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A review on alternative fuels in future energy system

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ABSTRACT

Transition and decarbonization of the energy sector require the utilisation of new technologies and energy sources. Higher penetration of intermittent renewable energy sources implies the installation of energy storage, to store electricity excess and enhanced system efficiency. These electricity surpluses that will occur more often in the future energy system could be effectively utilized for the production of alternative fuels. Most of the alternative fuels that are considered for future applications are already known chemicals or products, nowadays used for other purposes. Another great advantage of some alternative fuels lies in their possibilities to act as an energy carrier. This feature might be crucial while discussing their utilisation potential and further development. Fuels which can simultaneously be used for power generation and as an energy carrier will have a more important role in the future and are likely to be utilized on a greater scale. Renewable energy source like biomass, on the other hand, is already widely used, and their role in the future system is not questionable. Even though significant increment in biomass consumption raises serious concerns about its sustainability, and seeks for new approaches. In this work, the authors tried to review alternative fuel characteristics, alongside their utilisation and production opportunities. To come up with the optimal solutions, the authors compared various proposed alternative fuels, alongside their advantages and drawbacks with an aim to find the most appropriate role for each fuel.

1. Introduction

The transition toward a 100% Renewable Energy System is a complex process with different technical and economic challenges. In order to achieve predetermined goals, several steps should be carried out simultaneously, including increment of energy efficiency, savings in primary energy consumption, and finally, deployment of variable renewable energy sources (VRES) [1]. A high share of intermittent renewables like wind and solar in the electricity mix consequently affects the grid stability and requires the flexible operation of conventional, baseload power plants [2]. Moreover, a higher share of VRES indicates that the periods with an excess or lack of electricity production will be more often; therefore, it is necessary to include short- and long-term energy storage [3]. Fig. 1 illustrates the penetration of VRES into the power system for the case of the European Union (EU28). It is known that about 30% of VRES can be balanced by the grid. Up to 80% of VRES can be integrated using demand response technologies like vehicle-to-gird (V2G), thermal storages, and other types of short-term storage. To integrate 100% of VRES, long-term energy storages are a necessity. Hydropower and biomass are renewable energy sources, suitable for flexible operation in a decarbonized energy system. Nevertheless, these resources may be scarce in some countries or geographical regions, and even more, their over-usage to fill the remaining gap of 20% may be unsustainable [4]. Lately, the chemical conversion of electricity surplus into some form of alternative fuel (Power-to-X) is introduced as a promising solution since they can act as an energy source or carrier, but also as long term energy storage [5].

Alternative fuels may vary by its origin and production process, but the common for all of them is that they are produced through the sustainable and clean procedure, without the additional emissions of Carbon dioxide (CO_2) [6]. There are two main pathways for the synthesis of alternative fuels: direct utilisation of electricity surplus and thermochemical conversion of raw feedstock. For the former one, the term electrofuels has lately been introduced to clearly emphasize the production route and usage of electricity [7]. Electrofuels are carbon-neutral fuels synthesized from the VRES electricity surplus in a

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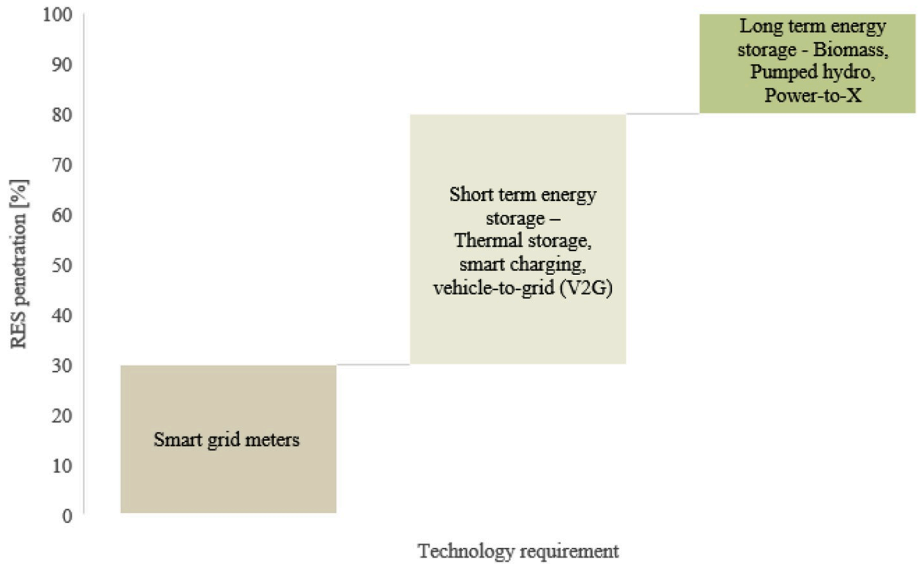

Fig. 1. Integration of Renewable Energy Sources into Electrical Grid [4].

gas, or liquid form, and carbon neutrality is achieved by closing the loop in a way that used CO_2 is captured from the exhaust gases or directly from the air [8]. In addition, electrolysis which is a crucial technology for the synthesis of electrofuels can be operated in a flexible mode in accordance with the production of the renewables, increasing the overall system efficiency and simultaneously allowing higher penetration of VRES [9]. The basic synthesis components of electrofuels are Hydrogen (H_2) and CO_2; therefore production targets are synthetic hydrocarbon gases like methane (CH_4) or butane, or in liquid form alcohol fuels like methanol (CH_3OH) [10]. Another, aforementioned, pathway for the synthesis of alternative fuels is through the thermochemical conversion of a raw feedstock into useful gaseous or liquid fuels [11]. These processes are widely investigated nowadays since they can convert different waste materials or raw feedstock into valuable alternative fuels or chemicals. The main challenge for broader application of thermo-chemical conversion is to couple synthesis process with VRES and lower the production costs. On the other hand, the main advantage of alternative fuels is derived from the fact that once produced; they can easily be stored and distributed where needed [12]. Fig. 2 presents potential pathways for the clean synthesis and utilisation of alternative fuels in future energy systems.

Alternative fuels can be synthesized in a liquid, gaseous or solid

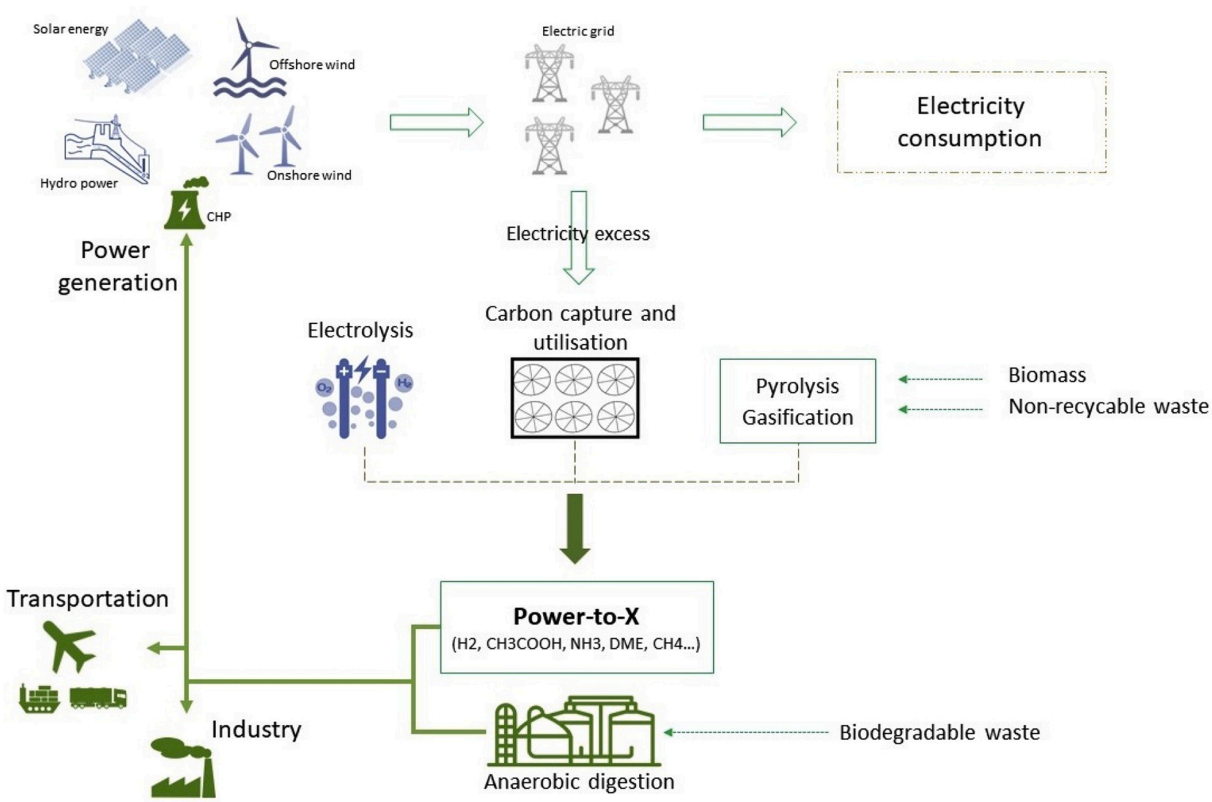

Fig. 2. Production pathway for Alternative Fuels synthesis using VRES.

Table 1
Recent review papers on various alternative fuels.

