrogen-air ratio, temperature, and other parameters. In [61], the authors performed an experimental and numerical investigation on the hydrogen knock resistance with the ASTM's RON method. The results of the above work showed that the hydrogen has a significantly higher knock resistance than standard gasoline when providing similar energy to the mixture, but on the other hand, the ASTM standards resulted to be poorly suited for the analysis of richest conditions. In fact, the high flame laminar speed of the hydrogen resulted in an immediate rise of the pressure inside the combustion chamber, which can be mistakenly identified as knock occurrence. In this context, it is noteworthy to mention the existence of the methane number (MN) that is used to characterize the knock resistance of gaseous fuels. Unfortunately, this method uses the methane as the fuel of reference with a MN of 100 and hydrogen with a MN of 0 per definition [63]. Obviously, this is a clear contradiction with respect to the above statement, thus making the latter method unsuitable for the hydrogen knock resistance evaluation.

Anyway, the knock has been deeply investigated and several studies evaluated the main operating parameters that contribute to the achievement of the conditions for which the abnormal combustion occurs. In [64], the authors performed an experimental study on a multicylinder spark ignition engine fuelled with hydrogen for analysing the effect of knocking and how the latter was correlated with the backfire. Based on the abovementioned study, the backfire increased in frequency when knocking occurred in earlier cycle, mainly due to the increased temperature of the intake valves. Joel et al. carried out an analysis on the water injection strategy so as to extend the knock limits on a heavy-duty hydrogen-fuelled engine [34]. The work highlighted how the addition of water affects the knock limits via three potential routes, i.e., charge cooling effect, thermophysical effect, and kinetic effect. From the first to the following two with a progressively decreased contribution, the water injection was experimentally observed to significantly extend the knock limits, thus increasing the power output and bringing it on a comparable value to the diesel with a limited loss of efficiency by almost 1%. Anyway, the phenomena led to the development of different models, but as matter of fact, most of the available works are based on the approach proposed by Livengood and Wu in [65], where the self-ignition occurs when the integral time between the intake valve closing and knock of the inverse of the induction time of the mixture reaches the unit. However, this approach relies on phenomenological formulations intended for liquid fuels. Therefore, it is not rare to find studies in which the authors followed alternative paths. For instance, Millo et al. proposed a numeric approach to evaluate the self-ignition time using a 0D chemical reactor simulation developed in CONVERGE environment [66]. The time needed to detect an increase of 400 K from its initial value was defined as the induction time.

5.1.4. NOx Emissions. When considering the pure hydrogen-running mode, thanks to the absence of carbon particles, the regulation about the harmful emissions should be theoretically limited to only nitrogen oxides. Nevertheless, albeit the hydrogen is carbon free, there is always a small amount of carbon particles coming from the wears of the gaskets and lubricant oil. In [35], the effect of lubricant oil on the particle emissions has been investigated using a hydrogen-fuelled spark ignition engine. The study highlighted the role of the temperature in the entertaining part of the lubricant oil into the combustion chamber, thus undergoing a process of oxidation and dissociation. Therefore, it is intuitable that when talking about H2-ICEs, it is an oversimplification of the statement for which these power units contribute to generate polluting emissions only for the

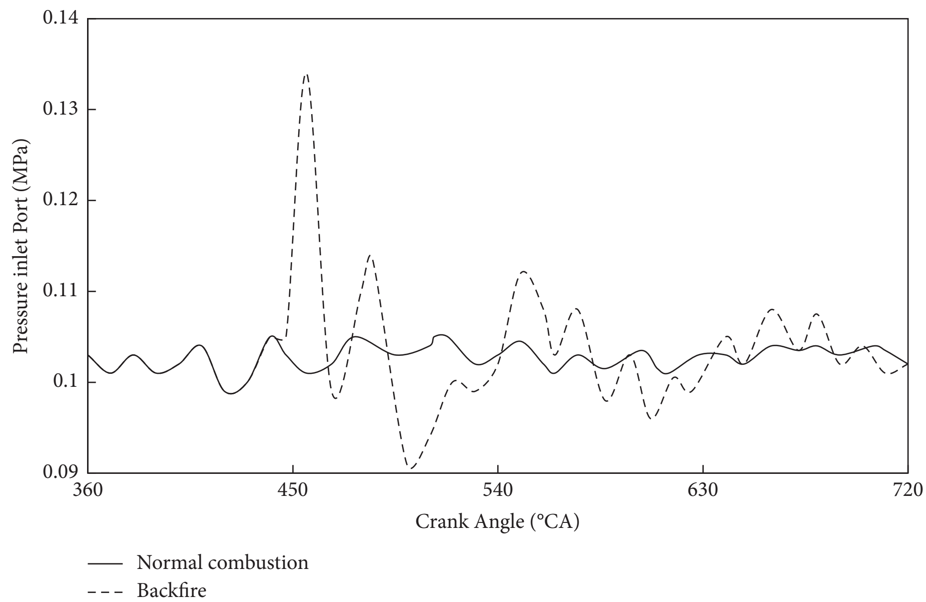

Figure 11: Pressure fluctuations in the inlet with and without backfire. Adapted from [60].

