Comparative Assessment of Methanol and Ammonia: Green Fuels vs. Hydrogen Carriers in Fuel Cell Power Generation

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Abstract

Methanol and ammonia emerge as two of the most important energy carriers in a new decarbonized society. In this work, a systematic assessment of the power generation based on these chemicals is performed using two different alternatives: direct utilization as green fuels in fuel cells or as carriers for hydrogen. Despite the need for a previous stage for hydrogen production, the use of these chemicals as hydrogen carriers demonstrates higher efficiencies (around 40%), mainly due to the higher degree of maturity of the hydrogen fuel cells. This is reflected in the cost of electricity for the different alternatives with around 700 €/MWh for hydrogen carrier options and about 1200 €/MWh for the direct utilization as green fuels. Compared to hydrogen, the use of methanol or ammonia has a higher electricity production cost. However, future improvements in the efficiency of fuel cell units could convert these fuels is competitive options. In addition, for different scenarios combining transportation and power generation, methanol and ammonia emerge as technically and economically feasible alternatives, especially for distances over 3000 km. Consequently, both hold a pivotal role in addressing the challenges associated with hydrogen within a future energy systems characterized by high renewable penetration.

Keywords: Ammonia, Energy carriers, Fuel Cell, Methanol, Power Generation, Renewable Energy

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1. Introduction

The energy sector is a significant contributor, responsible for approximately three-quarters of today's greenhouse gas emissions, demanding a profound transformation in the years to come [1]. A significant increase in the penetration of renewable energies is expected, in particular, wind and solar technologies. However, their inherent variability could jeopardize the stability and reliability of the electricity grid. In addition, direct electrification remains a significant hurdle in certain sectors such as heavy transport, shipping, and industrial heating. In this context, hydrogen emerges as one of the most powerful solutions for the green energy transition [2].

According to BloombergNEF [3], hydrogen and its derivatives have the potential to significantly reduce about one-third of the current global emissions from fossil fuels and industrial processes. Furthermore, it is projected that by 2050, they could supply around 24% of the world's final energy demand. Hydrogen is being considered as a means to decarbonize challenging-to-abate industrial sectors like steel production [4] or the chemical industry [5]. Additionally, hydrogen is gaining attention for its use in energy storage, particularly for medium and long-term applications [6], as well as for the production of green fuels [7]. Although the current costs are relatively high, averaging around $4-5 per kilogram, the advancements in electrolysis technology are expected to drive these costs down to a range of $0.7-1.6 per kilogram before 2050. However, it is essential to note that hydrogen has limitations, primarily due to its low volumetric energy density and challenges associated with its storage conditions, such as leakage and corrosion. Consequently, alternative hydrogen-derived products have been suggested for the applications mentioned above, offering more manageable storage and transportation conditions. Among these alternatives, methanol and ammonia emerge as the most appealing liquid options [8]. These chemicals have been explored for applications such as energy storage [9] including their combustion in different devices [10] or their use in fuel cell units [11]. Another important area of application is as alternative fuel in sectors such as maritime transport [12] or aviation [13]. Although the use of these fuels could reduce the global energy efficiency compared to hydrogen, the improved properties in terms of storage and transportation make them attractive for introduction into the future energy system. In their production, synergies between the use of these species as

chemicals and as fuel should be explored [14].

Ammonia and methanol can be used for different applications through two approaches: as green fuels or as hydrogen carriers. In the first scenario, these chemicals are produced from green hydrogen and subsequently utilized directly in various end-use sectors, such as maritime transport [15] and power generation [16]. In the second role, ammonia and methanol function as intermediaries, simplifying the storage and transportation of hydrogen. This involves the initial synthesis of methanol and ammonia from green hydrogen, followed by the inverse transformation to regenerate hydrogen for final applications [17]. Ammonia decomposition stands out as the most extensive method for hydrogen production from ammonia [18]. Regarding hydrogen generation from methanol, methanol steam reforming is considered due to its high conversion rates, selective formation of H_2, minimal CO generation, and manageable temperature control [19].