Type of review	Authors	Content
Electrofuels	McDonagh et al. [18]	• Production of electrofuels using curtailed energy from VRES
	Lehtveer et al. [19]	• Higher penetration of VRES might not be sufficient enough to achieve cost-competitiveness
Hydrogen	Abdalla et al. [20]	• Production, transportation, storage and application challenges
	Parra et al. [21]	• Role of hydrogen for deep system decarbonization
Ammonia	Giddey et al. [23]	• Sustainable synthesis and transport application
	Valera-Medina et al. [22]	• Highlights of previous research regarding utilisation of Ammonia as a viable energy vector for power applications
Biodiesel/Biomass	Chandra Bhan, Lata Verma, and Jiwan Singh [32]	• Review on alternative biofuels
	Bajwa et al. [14]	• Review on solid densified biomass products
	Perkins et al. [25]	• Fast pyrolysis for the production of liquid biofuels
	Widjaya et al. [26]	• Biomass gasification
	Sher et al. [33]	• Thermal and kinetic analysis of six different biomass fuels for power generation
Alcohol derived fuels	Verhelst et al. [34]	• Methanol as an IC engine fuel
	Svanberg et al. [28]	• Methanol for shipping
	Çelebi and Ayday [27]	• Review on light alcohol fuels
	Awad et al. [35]	• Alcohol and ether alternative fuels
Non-recyclable waste	Makarichi et al. [29]	• Review on waste incineration
	Al-Salem et al. [30]	• Pyrolysis of waste plastics
	Hassan et al. [31]	• Co-pyrolysis of biomass and plastics

phase, depending on the application needs and production processes. Liquid and some gaseous fuels are the most promising solution for the transport sector [13], while solid fuels are likely to be used for stationary needs in power plants [14]. Additionally, fuels that might be utilized in more than one form, and simultaneously being used as an energy carrier or storage will be deployed on a greater scale. To maximise fuel and overall system efficiency, cross-sectoral integration is mandatory [15]. This implies, combined heat and power (CHP) production, but even more, deeper integration of transport and industry within the power generation sector [16]. Cogeneration plants have notably higher efficiency compared to conventional power plants; therefore, they are preferred in the future energy system. Moreover, waste heat can be utilized for district heating or industrial purposes, or directly for the production of alternative fuels. Term alternative fuels will be used for all considered fuels in this review, including electrofuels, to avoid potential confusion.

The majority of alternative fuels still haven't reached the commercial scale of application due to the limitations in production or consumption processes and technologies. Mainly, this is related to a high energy penalty which fuels need to undergo during the life-cycle or the economic viability of the production process itself [13]. At the moment, biomass is the only one commercially used, while its consumption is expected to increase even more. Other alternative fuels like hydrogen, ammonia, methanol, biodiesel, biogas, waste-derived fuels, etc. still haven't reached commercial maturity, and their current consumption is almost negligible [17]. Table 1 presents recent reviews on considered alternative fuels with a brief description of the main objectives. McDonagh et al. [18] analysed the cost and efficiency of electrofuels production using curtailed energy when VRES penetration is between 40 and 60%. It was shown that up to 56% more could be achieved in production with approximately similar cost reduction. Lehtveer et al. [19] analysed the cost-competitiveness of electrofuels in future energy systems, showing that they are unlikely to become feasible even with higher penetration of VRES. Abdalla et al. [20] and Parra et al. [21] reviewed the role of hydrogen for deeper system decarbonization, concluding that pronouncedly more needs to be done by policymaking to boost up the broader deployment of hydrogen as an alternative fuel. Valera-Medina et al. [22] and Giddey et al. [23] evaluated the role of ammonia in the future energy system. They find that ammonia might have an important role as energy storage or carrier. Biodiesel and biomass were widely investigated over the years as a carbon-neutral energy source. Lately, the research focus was shifted to the solutions that could significantly improve the properties of biofuels and enhance their efficiency. The utilisation of waste biomass feedstock [24] through thermochemical conversion processes such as pyrolysis [25] or gasification [26] could significantly improve the sustainability of biomass consumption. Various alcohol derived fuels are widely investigated as a potential substitute for IC engines [27]. Especially interesting is the methanol, as the simplest alcohol, which has great potential for utilisation in the shipping sector [28]. Finally, non-recyclable waste could be effectively utilized as a feedstock for fuel production overcoming the problems related to waste incineration [29]. Waste plastic materials are lately investigated for fuel production [30], especially to improve the properties of bio-oils through co-pyrolysis processes [31]. The list of alternative fuels is extensive, and this paper covers mainly the most promising at the moment.

This review paper aims to present and analyse the most prominent alternative energy sources, which are nowadays widely investigated as a potential alternative fuel, and energy carriers or storage. Up to now, various alternatives fuels have been investigated and detailed reviews have been carried out as summarised in Table 1. Nevertheless, comprehensive review which would summarise and evaluate considered alternatives with their advantages and drawbacks, as well as the prospective for greater deployment is widely missing. In addition, alternative fuels are often compared in competitive way, promoting the usage of one fuel for all applications. In this work, the authors analysed the most prominent chemicals, biofuels and alcohol derived fuels with a goal to find a complementary role for each of them in future energy systems. Finding a complementary role is especially important to continue with the research in a way which would maximise application potential of each considered fuel.

2. Materials and methods

The research method is based on a three-step procedure, consisting of (i) systematic literature review and information synthesis, (ii) grouping of studies by selecting the most prospective and promising solutions and (iii) assessment of accuracy and topic relevance. The literature search was done by searching scientific databases Scopus and Web of Science. Fig. 3 presents a flowchart of how the literature review was done. Firstly, the scientific databases were searched for general terms like alternative fuels, synthetic fuels and electrofuels by keywords, abstract and title. The great number of publications can be found when these terms are searched, and most of the studies are not directly relevant to the topic

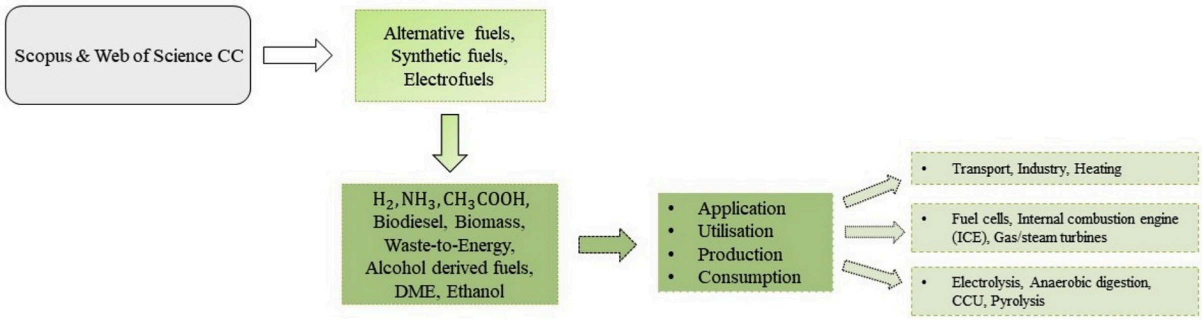

Fig. 3. Flowchart of used methodology for literature review.

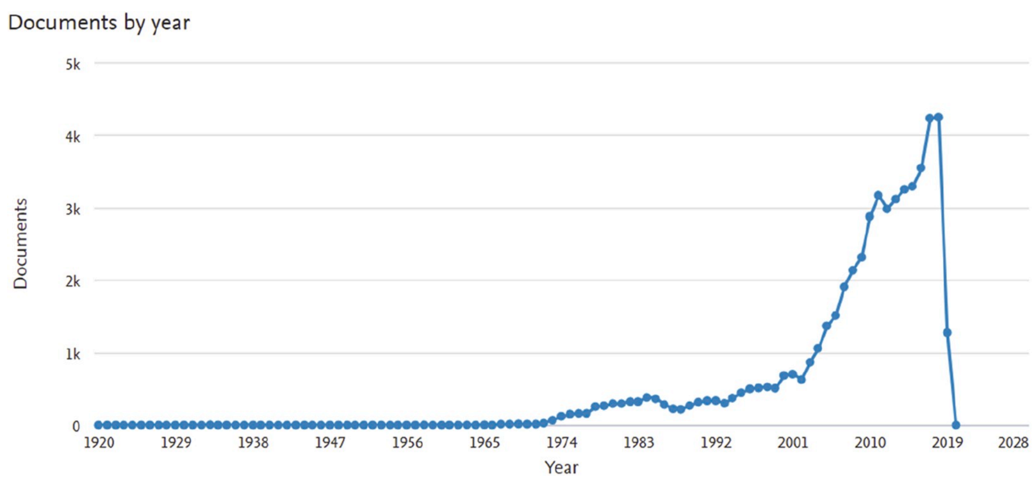

Fig. 4. Number of publications for alternative fuels in the Scopus database [36].

and objectives of this work. Therefore, additional refinement related to the field of energy was applied, narrowing the results to approximately 5000 recent studies which were marked as promising by scanning the title, and keywords. Based on the obtained and synthesized results from the last 5 years, the most promising alternative fuels are selected. This selection was based on the research activity and a number of available publications. Each fuel was additionally investigated and reviewed regarding the application needs, utilisation technologies and production routes.

Fig. 4 presents the number of publications per year that can be found in the Scopus database regarding alternative fuels. From the figure, it can be seen that alternative fuels are gaining research momentum since the 2000s.

3. Review of alternative fuels and utilisation possibilities

To present current and future energy demand, "Global Energy Transformation: A roadmap to 2050" 2018, by International Renewable Energy Agency (IRENA) was used [17].

3.1. Overview of current and future energy demand

According to the IRENA roadmap, the share of renewable energy in total primary energy supply (TPES) was 15% in 2015. This should be increased by two-thirds of overall consumption to meet goals for 2050, while TPES should remain at nowadays level. In 2017, the share of all renewable sources (RES) in the power sector was 25%, with an aim to increase this share to 85% by 2050. This will ensure that electricity from RES accounts for 60% of total renewable energy (RE) in TPES. In a reference case for 2015, electricity accounts for about 20% of total final energy consumption (TFEC), while the rest are other sources, mainly fossil fuels. To meet projected goals, more than 13 000 new, renewable gigawatts needs to be installed. The major increment is expected from VRES, wind and solar photovoltaic (PV) energy, where most new capacities will be installed. The high share of VRES indicates more periods with excess or lack of electricity production, requiring some form of energy storage. Synthesis of alternative fuels from electricity surplus can offer multiple benefits, especially in terms of transport and industry, where very little has been done so far. In 2015, the share of renewable energy in the transport sector was around 4%, while this is expected to increase to 58% by 2050. The most are expected from electric vehicles (EV), especially for light-duty transport; while decarbonization of aviation, shipping, and high-duty vehicles seeks for different solutions. This gap may be filled with high-energy density alternative fuels like hydrogen, advanced biofuels or electrofuels. Transition and decarbonization of an industry sector will be a particularly challenging task. The share of renewable energy for the industry was approximately 14% in 2015, with biomass and renewable electricity equally represented. Electrification of the low-temperature processes will significantly contribute to decarbonization of the sector, while high-temperature processes require the introduction of alternative fuels. Besides biomass, a higher contribution is expected from emerging alternative fuels like hydrogen, enhanced bioenergy and similar. The overall share of renewable energy in the industry is expected to be 60% of TFEC in 2050 [17]. Fig. 5 illustrates the current and predicted renewable energy and electricity consumption according to the IRENA scenario. Current and expected share of renewable energy is on the left, while the share of electricity is on the right side for each sector. In case of power generation, the number refers only to share of renewable energy.