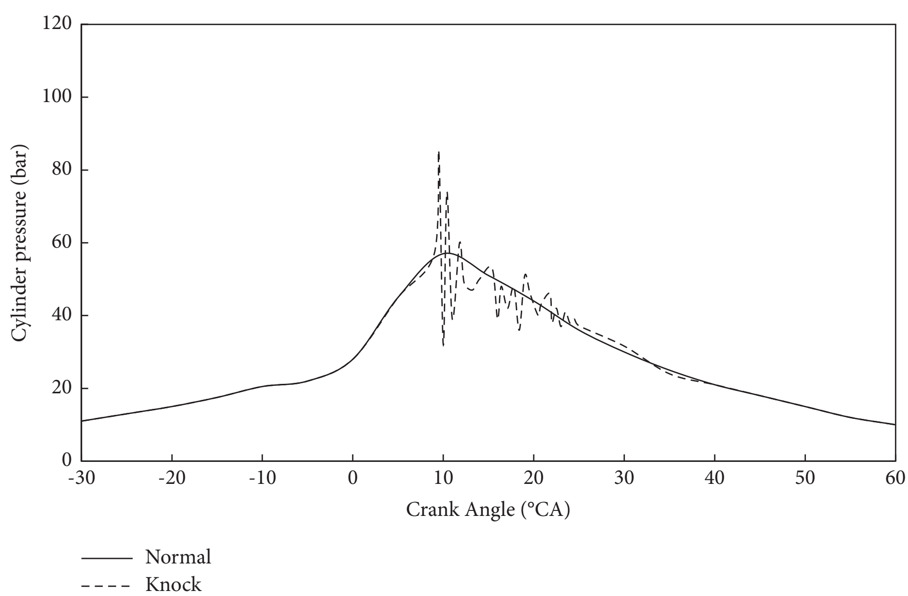

Figure 12: Comparison of two cycles whereby is highlighted the typical effect of knock occurrence on in-cylinder pressure.

nitrogen oxides. Anyway, several studies deeply examined the mechanism underlying the formation process of the NOx, how the latter is influenced by the main engine control parameters, and which strategies can be used to reduce them, e.g., exhaust gas recirculation (EGR) and water injection.

Vipin D. and K.A. Subramanian performed an experimental study on a multicylinder spark ignition engine fuelled with hydrogen [67]. In this work, the authors compared EGR and water injection strategies to observe the effect on NOx emissions. Both the increase of the EGR rate and the water-hydrogen ratio were found to have a significant role on the NOx emissions with the main difference that the first one was limited to 25% so as to not affect the engine performance, then resulting in a maximum reduction of the emissions of 57% with respect to the running conditions without recirculation. On the other hand, the water-hydrogen ratio returned an almost complete reduction of the NOx, up to 97% compared to the baseline condition without affecting the performance in a relevant way.

Other studies focused on the development of the detailed model to simulate and reproduce the mechanisms that bring to the increase in NOx emissions [68]. In this sense, the 3D-CFD code simulation returned to be an effective tool to obtain accurate results about the main mechanism behind the NOx formation process, shedding light on the role of the flame front temperature on the decomposition of NO and

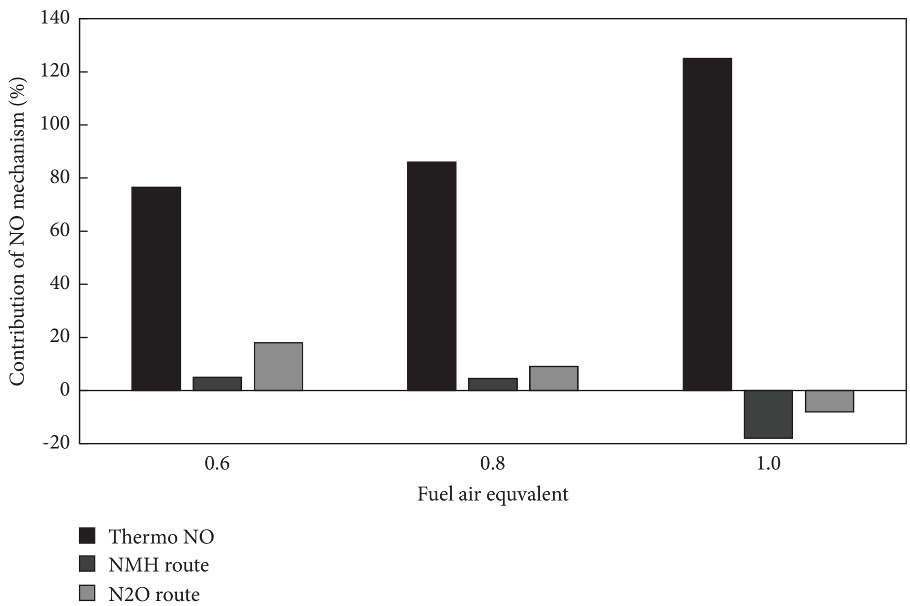

Figure 13: The contribution rate of the three reactions on NO under various fuel air equivalent ratios. Adapted from [36].