Within this framework, methanol and ammonia can be converted into power through two distinct pathways. The first approach entails a thermochemical conversion, where methanol or ammonia are combusted with air to generate power, typically in a gas turbine [20]. It is worth noting that the addition of a co-fuel may be necessary to address flammability concerns related to the combustion of methanol and/or ammonia [21]. In this area, Park et al. [22] evaluate the use of methane/ammonia and hydrogen/ammonia blends in a large-scale industrial gas turbine. A maximum ammonia ratio of 76% is proposed highlighting the need for a large-scale NO_x treatment to introduce ammonia as eco-friendly fuel. Su et al. [10] introduce an integrated scheme where a mixture of $H_2/N_2/NH_3$ is introduced into a gas turbine. The exhaust gases are used as heat source in ammonia craking and evaporation reaching a thermal efficiency around 35%. For methanol, Shen et al. [23] assess the combustion of this fuel in gas turbines. Methanol is partially decomposed to improve the combustion performance allowing a stabilized flame.

Alternatively, an electrochemical route can be employed, involving the feed of methanol or ammonia into a fuel cell. While this alternative holds great promise, it is essential to recognize that current efficiencies for direct methanol and ammonia fuel cells are relatively low [24, 25]. Vecchio et al. [26] experimentally evaluated a alkaline

direct methanol fuel cell. The highest energy density achieved was around 50 mW/cm^2. Different anode designs was proposed by Okech et al. [27]. In the best scenario, the new alternative anode pattern duplicates the energy density versus the reference design and allows a better evacuation of CO_2 in the unit. Liu et al. [28] studied a direct ammonia fuel cell with an anion exchange membrane with a peak power density of around 25 mW/cm^2. The authors have analyzed different operating parameters such as temperature, ammonia and KOH concentrations, flowrates, etc. The modeling perspective of direct ammonia fuel cells is investigated by Li et al. [29]. The use of solid oxide fuel cells (SOFCs) is primarily investigated for hydrogen applications [30], although new designs have also been proposed for ammonia [31] and methanol [32]. Different solid oxide materials and electrolytes are being tested to improve the performance of the system [33] and the search for low temperature conditions is also attracting attention [34]. For direct fuel cell units, substantial improvements in stability and durability are necessary to achieve full industrial implementation of this technology.

The second approach to generate power involves utilizing methanol or ammonia as hydrogen carriers. Initially, methanol is reformed, or ammonia is decomposed to release hydrogen. Subsequently, this produced hydrogen is employed to generate electricity, primarily through the use of a fuel cell. This alternative introduces an additional step for hydrogen production, potentially increasing system complexity and reducing energy efficiency. However, it is crucial to highlight that current hydrogen fuel cell technologies demonstrate significantly higher efficiency when compared to direct methanol or ammonia fuel cells, making this option currently attractive [35, 36]. Zheng et al. [37] evaluated four different catalyst (nickel, iron, copper and 304 stainless steel) for ammonia decomposition. The final objetive was provided the produced hydrogen into a SOFC. The best performance is obtained with nickel yielding a conversion rate of 55% at 1073K. A detailed reactor model analysis is proposed by Hu et al. [38] for methanol steam reforming. The composition requirements for different fuel cell technologies are met using the proposed methanol reforming reactor. Methanol conversion is mainly limited by the operating temperature and the steam to carbon ratio.

In the context of power production from methanol and ammonia using fuel cells, a significant trade-off emerges. On the one hand, methanol and ammonia can be directly