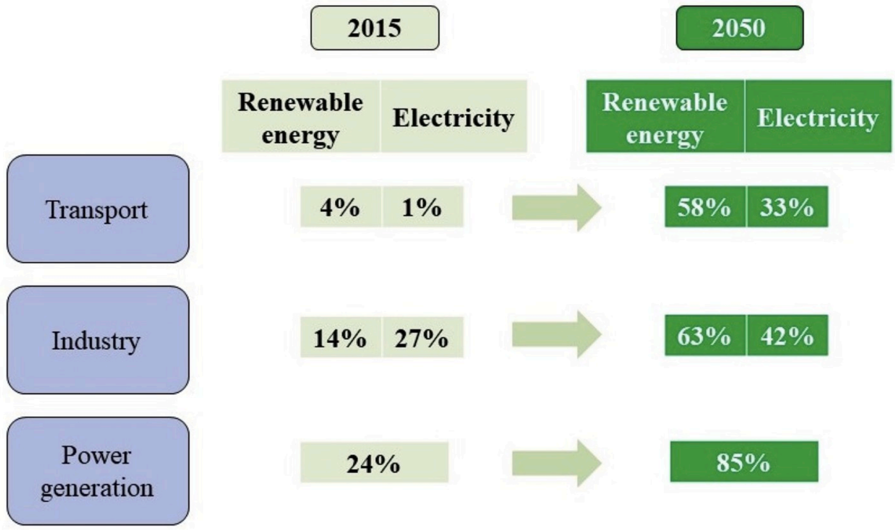

Fig. 5. Current and predicted renewable energy and electricity consumption by the sector [17].

3.2. Alternative fuels

3.2.1. Hydrogen

Hydrogen is the cleanest known energy source that can be produced from various energy sources like fossil fuels, nuclear energy or VRES [20]. Currently, hydrogen is widely used as rocket fuel in the aerospace sector [37], as a refining material for the petrochemical industry as well as in multiple other industrial processes [38]. Almost 50% of hydrogen is globally used only for the production of Ammonia (NH3) [39]. When used as a fuel, hydrogen oxidation releases only water and heat, without additional emissions (Equation (1)). Even though hydrogen is the most abundant chemical element in the universe, its natural, elemental occurrence on earth is seldom. Nevertheless, hydrogen can be found in various hydrocarbons, water or synthesized chemicals.

$$2H_2(g) + O_2(g) \rightarrow 2H_2O(g) + heat \quad (1)$$

One of the biggest advantages lies in high energy density which varies between 120 and 142 MJ/kg [40]. High energy density coupled with the maturity of production processes promotes hydrogen as potential seasonal storage in the future energy, as well as the alternative fuel [21]. Fuel cells look like the most promising solution for hydrogen utilisation for both portable and stationary use [41]. Nevertheless, due to the low volumetric energy content, the efficient application requires liquefication at −253 °C, or compression to 700 bars. Both processes are highly energy-intensive, resulting in energy losses around 10% for compression, and about 40% for liquefaction [39]. In addition, high flammability requires cautious handling procedures and raises several safety issues. Materials used for hydrogen storages must not react with hydrogen in any form and simultaneously serve as an excellent heat insulator [41]. In addition, problems with a hydrogen distribution network are even greater, and it is estimated that new infrastructure would costs over several billion dollars in the coming decades [39]. Even though serious issues are ahead of hydrogen utilisation as a fuel, strong strategic pushback by policymakers and notable research efforts, presume that hydrogen will have a role in the future. To overcome existing problems and open the path for broader application, an appropriate distribution network needs to be developed, and cost-competitive production from renewables should be met.

3.2.2. Ammonia

Ammonia (NH_3) is an entirely carbon-free chemical compound widely used as a fertilizer, which recently gained significant attention as a potential energy carrier or alternative fuel [23]. Ammonia is nowadays widely used chemical and its production accounts for approximately 200 million tons yearly. Currently, the primary feedstock for the synthesis via the Haber-Bosch process are fossil fuels like natural gas, coal, and oil as well as nitrogen from the air [22]. Ammonia is at room temperature, and 10 bar pressure in the liquid phase and its storing is quite easy with already developed distribution infrastructure. The energy density of ammonia is around 22.5 MJ/kg, with one of the highest gravimetric hydrogen densities (17.8 wt%), making it an ideal energy carrier for hydrogen fuel [23]. Sustainable usage of ammonia implies that electricity surplus from VRES is utilized for electrolysis and production of hydrogen, which is then synthesized with the nitrogen from the air. Where needed ammonia is once again converted to the hydrogen and then utilized for power generation [23]. Even though this process is highly energy-intensive and results with a significant energy penalty, the procedure is quite easy, and infrastructure is already in place [42]. Moreover, ammonia can be effectively used as energy storage since its price is more competitive than storing pure hydrogen. According to the study, storing hydrogen in the form of ammonia for 182 days costs 0.54 $/kgH_2$, compared to the 14.95 $/kgH_2$ if the pure hydrogen is stored [43]. There are already existing storage facilities in Qatar that use ammonia for storing hydrogen [44]. If the ammonia is solely used as a fuel, its energy content is equal to H_2 energy content. Complete ammonia oxidation is clean since the products are nitrogen, water and release heat (Equation (2)).

$$4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_20 + heat \quad (2)$$

The main problem of using ammonia is its high toxicity and hazardous nature. Ammonia is a colourless gas with a sharp odour, lighter than air, and it can cause serious health issues. In the liquid phase, ammonia is strongly corrosive, especially if mixed with water [45]. Moreover, incomplete combustion of ammonia leads to the formation of pollutant NO_X emissions. Issues related to the direct application of ammonia in IC engines or gas turbines are related to the high ignition temperature (~650 °C), and comparably lower energy density than gasoline which requires engine modifications [23]. Moreover, ammonia has low burning velocity and often needs additives like H_2, CH_4, or diesel to be ignited. Direct application in fuel cells is only feasible for solid oxide fuel cells (SOFCs) due to the high working temperatures, where ammonia could be cracked and utilized through hydrogen [42]. The utilisation of ammonia as fuel has several concerns; nevertheless, the International Environmental Agency (IEA) classified ammonia as a potential energy carrier and remarkable efforts are conducted globally to establish clean production.

3.2.3. Biodiesel

Biodiesel consists of monoalkyl esters; a long chain of fatty acid oils derived from renewable lipid sources such as non-edible vegetables, lignocellulose biomass or animal fats [46]. There are four generations of biodiesel, even though only two of them reached commercial scale. The 1st generation biodiesel was firstly introduced biofuel, produced from food crops like corn, sugar cane, wheat, and vegetable oils. The second generation is produced from energy crops and non-edible vegetables, waste oils and lignocellulose feedstock. It is important to emphasize that biodiesel can only be produced sustainably if production does not compete with the food supply chain. The 3rd and 4th generations of biodiesel are still emerging, and they include algal biomass and genetically modified microorganisms, respectively [47]. Up to know, biodiesel was successfully applied for the transport sector in fuel blends with conventional oil. There are two standards for biodiesel production, for the EU (EN14214) and for the U.S. (ASTM 6751) [48]. The calorific value of biodiesel is between 38 and 45 MJ/kg, which is comparable to conventional diesel [49]. Problems with biodiesel are mainly related to its higher viscosity and density resulting with fuel injection problems. For this reason, biodiesel is blended with diesel to improve cold start and fuel intake. In addition, lower energy density implies slightly higher fuel consumption [50]. On the other side, the performance of biodiesel in conventional IC engines is quite remarkable [51]. The reduction of pollutant emissions can be up to 78%, depending on the fuel quality and blend ratio [52]. Particularly, biodiesel combustion decreases the formation of Carbon monoxide (CO), CO_2, particulate matter (PM), and unburned hydrocarbons emissions, while NO_X emissions are slightly higher [53]. It was shown that engine performance could be increased by 3% when 20% of biodiesel was mixed with gasoline [54]. Currently, biodiesel is produced through transesterification, where feedstock is mixed with methanol or ethanol [49]. Pyrolysis might be a new potential method for the production of high-quality biodiesel fuels from the various feedstock [55]. The yield of bio-oil in such a process is up to 75%, with a heating value between 36 and 42 MJ/kg depending on the feedstock type, while the process is carried out on mild temperatures between 400 and 600 °C, with the feedstock that contains low moisture content [25]. The interesting research topic is upgrading the bio-oils through the co-pyrolysis process with waste materials to improve quality and fuel properties [56].

3.2.4. Alcohol derived fuels

Alcohol derived fuels like methanol, ethanol, and Dimethyl Ether (DME), have already been successfully deployed for internal combustion engines (ICE). Due to the application limitations, alcohol fuels are often introduced in fuel blends where it shares does not exceed 20% [53]. This review covers methanol as the simplest form of alcohol, ethanol as the commercially used fuel, and DME as the prominent fuel to be used in IC engines in the future.