the variability of its composition with the load [36]. More in detail, the thermos-NO, NNH, and N2O routes contribute to the total NOx emissions in a different way depending on the load condition but with a constant predominant role of the thermal component, further highlighted at high loads, with a niche effect of the other two. In addition, with increased temperature, the backward reaction rate constants of NNH and N2O routes rise so much that the NO starts decomposing faster than the formation process, thus giving a negative contribute to the NOx production rate (Figure 13).

5.2. Advanced H2 Combustion Strategies. In recent years, several advanced strategies of combustion have been investigated in relation to the use of hydrogen into internal combustion engines. For example, in the so-called plume ignition concept (PCC), the plume tail of hydrogen jet gets ignited immediately after the end of injection. The combustion of this local rich mixture, depending on the global value of lambda, can lead to an important reduction of the NOx and unburned fraction of hydrogen, while keeping the thermal efficiency and performance of comparable value of conventional strategies. On the other hand, this technique involves the use of a high-pressure injector capable of very high flow rate to shorten the injection duration enough to adapt to the entire engine map. Not only that, talking about alternative strategies, in the present literature, there is also an increasing number of studies carried out on the use of ammonia as fuel with hydrogen. The use of H2-NH3 blends mainly focus on overcoming the issues related to the nitrogen-based pollutant emission and the bad combustion properties mainly related to the ammonia low laminar burning velocity. Thus, a small addition in hydrogen allows performance and stability improvement, resulting to be beneficial for the early stages of combustion process where the low reactivity of the ammonia usually limits its use in pure fuelling mode [37]. More in detail, it was observed that the addition of hydrogen provides the concentration of H-free radicals in the first phase of combustion necessary to promote the generation of OH free radicals, thus accelerating the consumption rate of NH3 [38]. Another strategy to tackle down the well-known hurdle regarding the knocking occurrence is the use of low temperature combustion (LTC) techniques. The latter normally refer to the employment of delayed pilot fuel injection timing, the use of water injection, or the reduction of the compression ratio. Except the latter that depends on the design of the power unit, the other two strategies have been thoroughly studied and proved to have a beneficial effect, increasing the amount of hydrogen energy shared thanks to the reduced reaction rate between fuel and air. On the other hand, these strategies cannot be exploited during high load operation, and if the engine is running in the dual-fuel mode, it must be considered, against the reduction of NOx, the drawback linked to the increase of HC and CO emissions [69].

6. Summary

In this review, the hydrogen as fuel and energy carrier is discussed with particular attention to its application for internal combustion engines. In view of what has been reported in the various sections abovementioned, hydrogen appears to be a viable solution as an intermediate step towards the net zero emissions goal, both as alternative fuel for the road transport sector and also as an energy carrier to face the future energy demand. Its use in ICEs is already a subject of different studies that have demonstrated its benefits such as improved thermal efficiency with comparable performance to conventional fuelling modes and the (almost) absence of carbon particle emissions. The unique properties of hydrogen make it an attractive and challenging fuel for pure running conditions and dual-fuel strategies, where it was observed to increase the flame laminar speed, thus extending the optimal range of working conditions of conventional fuels, especially of methane. Nevertheless, since its production stage, there are several issues and criticisms that still limit its widespread, from the strong dependence on fossil fuels (i.e., the steam methane-reforming technique is widely the most used to produce hydrogen) to the combustion anomalies that involves the use of complex combustion/mixture strategies to avoid engine damage or loss in performance. In particular, among the well-known properties of the hydrogen, the low density is probably the most critical one that makes necessary the design of specific storage systems able to guarantee satisfactory energy content to ensure a range comparable to that of conventional vehicles, while complying with all the safety standards required by technical regulations. Anyway, the renewed interest that this element, fuel, and energy carrier, is undergoing, combined with the development of ever more accurate and effective simulation software, is leading towards a constant update of its the state of art. There are more and more studies suggesting the future on road application of this technology, especially in the heavy-duty sector where the shortcomings of BEVs place hydrogen into a key role position.

Data Availability

No data were used to support the findings of this study.

Conflicts of Interest

The author declares that there are no conflicts of interest.

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