employed in direct methanol (DMFC) or direct ammonia (DAFC) fuel cells, offering a straightforward approach with minimal component involvement. This simple use of this hydrogen-based chemicals makes it suitable for both stationary and mobile applications. However, it is important to note that fuel cell devices for methanol or ammonia are still under development, requiring further research to enhance their energy performance. On the other hand, methanol and ammonia can serve as hydrogen carriers within the power production scheme. In this scenario, an additional section is introduced, involving ammonia decomposition or methanol reforming. This process is more complex, demands higher investment, and involves energy losses associated with the reforming/decomposition reaction. Moreover, the hydrogen produced can be utilized in a hydrogen fuel cell, a more mature technology with superior technical and economic performance. Presently, research has predominantly focused on two main areas. DMFC and DAFC have been investigated, especially, through experimental approaches, concentrating on fuel cell design and material selection for cathodes and anodes, typically at laboratory scale [39, 40, 41, 42]. For power production using hydrogen as intermediate, research has mainly focused on ammonia decomposition and methanol reforming since hydrogen fuel cells are a more mature technology. Various catalysts have been tested to optimize reactor design for hydrogen generation using ammonia decomposition [43, 44] or methanol reforming [45, 46]. Nevertheless, obtaining a comprehensive overview of the entire process is imperative to thoroughly analyze the performance of both alternatives and derive practical implementation results. This approach facilitates an equitable comparison among different fuels, leading to precise findings from both technical and economic points of view. To the best of the authors' knowledge, there is a notable absence of research in this area, in particular, from a process system engineering perspective. Therefore, this work aims to systematically analyze the use of methanol and ammonia as green fuels or hydrogen carriers to produce power from a process-scale perspective. In particular, five process alternatives have been considered: direct methanol fuel cell, methanol reforming coupled with a hydrogen fuel cell, direct ammonia fuel cell, ammonia decomposition associated with a hydrogen fuel cell, and a hydrogen fuel cell. The inclusion of the last alternative is proposed to compare the outcomes of using methanol and ammonia in fuel cell devices with the most mature

current system, which directly uses hydrogen in these units. Detailed reactor modeling for the methanol reforming and ammonia decomposition units are incorporated, enabling the derivation of precise results for these complex units. These alternatives have been evaluated from both technical and economic perspectives to provide comprehensive insights into the deployment of this power generation technology.

The remainder of the work is organized as follows. Section 2 provides a simplified description of the processes considered in this study to transform methanol, ammonia and hydrogen into power. Section 3 includes the main technical and economic results for the proposed alternatives. Finally, Section 4 draws some conclusions.

2. Process description

This work presents the assessment of two distinct approaches for harnessing methanol and ammonia in power generation using fuel cell technology. The first method involves utilizing these substances as green fuels by directly introducing them into the fuel cell. Alternatively, methanol and ammonia can serve as carriers for hydrogen. Consequently, the process begins with the production of hydrogen from these carrier compounds, followed by the feed of this hydrogen into a fuel cell. To enable a comparative analysis, an additional alternative is also explored, entailing the direct integration of hydrogen as a fuel source into a fuel cell. A scheme of these proposed alternatives is presented in Figure 1. The full process flow diagrams for the different transformation alternatives are included in the Supporting Information. In this section, a general description of the processes involved and the associated modeling is presented. Further details are also included in the Supporting Information file.

2.1. Methanol based alternatives

The first option involves the direct utilization of methanol as a fuel source through a direct methanol fuel cell (DMFC). This power generation scheme comprises two distinct sections: power generation and CO_2 separation. The power production process occurs within the DMFC, which features a proton exchange membrane [24]. It operates by utilizing an aqueous methanol solution (1 M) and air as its primary input streams [47]. This concentration is reached by blending inlet methanol (100%) with two recycled streams