3.2.5. Methanol

Methanol, known as methyl or wood alcohol is one of the simplest alcohols which oxides as a clean fuel when produced with recycled CO_2 (Equation (3)) [57]. Currently, the primary market for the methanol is the chemical industry, even though significant efforts are given for utilisation as an automotive fuel as well (around 20 million tons/yearly for fuel blends) [34]. At the standard room temperature and pressure, methanol is in a liquid state, which makes it easier for handling and distribution. Nowadays it is mainly produced from catalytic conversion of carbon monoxide and hydrogen from natural gas, or from the gasification of coal. To be used in the future decarbonized energy system, the production process must shift toward cleaner solutions like Power-to-Liquid, which involves CO_2 capture technologies and electrolysis of water [58]. The alternative solution includes the biomass-to-energy approach where bio-methanol is produced [59], or solar production [60]. If the sustainable and cost-effective production is met, there are no further technical barriers for greater usage of methanol as a fuel, especially in the shipping sector [34]. Methanol has been widely shipped over the globe, which encouraged investigations for its utilisation as a fuel. Tanks and IC engines can easily be modified, while several refilling stations have already been installed [28]. Toxicity and high corrosion potential (higher than gasoline), as well as the swelling and shrinking of polymers, represents the main drawback of its utilisation [61]. Besides, methanol energy density is halved compared to conventional marine fuels, which makes it unsuitable for long voyages [62]. Lower energy density implies multiple refilling or the installation of additional tanks. Fuel blend of methanol and diesel can reduce NO_X emission by 30%, while methanol can increase overall engine performance and efficiency [34]. Up to know methanol was used in existing IC engines, while specific methanol engines are under development for smaller vessels, road and commuter ferries [61]. Methanol can also be utilized in the fuel cells, even though this produces relatively lower voltage and has poor conversion efficiency [63]. It should be mentioned that if methanol is not produced from renewable sources, the GHG cycle is even higher than conventional heavy fuel oils. Methanol is also investigated as a potential hydrogen carrier in Power-to-X systems, due to the fact that is it the simplest form of electrofuels [64].

$$2\,CH_3OH + 3\,O_2 \rightarrow 2\,CO_2 + 4\,H_2O \quad (3)$$

3.2.6. Ethanol

Ethanol or ethyl alcohol is the simple form of alcohol, commonly produced from the fermentation of biological matter. Today, a tremendous amount of ethanol is used for the medical application, as well as for the production of alcoholic beverages. Efforts to utilize ethanol for the IC engine started in the 1930s in the USA, with an even greater increase following the oil crises in the 1970s. In that period, significant importance ethanol gained in Brazil, where a national program for the production of alcohol fuels was established alongside subsidies for blending conventional fuels with ethanol. As a result, around 20% of the cars in Brazil are operated solely on ethanol, while the rest can have ethanol share up to 20%. The heating value of ethanol is around 27 MJ/kg, which is pronouncedly lower compared to gasoline (44 MJ/kg) and requires the installation of bigger storage tanks [65]. Besides, oxygen content in ethanol is around 35%wt., followed up by high latent heat of vaporization, indicating problems with a cold start. Ethanol oxidation releases CO_2, H_2O, and heat, as presented in Equation (4). Since the fuel is produced from biological feedstock and crops, CO_2 emissions might be considered neutral [66]. Nevertheless, if higher consumption of such fuel is expected in the future, problems with sustainability may arise due to over usage of biomass feedstock. Another drawback of ethanol combustion in IC engines is related to uncomplete combustion where significant amounts of formaldehyde emissions are released, which promotes the formation of ground-level ozone. The performance of an engine ran on ethanol fuel blend is satisfactory with efficiency similar to those powered by gasoline. Simultaneously, the reduction of CO_2 emissions could be up to 20% when "well to tank" is calculated [67]. Finally, in dedicated modified engines, ethanol performance is pronouncedly better, especially if a comparison is carried out for fuel blends or standard engines [68].

$$C_2H_5OH + 3\,O_2 \rightarrow 2\,CO_2 + 3\,H_2O \quad (4)$$

3.2.7. Dimethyl ether (DME)

Dimethyl ether is the simplest ether widely used as a precursor for the synthesis of a wide variety of organic chemicals. Lately, blending the DME with fossil fuels for spark-ignited engines has been proposed as an interesting method for the enhancement of combustion properties and improvement of engine thermal efficiency [69]. The DME can be produced in a two-stage process where firstly methanol is produced from methane steam reforming and then dehydrated to DME [70]. Sustainable production could be achieved if syngas is obtained from biomass gasification or methanol is produced using CCU technologies and electrolysis [71]. The DME is a non-toxic and non-carcinogenic compound with very low global depletion potential, which makes it an ideal substitute for fossil fuels in IC engines. In addition, the DME burns with a visible blue flame, and it has a sweet odour which is an important safety issue. It has the highest heating value of alcohol derived fuels (~29 MJ/kg), and cetane number similar to that of diesel (55–60), which marked him as a potential diesel substitute [72]. The main advantages of the DME utilisation as a fuel are the following: decreased emissions of NO_X, hydrocarbons, CO and complete absence of soot and SO_X emissions. Significantly reduced pollutant emissions promote the DME as a potential solution for the substitution of diesel fuel in IC engines [73]. A major drawback for wider application is related to comparably lower heating value, which implies the installation of bigger storage tanks. In addition, lover viscosity results with significant injection and leakage problems, demanding a new, dedicated fuel delivery system [74].

3.2.8. Biomass

Biomass is one of the few energy sources, simultaneously used as a fuel and feedstock for fuel production [75]. In 2010, total biomass consumption reached 56 EJ/yr, mainly for residential and building heating and cooking in individual, poorly efficient stoves. In addition, biomass is used as a fuel for cogeneration (CHP) power plants (4.5 EJ/yr), and also in industry and transport sector with the cumulative consumption of approximately 13 EJ/yr. It is expected that inefficient stoves will be replaced by 2050 with modern ones, and biomass will remain an important energy source in rural areas. In the future, demand for the biomass is expected to double by 2030 from nowadays levels to approximately 108 EJ/yr. The increase is expected in all sectors, and it is estimated to be ~31 EJ/yr in transport, and ~21 EJ/yr for the industry. The remaining 56 EJ/yr is foreseen for power generation and heating (individual and district heating) [75]. Traditional biomass (i.e. firewood) which is now widely used, strives for new approaches in order to find more appropriate solutions to enhance the sustainability of its consumption [76]. Firstly, the usage of traditional biomass for heating and cooking in rural areas should be minimized and replaced by electricity. Furthermore, the usage of traditional biomass with low exploitation properties should be abandoned, while the research focus should shift toward enhanced biofuels [11]. Such biofuels have improved combustion properties, easier are for handling and distribution, and finally, can be produced from waste biomass residues. Waste biomass sources like agricultural waste, sawdust, tree shavings, cutters, and wooden chips, are bulky by-products of some other industrial activity, but most importantly they could be efficiently utilized in forms of densified fuels. The most prominent solutions are pellets, briquettes, and cubes. Densified, solid fuels share similar characteristics in terms of density (450–750 kg/m^3), moisture content (8–12%), and heating value (15–21 MJ/kg). The difference is that pellets are mainly used for heating stoves and individual boilers, while briquettes are used for industrial applications [77]. The main advantage of densified fuels over traditional biomass is in the lower moisture content (up to 40%), which enhance overall combustion performance up to 40–68%, depending on the wood type [78]. The promising solutions for upgrading the biofuels could be the pyrolysis [79] or gasification [26]. Obtained product are high-quality biochars, bio-oils, and syngas. Biochar can be used as an environmentally friendly soil fertilizer, bio-oils can be further refined for biodiesel, while syngas can be utilized in gas turbines. Biomass pyrolysis occurs in the temperature range between 300 and 600 °C, in the absence of oxygen, while gasification is carried out between 800 and 1000 °C with controlled air and oxygen content [26]. Some catalysts are used to enhance the selection of product yield [79]. Lately, microalgae are examined for the production of biogas, composed of typical syngas compounds (CO, CO_2, CH_4, H_2O, H_2) with a calorific value between 10 and 35 MJ/kg [80]. Even though the cultivation of algae still didn't reach commercial applications due to the production costs, the idea looks promising since they are not competing with food production. Finally, biomass can be upgraded through co-pyrolysis with waste materials in order to enhance the synergistic effects of individual components and to obtain high-quality products [81]. More on this will be discussed later.

3.2.9. Non-recyclable waste

Firstly, it needs to be stated that Waste-to-energy should be the last measure in waste management systems. Prior to energy recovery, reusing and recycling are preferable, while waste incineration should be applied for the non-recyclable waste only. Currently, a widely used energy recovery method is waste incineration for cogeneration of electricity and heat [29]. Waste is used in the form of solid recovered fuel (SRF), refuse-derived fuel (RDF) or through direct combustion of municipal solid waste (MSW) [29]. Since the waste generation is inevitable and will be generated at higher rates in the future, sustainable solutions for waste management practice is necessary. Thermochemical conversion is a highly efficient method for reduction of mass and volume, but higher SO_X, NO_X and other pollutant emissions raise serious environmental concerns [82]. Decreasing NO_X emissions is especially important since they are a source of multiple health issues [83]. Thermo-chemical treatment of waste is lately introduced as a method to deal with waste materials that reached recycling potential, or their recycling is economically inefficient (low-quality plastics, composite materials, end-of-life plastics). Such materials might be used as feedstock to improve the exploitation properties of biomass or MSW [84]. It was shown that plastics could significantly enhance biomass properties through a synergistic effect when optimal fuel blend is pyrolyzed [85]. In addition, various waste, like rubber [86], MSW [87], or sewage sludge [88] have been co-pyrolyzed with biomass, and again it was shown that fuel blends products (liquid, gas, char) are noticeably upgraded compared to the individual pyrolysis [89]. Using non-recycling waste to upgrade biomass properties offers several benefits. Firstly, over usage of biomass could be prevented since the feedstock needs are partially satisfied with waste. Secondly, the waste management sector can be effectively integrated into the energy system in order to find an appropriate and sustainable disposal solution [90]. Finally, obtained products of high quality can be further utilized where appropriate (bio-liquids for biodiesel, syngas for steam generators). General characteristics of waste fuels could not be provided since the composition of waste significantly varies over the regions and countries, but also over time. This is one of the main drawbacks of waste utilisation as a fuel since the multiple investigations should be continuously carried out to determine the waste composition, characteristics, and appropriate pre-treatment methods. Furthermore, exhaust gases may contain toxic and harmful compounds that require complicated and expensive after treatment [91]. Nevertheless, since the generation of waste is inevitable in the future, sustainable solutions for its disposal should be found. Energy recovery seems the most promising and cost-effective solution, even though public acceptance of this method is still mostly missing. In further chapter to avoid confusion, when implying to energy recovery of non-recyclable waste, "waste fuel" expression will be used.