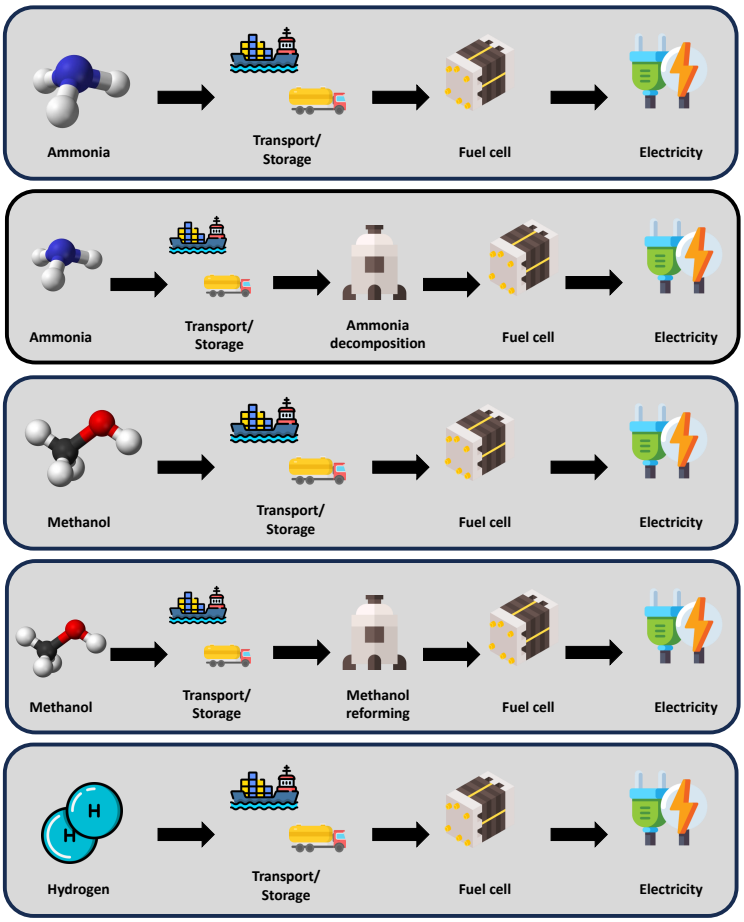

Figure 1: Overview of the analyzed alternatives

from the outlet gas treatment (formed by methanol and water). The methanol solution is fed at the cathode, where the following chemical reaction takes place:

$$CH_3OH + H_2O \rightarrow CO_2 + 6H^+ + 6e^- \tag{1}$$

Protons diffuse across the membrane and CO_2 is produced as main product. At the anode, protons undergo a reaction with oxygen (sourced from the air), resulting in the formation of water, as depicted in the following reaction:

$$6H^+ + 6e^- + \frac{3}{2}O_2 \rightarrow 3H_2O \tag{2}$$

The operating temperature for this device is set at 333 K, and both the incoming streams (an aqueous methanol solution and air) need to be heated to this temperature [48]. The unit operates at atmospheric pressure. As indicated by the outlined reactions, the effluent leaving the anode primarily consists of CO_2, water, and methanol. Additionally, the cathode yields a mixture of water, air, and CO_2. In this cathodic stream, water results from the chemical reaction occurring at the cathode, but there is also some water permeation through the membrane from the anode [49]. Furthermore, the presence of CO_2 in this stream is due to the methanol crossover within the membrane, as methanol migrates from the anode to the cathode, where it undergoes an oxidation reaction [50].

The effluent from the anode is subjected to separation through a flash system. The gaseous phase primarily consists of carbon dioxide and could be recycled for reuse in the synthesis of methanol. Meanwhile, the liquid fraction comprises water and methanol, which are the essential raw materials for the DMFC. A complete separation between the gaseous (CO_2) and the liquid (water and methanol) species is assumed. As a result, this liquid stream is combined with the fresh feedstock to feed the fuel cell. On the cathode side, the mixture of CO_2, N_2, O_2, and water is separated assuming the complete separation of the gas compounds. A fraction of the condensed water is recycled to adjust the final composition of methanol before the fuel cell unit.

The second methanol-based alternative involves using methanol as a hydrogen carrier. This process starts with the production of hydrogen by reforming methanol with steam. The subsequent step involves introducing the generated hydrogen into a fuel cell to produce electricity. Methanol reforming takes place within a fixed-bed, adiabatic catalytic membrane reactor, and the following reactions are part of this process:

$$CH_3OH + H_2O \rightleftharpoons CO_2 + 3H_2 \tag{3}$$

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \tag{4}$$

$$CH_3OH \rightleftharpoons CO + 2H_2 \tag{5}$$

The catalyst chosen for the methanol reforming reactions is $Cu/ZnO/Al_2O_3$ due to its favorable catalytic activity, rapid kinetics, high selectivity, and cost-effectiveness