3.3. Form of utilisation (solid/liquid/gaseous fuels)

Form of utilisation implies the state of matter in which fuel could be utilized. The most of considered alternative fuels might be utilized in more than one state, with the different efficiencies. This section briefly discusses the possible form of utilisation for considered fuel alongside their advantages and drawbacks.

Solid fuels are nowadays widely used for stationary purposes in power plants, or for satisfying high-energy demand in industrial processes [92]. Solid alternative fuels might have an especially important role in the decarbonization of heavy industry, currently dependable on fossil fuels [93]. Alternative solid fuels, like biomass or waste-derived fuels, could be an adequate substitution for fossil fuels without significant infrastructure modifications [94]. Besides space and dry conditions, no additional requirements are needed. Application of solid fuels for the power generation will most likely be in CHP power plants (i.e. district heating), while notable consumption of biomass is expected to remain in rural areas as well [76]. Biomass is already used in the form of densified fuels like firewood, wood chips, pellets, briquettes for heat and power production on a commercial scale [95]. In addition, biomass is often used in fuel blends to decrease GHG emissions of fossil fuels like coal [96]. To achieve sustainability in biomass consumption, new approaches and utilisation technique are necessary. This includes gasification, pyrolysis, and anaerobic digestion of raw biomass with an aim to enhance the properties of derived products. Similar to biomass, waste is also already used as an energy source [97]. Nevertheless, current waste management practice relies on unsustainable methods, where waste is incinerated in CHP power plants or cement kilns without appropriate pre-treatment [93]. This implies that the pre-selection process, where valuable materials would be recovered is skipped, resulting in economic losses as well [98].

Liquid fuels like gasoline, diesel, and heavy oils are conventionally used in IC engines for all types of transport (road vehicles, shipping, aviation) [99]. Even though it is expected that electric vehicles (EVs) will dominate the future transport sector, additional alternative fuels are needed as well [100]. This is due to the fact that heavy, cargo vehicles need high-density fuels for a drive, or propulsion [101]. In addition, battery capacities are still not enough for long-range voyages since they demand multiple charging stops. This becomes a severe issue for overseas transport since multiple stops for charging are unpractical and time-consuming [40]. Alternative fuels that can be utilized in the liquid state are biodiesel and ethanol on a commercial scale, and methanol and DME in the concept proof stage [46]. Pyrolysis oil could also be utilized in a liquid state, even though more research is required to find an appropriate application and production procedure. Finally, hydrogen and ammonia, as potential transport fuels are both facing storage problems when liquified. While ammonia is strongly toxic and usage raises safety concerns; cryogenic technology is necessary to liquefy hydrogen below the critical point of −252 °C, resulting with high energy penalty [22].

Gaseous fuels are important transition fuel, while their importance will increase even more since they can be used in a flexible ramping mode. This is especially important for grid balancing once when a high share of VRES is achieved [102]. Gaseous fuels are utilized in gas turbines or steam boilers, preferably in the CHP cycle with high efficiency [103]. Syngas and biogas are the most prominent alternative fuels to be used for stationary applications like CHP [104]. They are obtained through conventional gasification [105], pyrolysis [106] or anaerobic digestion [107]. The main component of gas fuel is methane, while a notable portion of CO, CO_2, H_2, and higher hydrocarbons are obtained as well [108]. The main drawback of such fuels is inconstant and lower heating value (10–35 MJ/kg) compared to natural gas (19–21 MJ/kg) [109]. On the other hand, hydrogen is the most prominent gaseous fuel to be used for mobile applications, and it is already utilized for automotive purposes, using fuel cell technology [110]. In addition, a lot is expected from hydrogen as a fuel in aviation, heavy-duty vehicles and long-range shipping. Even though hydrogen needs to be compressed to 700 bars, this is still a more appropriate and practical solution for the commercial application then cryogenic liquefication [111]. Used storages are made entirely from composite materials (IV carbon-composite technology) which endures high pressures, and deformation in case of crushing [20]. Lastly, if ammonia is going to be utilized as a fuel, most likely, it will be in the gaseous state [23]. In the gas phase, ammonia can be co-fired with similar gas fuels to improve combustion performance and to overcome problems related to liquid ammonia. Fig. 6 presents the potential application and utilisation technologies for considered alternative fuels. As it was already mentioned, some fuels might be utilized in more than one form and in different technologies. Nevertheless, the efficiency of utilisation in each technology is pronouncedly different, requiring additional insights and research to find the most appropriate solution. More on this will be discussed in the next section.

3.4. Utilisation technologies

This section aims to present the efficiency of considered alternative

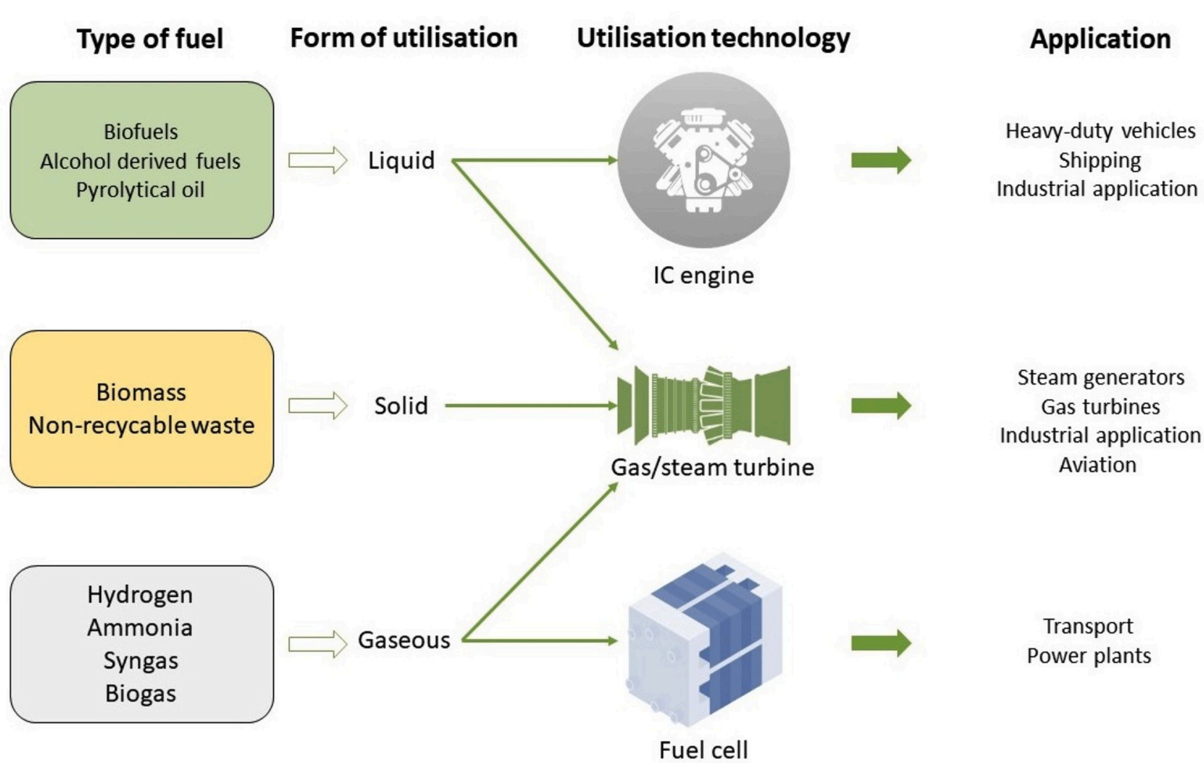

Fig. 6. Form and Technology utilisation perspectives for Alternative Fuels.

fuels demonstrated on commercial or research scale. Majority of considered alternative fuels were tested for all presented technologies with different success. Technologies and alternative fuels that are used commercially are discussed briefly, while more attention is given to the emerging ones.

3.4.1. Fuel cells (FCs)

Fuel cells become widely discussed and investigated technology when hydrogen was introduced as a potential alternative fuel. Proton-Exchange Membrane Fuel Cell (PEMFC) and Solid Oxide Fuel Cell (SOFC) are the most attractive and investigated nowadays [40]. Hydrogen utilisation in fuel cells has gone farthest, and it is already commercially available, with the Toyota Mirai as a notable example of a hydrogen-powered vehicle [112]. Fuel cells are also foreseen for other types of transport, including shipping and aviation sectors [110]. They are relatively small in size, and therefore ideal for portable applications. For the hydrogen case, both fuel cells show a similar efficiency of approximately 50–60% depending on the fuel purity. While PEMFCs seems like a logical solution for portable applications due to the low operating temperature (up to 100 °C), the SOFCs could be the solution for stationary use. High working temperatures (500–1000 °C) of SOFCs requires longer start-up time, therefore more practical application for this technology is in power plants. The efficiency of compressed hydrogen used in PEMFC with all loses is about 40% [9]. Methanol can also be utilized in PEMFC, without reforming, making a new subgroup of proton-exchange fuel cells, called direct methanol fuel cells (DMFC). Operating conditions of these FCs are relatively similar to those of PEMFC, while conversion efficiency varies between 13 and 29% [63]. Ammonia is the last alternative fuel tested with fuel cell technology. Due to the high operating temperature of SOFCs, ammonia can be directly utilized without reforming, with the efficiency between 39 and 50% [22]. Highest efficiency is achieved when ammonia is used for stationary CHP production directly without reforming. If ammonia is used as a vehicle fuel, PEMFC is required due to the lower operating temperature, implying that ammonia is used as an energy carrier, and before being introduced to FCs needs to be reformed to pure hydrogen, which results with significant energy penalty. In the end, the net efficiency for the best-case scenario is between 11 and 19% [23].

3.4.2. Internal combustion engines (IC engines)

Biodiesel is the only fuel utilized in the conventional IC engine on a commercial scale. In addition, biodiesel can be used solely as a fuel or in blends with the conventional diesel. When the bottom one is applied, the share of biodiesel is indicated with factor “B" and the respective share (i.e. B20, indicates that share of biodiesel is 20%, while rest is diesel) [52]. The share of biodiesel in the fuel mix is limited by the engine itself, quality of fuel, and requirements that need to be satisfied. The especially important criterion is fuel quality which mainly depends on the feedstock used for the production, and it is determined based on fuel viscosity, flash point, calorific value, and specific density [54]. Quality can be controlled during the production process by appropriate pre-treatment methods and processes parameter manipulation (temperature, pressure, or used catalyst) [113]. The overall performance of the engine can be enhanced, while concentrations of exhaust emissions may vary. Even though it can be stated that the overall reduction of GHG emission can be achieved when biodiesel is blended with conventional diesel, this strongly depends on operating conditions. While NO_X emissions in most cases are decreased, CO and CO_2 emissions seem to be slightly increased [114]. Nevertheless, it is expected that biodiesel will be used in the future for IC engines since it has been proven in the operating environment, and it is widely discussed as a potential fuel for the aviation sector in the form of bio-jet fuel [115]. Usage of methanol for IC engines has been discussed for a long time with some actual examples of implementation. The problem of methanol deployment for IC engines is related to its high corrosive potential, which requires engine modifications [116]. Finally, methanol has lower energy content compared to petroleum fuels which imply a need for larger tanks. Nevertheless, simple production procedures, coupled with the increased engine performance and efficiency, opens the possibility to use methanol in the shipping sector as a partial substitution for fossil fuels. This is supported by the fact that methanol can reduce NO_X emission by up to 30%, which is a remarkable success for the shipping sector [28]. Ammonia was tested for IC engine applications as well [110]. The main problem of using ammonia in the IC engine is related to the high burning temperatures, which require the addition of some other fuel like diesel to enhance the start-up process [117]. These problems are prevailing when spark-ignition engines are used [23]. Generally, when ammonia is used as fuel for the IC engine, it must be in conjunction with some other conventional fuel to ease the start of the combustion process. Relatively low reactivity followed by high auto-ignition temperature and low flame velocity limits the application of ammonia solely as a fuel. Achieved overall efficiency of ammonia combustion in IC engines is between 35 and 40% [22]. The advantage of using ammonia in the IC engine is derived from the fact that high octane numbers (~130) can reduce knocking and improve combustion properties. The main issue related to ammonia application in IC engines is in fact that potentially higher NO_X emission can occur if there is incomplete combustion.

3.4.3. Gas and steam turbines

Biomass is already used in the CHP cycle, and its consumption will only increase [114]. The great advantage of biomass is that it can easily be introduced to existing power plants where can be combusted solely or in fuel blends with fossil fuels. Even though the efficiency is slightly lower, a remarkable reduction of pollutant emissions in exhaust gases might be achieved, especially in terms of NO_X, SO_X, and particulate matter emissions [118]. Furthermore, emitted CO_2 can be considered neutral since it was consumed during plant life. If there is high moisture content (i.e. firewood), combustion efficiency is notably lower due to the fact that a considerable amount of energy is used for vaporization [119]. Lately, significant efforts are noticed in the research, to achieve synergistic effects of biomass and other types of solid fuel in order to enhance fuel quality and properties [120]. Such fuel blends (i.e. biomass-plastics) could be effectively applied in power plants since the treatment systems for exhaust gases are already in place [121]. Biogas and syngas, as the products of biomass upgrading, can be utilized in gas turbines for the combined cycle as well [122]. The quality of biogas obtained from anaerobic digestion (AD) depends on feedstock type, but even more on production conditions [123]. More on AD will be discussed in the following section. Syngas is, on the other hand, derived from biomass gasification (800–1000 °C) or pyrolysis (300–600 °C) and again, slight shifts in the temperature region significantly affect its composition [26]. This is directly reflected in its calorific value and consequently, overall efficiency. When obtained gaseous fuels have a higher share of hydrocarbons and hydrogen, combustion characteristics are better, and efficiency is higher [124]. If gaseous fuels are synthesized from renewables, emitted CO_2 can be considered carbon neutral. In order to decrease the share of CO_2 in biogas composition, a further upgrade is required. This implies amine scrubbing for CO_2 removal or co-pyrolysis of biomass with high calorific waste on high temperatures, to increase hydrocarbon content [125]. The utilisation of biogas in power plants has a significant drawback since it may cause acidification and eutrophication several times higher compared to fossil fuels [126].

Waste incineration is a long-time used practice for energy recovery of waste materials. Solid waste is introduced to the power plant where it is burned at high temperatures between 750 and 1100 °C [98]. Because of the feedstock content, exhaust gas contains various pollutants like SO_X, NO_X, CO_X, Polycyclic Aromatic Hydrocarbons (PAHs) and heavy metals. This requires complicated and expensive treatment of flue gases, and it is considered as a major drawback. Nevertheless, stringent control emissions make this process quite effective for waste management, simultaneously producing heat and electricity with an efficiency of up to 80% [127]. Hydrogen and ammonia could also be utilized in gas turbines,

Table 2
Summary of main fuel characteristics.

Type of fuel	Chemicals		Biofuels				Waste
	H_2	NH_3	Alcohol derived fuels	Biodiesel	Biomass	Syngas/biogas	
Calorific Value (MJ/kg)	120–140	22.5	~19 (Methanol) ~27 (Ethanol) ~29 (DME)	38–45	15–21	10-35 Syngas 15-22 Biogas	Vary
Feedstock and production	Electrolysis, Biomass and waste gasification	H_2 (electrolysis) + N_2 (air separation)	Biomass, CO_2 (CCU), H_2 (electrolysis)	Energy crops, waste oils, lignocellulosic plants	Sawdust, agricultural waste, tree shavings and cutters and similar	Biomass, non-recyclable waste, biodegradable waste	MSW, RDF, SRF, non-recyclable waste, sewage sludge
Combustion products	H_2O, Heat	H_2O, N_2 Heat	CO_2, H_2O	CO, CO_2, NO_X	CO_2	CO_2, CO, NO_X	CO_2, NO_X, SO_X, various pollutants
Utilisation efficiency							
Fuel cell	50–60% (PEMFC/SOFC)	11–19% (SOFC)	13–29% (DMFC)	–	–	–	–
IC engine	–	35–40%	Up to 40% (depends on the type of engine)	Varies	–	–	–
Gas and heat turbines	–	55–60%	–	–	Up to 80% (CHP), Electricity production; 30–34% dry biomass, 45% co-firing		Up to 80% (CHP); only electricity ~22%

even though seldom work was done in this field. Ammonia was used in various fuel blends and achieved efficiency in combined cycle with gas turbines is between 55 and 60% [23]. Problems reported with the application of ammonia for IC engines are similar in this case as well. Direct combustion of hydrogen in gas turbines have a severe drawback related to its high reactivity which results in high burning temperatures, flame speed and similar. Therefore, hydrogen utilisation in gas turbines requires the development of dedicated technology [128]. Table 2 summaries all presented alternative fuels with their main characteristics such as calorific value, feedstock for production and derived combustion products. Between the considered alternative fuels, hydrogen has the highest calorific value without emission of greenhouse gases. Moreover, it can be produced from the completely clean procedure, if the electrolysis is powered by renewable energy sources. Ammonia and alcohol derived fuels, express the most disadvantaged characteristics required to meet fuel specifications. They have the lowest calorific values, and incomplete combustion might result in even higher emissions.

4. Production pathways

This section aims to present essential technologies and processes for the synthesis of considered alternative fuels. Water electrolysis might be a key technology for fuel synthesis, since it can be driven in flexible mode, allowing higher penetration of VRES. Even more, clean hydrogen is inevitable for the production of other forms of alternative fuels as well.

4.1. Sustainable methods for clean production of alternative fuels

4.1.1. Hydrogen production

Hydrogen production from fossil fuels is a known procedure where natural gas or coal is used as a feedstock. Today, hydrogen is most often produced from steam reforming of methane, while it can be produced from partial or autothermal oxidation or gasification as well [20]. Nevertheless, production from fossil fuels is not possible in the future decarbonised energy system, and procedure must shift toward sustainable solutions. Production from renewable energy sources implies pyrolysis or gasification of biomass [26] or water electrolysis from the electricity surplus from VRES [129].

One of the most prospective ways to produce clean hydrogen is water electrolysis (Equation (5)). Notable research efforts are conducted to bring this procedure on a commercial scale, and even though this accounts for only 4% of today's production, perspective is bright [129]. There are several types of electrolysers, divided by the nature of electrolyte they use. The most prominent ones are Polymer Electrolyte Membrane (PEM) electrolyser, alkaline electrolyser, and Solid Oxide Electrolysers (SOE) [129]. Electrolysers have the capacity to produce hydrogen with high purity (99.999 vol%) with the efficiency of between 70 and 85% [130]. Process efficiency mainly depends on the load factor of renewables and electrolyser efficiency itself. Since the water is carbon-free, and technology reached the maturity stage, the last step for broader deployment of electrolysis is the economic competitiveness of the procedure. At the moment, production costs of hydrogen from electrolysis (~$3/kg) are double than those from natural gas reforming ($1.2.-1.5/kg) [20]. Since the electricity is the main driver of electrolysis production costs, once when higher penetration of VRES is achieved, this procedure would be entirely competitive to steam reforming of fossil fuels. This is especially important in the future energy system, where it will be more periods with electricity surpluses, which can be effectively utilized for electrolysis. This would ensure grid stability, avoidance of production curtailment, and more importantly, clean production of hydrogen [128]. Furthermore, hydrogen can be directly produced from solar, nuclear or waste heat utilisation from industrial processes. If the hydrogen is produced directly from solar energy, concentrating solar power (CSP) seems like the optimal solution since higher temperatures are required [21]. Production using nuclear energy implies the integration of waste heat for high-temperature electrolysis, even though this requires further research efforts [131].

$$2\,H_2O \rightarrow 2\,H_2 + O_2 \quad (5)$$

Technological maturity implies that clean hydrogen production could be completely viable once when a higher share of VRES is integrated since this will cause a further reduction of electricity costs which are the main driver for full commercialization and broader application.