[51]. The inlet operational parameters for this unit include a temperature of 600 K, a pressure of 10 bar, and a steam-to-methanol ratio of 1.5. To enhance the conversion of methanol and produce a higher-purity hydrogen stream for downstream applications, a Pd-Ag membrane is employed. This material is widely documented in the literature for such purposes [52]. Given the significant endothermic nature of these reactions, four reactors are employed in succession. Before each of them, a heat exchanger is set to rise the temperature up to the selected for the operation of the unit (interheaters between adiabatic reaction stages). Additionally, a portion of methanol is combusted with air in a separate chamber, and the resulting gases serve as a heating agent in the reforming section, providing the high temperatures required in the mentioned heat exchangers. The kinetic model used is taken from Peppley et al. [53] and is based on the Langmuir-Hinshelwood model. This choice is justified by the fact that the adsorption of reactants on the catalyst surface represents the rate-limiting step (see equations 6-8).

$$r_R = \frac{k_R K^*_{CH_3O(1)} \left(p_{CH_3OH}/p^{1/2}_{H_2}\right)\left(1 - p^3_{H_2} p_{CO_2}/K^{eq}_R p_{CH_3OH} p_{H_2O}\right) C^T_{S_1} C^T_{S_1 a}}{\left(1 + K^*_{CH_3O(1)}\left(p_{CH_3OH}/p^{1/2}_{H_2}\right) + K^*_{HCOO(1)} p_{CO_2} p^{1/2}_{H_2} + K^*_{OH(1)}\left(p_{H_2O}/p^{1/2}_{H_2}\right)\right)\left(1 + K^{*1/2}_{H(1a)} p^{1/2}_{H_2}\right)} \tag{6}$$

$$r_W = \frac{k^*_W K^*_{OH(1)} \left(p_{CO} p_{H_2O}/p^{1/2}_{H_2}\right)\left(1 - p_{H_2} p_{CO_2}/K^{eq}_W p_{CO} p_{H_2O}\right) C^{T^2}_{S_1}}{\left(1 + K^*_{CH_3O(1)}\left(p_{CH_3OH}/p^{1/2}_{H_2}\right) + K^*_{HCOO(1)} p_{CO_2} p^{1/2}_{H_2} + K^*_{OH(1)}\left(p_{H_2O}/p^{1/2}_{H_2}\right)\right)^2} \tag{7}$$

$$r_D = \frac{k_D K^*_{CH_3O(2)} \left(p_{CH_3OH}/p^{1/2}_{H_2}\right)\left(1 - p^2_{H_2} p_{CO}/K^{eq}_D p_{CH_3OH}\right) C^T_{S_2} C^T_{S_2 a}}{\left(1 + K^*_{CH_3O(2)}\left(p_{CH_3OH}/p^{1/2}_{H_2}\right) + K^*_{OH(2)}\left(p_{H2O}/p^{1/2}_{H_2}\right)\right)\left(1 + K^{*1/2}_{H(2a)} p^{1/2}_{H_2}\right)} \tag{8}$$

The specific values of the constants used in these equations can be found in the Supporting Information file. The flowrate passing through the Pd-Ag membrane is calculated using the following expression, which is dependent on the hydrogen pressure gradient [54]:

$$J_{H_2} = \frac{Pe_0 exp\frac{-E_a}{RT}\left(P^n_{H_2,ret} - P^n_{H_2,per}\right)}{\delta} \tag{9}$$

In equation 9, J_{H_2} represents the hydrogen flux through the membrane, while the associated parameters (Pe_0, E_A, δ, n) can be found in the Supporting Information. A

system of differential equations, encompassing mass, energy, and momentum balances, is utilized to describe the system, enabling the determination of the composition, temperature, and pressure profiles throughout the reactor.

The hydrogen produced, which passes through the membrane, is directed to the fuel cell section to generate electricity. This stage relies on the utilization of a solid oxide fuel cell (SOFC). The selection of SOFC technology is based on its high efficiency and cost-effectiveness [55]. Moreover, SOFCs operate at elevated temperatures, making them particularly suitable for stationary applications, aligning well with the proposed decomposition scheme. In this section, hydrogen from methanol reforming is heated up to 800ºC. The outlet gases from the fuel cell provide the required energy allowing for the heat integration of the process. Air is the second reagent to be fed and must be heated before being introduced into the fuel cell. In the SOFC, the following reactions take place. In the cathode:

$$\frac{1}{2}O_2 + 2e^- \rightarrow O^{2-} \tag{10}$$

Oxygen ions (O^{2-}) are collected by the cathode and injected through the ceramic electrolyte material toward the anode where the following reaction occurs:

$$H_2 + O^{2-} \rightarrow H_2O + 2e^- \tag{11}$$

The electrical energy efficiency of the fuel cell is set to 60% and the fuel utilization ratio (refered to the amount of fuel electrochemically oxidized within the anode compartment of the cell stack) to 80% [56]. The streams from the cathode and anode are sent to the combustor where the unreacted hydrogen is burnt increasing the temperature of the outlet gases. The resultant high-temperature stream from the combustion chamber serve a dual purpose: initially, they raise the inlet hydrogen's temperature and subsequently adjust the inlet air's temperature to match the conditions required by the fuel cell.

The exhaust gases from the combustor still contain a significant amount of energy after heating the inlet hydrogen and air. Therefore, in order to increase the energy performance of the system, a organic Rankine cycle (ORC) is introduced [57, 58]. The re-

frigerant R134a is selected and the properties have been taken from Ding et al. [59].

2.2. *Ammonia based alternatives*

The first alternative explored with regards to ammonia utilization is the direct ammonia fuel cell (DAFC). In this study, an alkaline membrane fuel cell has been chosen as it is specifically recommended for ammonia-based fuel applications, enabling operation at lower temperatures [60, 61]. Within this device, the introduction of ammonia as vapor occurs at the anode, where the subsequent reaction takes place:

$$2NH_3 + 6OH^- \rightarrow N_2 + 6H_2O + 6e^- \tag{12}$$

The hydroxyl ions are produced during the reaction at the cathode:

$$\frac{3}{2}O_2 + 3H_2O + 6e^- \rightarrow 6OH^- \tag{13}$$

Furthermore, a fraction of the ammonia permeates the anion exchange membrane (referred to as crossover), undergoing oxidation at the cathode to produce nitrogen oxide (NO). The operating temperature of this system is 368K with ambient pressure. The inlet air must contain a relative humidity of 50% (at operating temperature).

The cathode exhaust gases primarily consist of air, water, and nitrogen oxide. To reduce the concentration of this pollutant, a selective catalytic reduction (SCR) unit has been incorporated into the process configuration. This reactor requires ammonia and air as reagents. The gases from the anode are mainly ammonia, water, and nitrogen. In this case, the gases are cooled down to be able to separate ammonia as liquid. The ammonia is recycled to be used again as fuel in the power production device.

The second alternative utilizing ammonia comprises two stages: ammonia decomposition and hydrogen fuel cell. The ammonia decomposition reaction is performed in a fixed bed, adiabatic, catalytic membrane reactor with Ni/Al_2O_3 as catalyst. This is a good economic alternative to ruthenium catalyst with a good performance. The kinetic expression governing the ammonia decomposition in this reactor is derived from the

Temkin equation [62]:

$$r = 3k_{reac}\left[K_p^2 a_{N_2}\left(\frac{a_{H_2}^3}{a_{NH_3}^2}\right)^{\alpha} - \left(\frac{a_{NH_3}^2}{a_{H_2}^3}\right)^{1-\alpha}\right]\Phi\Omega \tag{14}$$

where a is the activity of the components and the constants are specified in the Supporting Information file. The incorporation of a membrane aims to facilitate the in situ separation of H_2, thereby enhancing the thermodynamic equilibrium conditions during ammonia decomposition. A Pd-Ag supported membrane is selected for this purpose [63]. The permeation rate of H_2 through the membrane is expressed as a function of the gradient of partial pressure on both sides [64].

$$r_{H_2}^p = \left(\frac{28.84 \times 10^{-5}}{\delta}\right) exp\left(\frac{-1888.381}{T}\right)\left(\sqrt{P_{H_2}^r} - \sqrt{P_{H_2}^p}\right) \tag{15}$$

The ammonia decomposition reaction operates at a pressure of 10 bar and an inlet temperature of 900 K. These conditions have been set based on a preliminary analysis of the reactor conditions using the rigorous model proposed. A trade-off arises regarding pressure levels, as lower pressures promote the ammonia decomposition reaction, while a higher-pressure gradient enhances hydrogen permeation. The pressure on the permeate side is set to atmospheric. As in the methanol case, the ammonia decomposition reaction is endothermic. Therefore, as in the previous case, a strategy employing a sequence of four reactors along with their respective heat exchangers has been implemented to supply the requisite energy for the reaction. As heating agent, a fraction of the inlet ammonia is burnt separately and the hot flue gases are used for heating purposes. The design and operation of this unit rely on a set of differential equations that encompass mass, energy, and momentum balances.