4.1.2. Ammonia synthesis

The main constituents for ammonia synthesis are H_2 and nitrogen (N_2) via the Haber-Bosch process. Hydrogen is most often obtained from the reformation of natural gas, which accounts between 1 and 2% of the annual energy demand [45]. The Haber-Bosch process is energetically demanding and kinetically complex. It is important to emphasize that Haber-Bosch production of ammonia operates as a continuous process whereby each pass through the reactor converts only about 15% of the N_2 and H_2 to NH_3, yet with continuous recycling and overall conversion rates are around 97% [132]. This recycling implies that intermittency of VRES is not a severe problem since the feedstocks can be produced when there is electricity excess and stored for later use. Some types of "green" ammonia synthesis processes have been demonstrated in America, Australia, Africa, Canada, Germany, the Middle East, Norway, and the United Kingdom [133]. Moreover, a small-scale solar ammonia facility has been operating for a few years at Pinehurst Farm in Iowa. The ammonia thus produced is used as a fertilizer and as a fuel for tractors [134].

Switching to clean production implies the electrolysis and air separation, which can be entirely powered by VRES. Cryogenic air separation provides N_2, used for ammonia production and oxygen, which has other valuable applications. To maintain a fully green process, and avoid CO_2 emissions, the electrolyser should be powered by electricity surplus from the grid or by direct renewable solar energy installed in situ [22]. If the direct solar energy is used, due to the low solar conversion efficiency (~16%), the overall primary energy input increases, from 16.4 MJ/kg-NH_3 of methane to 236.7 MJ/kg-NH_3 of solar energy. Low efficiency leads to higher production costs due to higher energy demands. In the best-case scenario, 27.2 MJ of solar electricity displaces 16.4 MJ of natural gas, required to manufacture the same amount of ammonia (1 kg) [45]. Nevertheless, in regions endowed with wind and solar resources and with a high share of VRES, green ammonia could be competitive. In these ideal locations, the cost of solar and wind electricity is predicted in the range of $30/MWh, which translates into a cost-competitive $2/kg of H_2 from water electrolysis. In other words, if solar electricity is available, usage for ammonia production is suboptimal at least until the electricity mix becomes nearly 100% renewable [43].

4.1.3. Methanol synthesis

An innovative trend becoming increasingly evident in the scientific literature is the use of light to drive or assist chemical reactions and processes to produce clean methanol. The prospect of using solar energy, CO_2 and water to synthesize methanol could lead to an economically viable technology, capable of replacing fossil fuel heavy industry with a renewably sourced alternative [59]. There are different ways to produce light-assisted chemical products, including direct utilisation to convert CO_2 and water through solar thermochemistry, photochemistry, or photoelectrochemistry. Another potential solution is the gasification of biomass feedstock to produce syngas [64]. Solar concentrators in conjunction with complementary focusing elements, intensify the sunlight incident on the biomass gasification reactor. The temperatures thereby achieved should be sufficient to affect biomass gasification (~850 °C) without the need for external heating. Again, similar to ammonia, the low efficiency of solar-to-power technology is considerable constraint affecting overall processes efficiency. Therefore, an interesting solution might be coupling hydrogen from electrolysis and integration with CCU technologies utilising electricity surpluses from the grid [60]. An excellent example of sustainable and clean methanol production is in Reykjavik, Iceland. This industrial facility commissioned in 2007, annually produces 4000 metric tonnes of methanol made from captured CO_2 and H_2. This corresponds to 5500 MT of recycled CO_2 per year. The location of the facility allows utilisation of geothermal steam from the 75 MW_{el} Orka's Svartsengi power station to provide renewable heat and electricity., while captured CO_2 accounts for about 10% of total annual power plant emissions. Electricity is mainly used to power alkaline water electrolysis to produce H_2, which in turn reduces CO_2 in the presence of a catalyst, in a process operating at 250 °C and 5–10 MPa [129].

4.1.4. Anaerobic digestion

The anaerobic digestion is a process that includes four biometabolism steps (hydrolysis, acidogenesis, acetogenesis, and methanogenesis) in which biodegradable waste is converted into valuable biogas, consisting mainly of methane [123]. The AD is an optimal process for treating a biodegradable fraction of MSW, agriculture waste (animal manures, energy crops, algal biomass, harvest remains), food industry waste (food/beverage processing, dairy, starch) or sewage sludge [135]. The four-step process can be carried out into single-stage or multi-stage AD systems, even though the bottom one requires additional research. The overall process is determined by complex relations between various operating parameters, growth factors, system design, and the type of reactor [123]. Type of the feedstock is essential for the selection of system design and type of the reactor, as well as it affects growth factors and operational parameters [107]. During the process, pH is stringently controlled since it influences the bacteria efficiency, consequently the success rate of the process as well. Nowadays, most of the AD systems are operated in continuous single-stage mode, processing various biodegradable waste [136]. Even though anaerobic digestion is a complex process with higher investment and operational costs, installed capacities increased from 2 to 11 million tonnes, over the last two decades [123]. Installed capacities are expected to increase even more since the generation of biodegradable waste is inevitable, while AD looks like a promising waste management method [137]. Nevertheless, further research focus should be given to multi-stage AD, where high-quality biogas can be produced, and cost reduction to achieve economically viable production.

4.1.5. Carbon capture and utilisation

Carbon capture and utilisation technologies are an important part of the supply chain for the production of alternative fuels using recycled CO_2 emissions. The title indicates that carbon capture technologies are focused on extracting CO_2 emissions from the point source or directly from the air and then utilising it where needed [4]. Lately, these technologies have been marked to play a complementary role in future energy systems since they can be operated in flexible mode. This allows grid stabilization through Power-to-X (PtX) processes once when higher penetration of VRES is achieved [64]. PtX implies the utilisation of captured CO_2 into some form of electrofuels, reducing the need for battery storages, simultaneously producing a valuable liquid or gaseous fuels [138]. Major technologies for carbon capture, include pre-combustion capture, oxyfuel combustion, chemical looping combustion (CLC), post-combustion capture, capture from fermentation processes, and direct air capture (DAC). An extensive review of the presented processes is given by Mikulčić et al. [4]. Even though CCU technologies might have remarkable efficiency (up to 98% for amine scrubbing) in terms of CO_2 emissions, they inevitably affect overall system efficiency due to the high energy penalty for its operation. The techno-economic analysis which was carried out by Bhave et al. [139], estimates the cost at 145–185 €/t for 50 MW plant, with CLC being the least expensive, and pre-combustion being the most expensive. It should be mentioned that CCU technologies are mostly in the R&D phase, except post-combustion amine scrubbing and pre-combustion natural gas processing [140]. Since the introduction of electrofuels, the CCU technologies are assessed through their role in PtX production pathways, which might become an essential market for scaling up technology on a commercial level. Finally, even though carbon capture has an important role in the future energy system due to operational flexibility, meeting the cost-competitive price of operation is a crucial step for broader deployment [141]. The bottom one is especially important since the installation of a carbon capture system results with significant energy penalty and reduced overall system efficiency.

4.2. Fuel blends pyrolysis for enhanced characteristics of biofuels

Pyrolysis is a thermochemical conversion method where thermal decomposition takes place in the absence of oxygen. Derived products are carbonized residue, liquids, and gases. Lately, pyrolysis has been introduced as a promising technique for the conversion of waste materials into valuable fuels and chemicals [30]. Depending on the desired product distribution, pyrolysis is operated at different temperature ranges. In case the liquid yield is preferred, temperatures go up to 600 °C for most of the feedstock, while gasification is carried out on temperatures above 700 °C [55].

Biomass pyrolysis is most often carried out on temperatures between 200 and 450 °C, where feedstock is converted to high-quality liquids, pyrolytic gases, and carbon-rich char residue [79]. Product yield depends on operating conditions and the feedstock type, while obtained products usually need to undergo refinery processes prior to utilisation. In an example, bio-oils generally contain lower heating values and are unstable at a higher temperature, while pyrolysis gases may contain a high share of CO_2 [142]. Recently, significant research efforts are given to convert biomass feedstock into valuable fuels and chemicals. Especially interesting is the pyrolysis of waste materials like sawdust, agricultural waste, various straws, energy crops and similar [126]. Even though pyrolysis can significantly enhance biomass properties, further upgrade in terms of heating value, lower viscosity, high acidity, and thermal stability requires additional efforts. Interesting might be the synergistic effect that occurs during the co-pyrolysis of biomass with waste plastics [143]. Plastic has a high share of carbon and hydrogen, and the heating value similar to those of fossil fuels [144]. Besides, low share or complete absence of oxygen in the elemental composition reduces the yield of oxygenated compounds, marked as the main drawback of biofuels. Several research showed that co-pyrolysis significantly enhance the bio-oil properties in terms of heating value, thermal stability and viscosity. Since the chemical and mechanical recycling of plastics is expensive, while for some types not even feasible, co-pyrolysis seems like a promising method for waste management as well [145]. Besides, the different type of non-recyclable waste can be co-pyrolyzed with biomass, like sewage sludge (SS) [95], food waste [108], MSW [82], rubbers [86], etc. Even though conducted investigations showed that product properties are greatly enhanced in the co-pyrolysis process, more needs to be done to reduce the yield of various pollutants that constrain immediate utilisation. In Table 3, Ultimate and Proximate analysis of various waste materials, investigated as a potential co-pyrolysis feedstock is given. Characteristics given in Table 3 are essential for the feedstock selection and adjustments in co-pyrolysis or co-gasification process.

The most valuable pyrolysis product is bio-oil, which yield is favored when a high concentration of Volatile matter (VM) in the feedstock is present. This is found for waste plastic, marking them as an ideal feedstock for co-pyrolysis to enhance bio-oil properties. Moreover, the ash content and fixed carbon, which constrains liquid yield, is pronouncedly low for plastics. Finally, the pyrolysis of plastic yields a significant number of different hydrocarbons which is preferred in terms of heating value [144]. Nevertheless, using plastics in energy recovery raises several serious issues as well. Since the plastic materials are produced from fossil fuels and synthesized with different additives, toxic and hazardous compounds might be found in the obtained pyrolysis product [152]. Mainly, this is related to the formation of different PAHs, dioxins, furans, toxic hydrocarbons, and similar [125]. Moreover, a significant amount of chlorine-containing compounds might be found in both liquid and gaseous phases, which are not just toxic, but corrosive and therefore, unfavorable for further exploitation [153].

Conducted experimental investigations showed that the liquid yield of co-pyrolysis is of better quality than those of plastics and biomass pyrolysis alone [154]. Zhang et al. [155] investigated the catalytic co-pyrolysis of pine sawdust and plastics (polyethylene PE, polypropylene PP, and polystyrene PS) in order to maximise the production of aromatics and olefins. The best-case scenario showed that the overall yield of aromatics and olefins could be enhanced by 36%, and 35% respectively for PE/pine sawdust ratio 4:1 at 600 °C. Lu et al. [156], confirmed the thesis that the interaction of plastic and biomass leads to the reduction of oxygen and water content in the liquid fraction, and as a consequence, obtained oil has higher heating values and stability. Zhang et al. [56] investigated the potential for bio-jet fuel upgrade through the synergistic effect of biomass and plastic co-pyrolysis. Results showed that catalytic microwave pyrolysis could yield a sufficient number of hydrocarbons (42.66%) to meet jet-fuel specifications. There are numerous other examples of biomass/plastic co-pyrolysis under different conditions and with different goals. Conducted research showed that the synergistic effect significantly enhances individual characteristics, even though a cautious approach should be maintained due to the evolution of toxic and hazardous compounds. Besides plastics,

Table 3
Ultimate and Proximate Analysis of different biomass and plastic materials.

	Ultimate Analysis				Proximate Analysis			
	Volatiles	Moisture	Ash	Fixed carbon	C	H	N	O
	wt.% (different basis)				*wt.% dry basis*			
Miscanthus [146]	69	4.7	3.0	22.67	49.6	5.9	1.06	42.84
Grasses[a]	69.0	12.6	4.3	16.8	49.2	6.1	0.9	43.7
Straws[a] [147]	66.7	10.2	7.8	15.3	49.4	6.1	1.2	43.2
Shells and husks[a] [147]	64.6	12.4	18.6	4.4	50.2	6.3	1.4	41.9
Sawdust[b] [147]	84.6	–	1.1	14.3	49.08	6.0	0.5	43.7
Furniture waste [148]	72.9	12.1	3.2	11.8	51.8	6.1	0.3	41.8
Sugarcane bagasse [147]	76.6	10.4	1.9	11.1	49.8	6.7	0.2	43.9
Macroalgae [147]	45.1	10.7	21.1.	23.1	43.2	6.2	2.2	45.8
HDPE [149]	97.15	–	0.8	–	86.5	15.1	–	–
PP [149]	96.9	–	1.0	–	84.7	15.3	–	–
PET [149]	84.1	–	–	13.9	64.1	3.7	–	34.2
Rigid polyurethane foam [150]	83.2	–	6.2	10.6	62.7	6.3	6.4	24.0
Sewage sludge [151]	57.22	5.42	31.27	6.09	36.11	5.25	6.50	–

[a] Mean Value obtained after analysis of different samples from the respective group.
[b] Measured at the dry basis.

sewage sludge (SS) could be used in fuel blends with biomass in order to deal with its disposal problems. Pyrolysis of sewage sludge solely at 800 °C, yields around 55% of the gaseous phase with the methane, hydrogen, and CO as the main constituents, and the heating value of 19.27 MJ/Nm^3 [157]. The SS is not a potential candidate for a bio-oil upgrade, but it can be used to obtain high-quality syngas and char residue. While syngas could be further utilized in gas turbines, quality biochar (free of pathogens due to high temperature), could be used as a fertilizer. Furthermore, biomass and SS can be pelletized together and used for power generation. The benefits of this method are the following; reduction of energy demand for the production of pellets, while the breaking force and Meyer's hardness are significantly higher. In addition, moisture absorption of biomass-SS pellets was lower, ignition temperature was reduced, and combustion temperature and performance were enhanced [151].

4.3. Current challenges and future trends

Currently, there are numerous constraints for greater deployment of alternative fuels. First of all, the availability of fossil fuels makes it hard for alternative fuels to meet cost-competitive production costs. In the case of biofuels and waste fuels, a quality criterion is the main concern; lower heating value, higher acidity, thermal stability and similar limits wider deployment of current commercially available biofuels. Nevertheless, research in this field is ongoing for some time with the constant enhancement of produced fuels, implying that the role of such fuel is not questionable in the future. On the other hand, considered chemicals (H_2, NH_3 and alcohol derived fuels) have well-known production procedure, but they are predominately synthesized for industrial needs. This implies that higher production costs are not a concern for such an application, but the further reduction is expected if the intention is to use them as a fuel. Furthermore, deployment of new fuels requires modification on existing utilisation technologies. While biofuels and alcohol derived fuels could be effectively utilized in existing IC engines with slight modifications, development of new technologies or significant modifications are required in case of hydrogen and ammonia. Fuel cells, developed for hydrogen utilisation, shows excellent perspective to be deployed for both stationary and portable applications, even though additional work is required to optimise operating parameters and increase efficiency. The last obstacle for the broader deployment of alternative fuels is the production, which needs to shift toward clean and sustainable solutions. In the case of biofuels, this predominately implies utilisation of waste agricultural and industrial biomass residues to produce high-quality clean fuels. Simultaneously, to achieve carbon neutrality, production of synthetic fuels should shift toward new solutions which do not include processing of fossil fuels as a feedstock. Additionally, synthesis of alternative fuels should be coupled with VRES, allowing them higher penetration into the energy system, simultaneously reducing the carbon footprint of produced fuels. Coupling the synthesis with VRES could also reduce the production costs once when a higher share of intermittent renewable sources is achieved. A notable trend in research is the direct utilisation of solar energy for fuel synthesis. The main advantage of solar production is in fact that there is no need for an external energy source. Nevertheless, the low conversion efficiency of solar energy is greatly influencing the overall process efficiency, making solar production economically uncompetitive. In addition, significant research efforts are given to bring technologies that can be operated in flexible mode on a commercial scale. This is especially important for electrolysis and carbon capture technologies which are used to produce essential feedstock (H_2 and CO_2) for alternative fuels synthesis. Coupling these technologies with VRES would have multiple benefits like reducing the production costs, decreasing the curtailments in power production, and improving grid stability. While talking about thermochemical conversion methods for alternative fuel production, significant research efforts are given to bring such processes on a larger scale and commercial level. Pyrolysis and gasification are especially interesting since they can process various waste materials and convert them into valuable fuels or chemicals. Recently, the research focus is shifted to enhance biofuels properties through co-pyrolysis or co-gasification with high calorific waste materials (i.e. end-of-life plastics). This is not only important for fuel synthesis but as a waste management method as well.

5. Conclusion

Alternative fuels are inevitable in the future decarbonized energy system. Even more, alternative fuels are especially essential to decarbonize transport and industry sector, where electricity will have a much lower impact, or it is not suitable as a replacement. In this review, the main goal of the authors was to present current potential alternative fuels within their applications, and present prospective alternative routes for their production. The bottom one is significantly important since it can be seen that current production pathways mainly rely on fossil fuels in both terms, the feedstock and fuels. Following conclusion are derived from this review:

- Biofuels, especially biodiesel and solid biomass, are the only alternatives available on a commercial level and already utilized for transport and industrial needs. Since their consumption is expected to increase even more in the future, new solutions should be found to achieve sustainability. Thermochemical conversion of raw feedstock through pyrolysis or gasification, as well as the anaerobic digestion of biodegradable waste, looks like promising solutions where future research efforts should be given. Additionally, waste management can effectively be incorporated within the production of enhanced biofuels, simultaneously tackling environmental concerns and improving biofuels properties.
- Chemicals like hydrogen and ammonia were tested as an alternative fuel for various utilisation technologies. Hydrogen has high energy density which marks it as a potential solution for high-temperature industrial processes or transport sector that requires such fuels. Nevertheless, hydrogen is widely used for other purposes as well, which implies that only a limited amount would be available for fuel application. Moreover, a new distribution network is required for greater deployment of hydrogen, which presents serious drawback. Ammonia, on the other hand, has a lower heating value, several safety concerns, and poor combustion properties. This suggests that role of ammonia as an alternative fuel will be very limited. Nevertheless, ammonia has a great hydrogen gravimetric density and could be used as an energy carrier or storage since the distribution is not a concern.
- Alcohol derived fuels are known alternative for some time. Nevertheless, commercial application on a greater scale is doubtful. Besides, lower heating values, which imply higher fuel intake, additional modifications or the development of dedicated IC engines, is necessary to achieve higher efficiencies. Nevertheless, such fuels show interesting characteristics when used in fuel blends, especially in terms of reducing pollutant emissions. In addition, methanol, as the simplest alcohol was successfully tested for marine application, with encouraging results regarding the engine performance and reduction of exhaust emissions.
- Greater deployment of alternative fuels can be expected once when the cost-competitive production is met. Strategic pushback can have a significant effect on this; nevertheless, the final price of produced fuels should be similar to conventional fuels. Higher penetration of VRES would allow this cost reduction since there will be more periods with an excess of electricity production, which can be effectively utilized for alternative fuel synthesis. Simultaneously, this would allow even greater penetration of intermittent renewable sources, since the produced alternative fuels can act as energy storage.

- Finally, production pathways should shift toward sustainable solutions and coupling with VRES. Predominantly this implies direct utilisation of solar energy to drive the production process or integration of various technologies like electrolysis and carbon capture with the VRES to achieve clean production of feedstock used for fuel synthesis.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Prof Neven Duic, which is co-author of this work, was blinded from the review process.

CRediT authorship contribution statement

H. Stančin: Writing - original draft, Writing - review & editing, Visualization, Conceptualization, Methodology. **H. Mikulčić:** Writing - original draft, Writing - review & editing, Methodology. **X. Wang:** Supervision, Writing - review & editing, Formal analysis. **N. Duić:** Supervision.

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