Hydrogen from ammonia decomposition is fed to the fuel cell section. As in the case of methanol reforming, a solid oxide fuel cell (SOFC) is selected with identical operating parameters as presented in the previous section. After the fuel cell device, the hot gases are used to heat up inlet hydrogen and inlet air to the fuel cell. After that, the remaining thermal energy is harnessed using an organic Rankine cycle, maximizing the utilization of available heat within the system.

2.3. Hydrogen based alternative

In this alternative, hydrogen serves as the inlet fuel, directly introduced into the fuel cell unit. The process proposed for this option is similar to the previous scheme for hydrogen when it is produced from methanol or ammonia. As previously mentioned, a solid oxide fuel cell is employed with identical reactions and operating parameters as in the preceding scenarios. The outlet gases from the fuel cell are burnt and, after providing the required thermal energy to the inlet hydrogen and air, are introduced into the ORC to increase the energy efficiency of the overall process.

2.4. Solution procedure and economic analysis

The different alternatives to produce electricity have been modeled and optimized using an equation-based approach. A nonlinear programming problem (NLP) is formulated and implemented in GAMS to determine the best operating conditions of the proposed systems under an economic objective function. During the economic assessment, the capital (CAPEX) and operating (OPEX) costs are estimated based on approximate correlations [65]. Regarding the CAPEX, the first step consists of calculating the total purchase cost of the major equipment within the proposed facilities. For the ammonia decomposition and methanol reforming reactors, the cost is estimated based on tree contributions: the steel vessel, the catalyst, and the membrane. In the case of ammonia decomposition reactor, the price of the Ni/Al_2O_3 is set at 30 €/kg [66] and the Pd/Ag supported membrane at 1500 €/m^2 [67]. In the case of the methanol steam reforming reactor, the price for $Cu/ZnO/Al_2O_3$ is also set at 30 €/kg [68]. For the fuel cell units, the unit price per kW of installed power is considered to be 1494 €/kW for direct ammonia fuel cells [69] and 1650 €/kW for direct methanol fuel cells [70]. For hydrogen solid oxide fuel cells, a baseline capacity of 100 kW with an associated cost of 480,000 € with an scaling index of 0.72 is selected [71]. For the SCR reactor, the selected catalyst is Pd/Al_2O_3 with a price equal to 2501 €/kg [72]. For the calculation of the operating cost, the main inlet parameter is the cost of the fuel. For methanol, the baseline price is set to 1200 €/t, for ammonia, 1060 €/t and, for hydrogen, 5 €/kg. These costs are selected based on the ranges proposed by Nemmour et al. [73]. Further details about the cost calculation methodology can be found in the Supporting Information file.

3. Results and discussion

This section begins by outlining the main operating results for the different processes analyzed to produce electricity. Section 3.2 contains comprehensive details regarding both the capital and operating cost of the proposed facilities. Within Section 3.3, a sensitivity analysis is presented focusing on the most important parameters. Finally, an overall evaluation including the transportation of the chemical and its transformation into power is presented.

3.1. Main operating variables

This section outlines the main operating variables obtained from the optimization procedure for the four distinct alternatives analyzed, utilizing both ammonia and methanol. In addition, the baseline scenario of a H_2 fuel cell is also presented for comparative purposes. These operating results are computed in all cases for a fuel cell with a power capacity of 1000 kW. In Table 1, some of the main operating results for the use of methanol and ammonia as hydrogen carriers are shown.

As outlined in the process description, both the ammonia decomposition and methanol reforming reactions are endothermic processes. Consequently, the management of the heat requirements stands as a pivotal aspect in the design and operation of these alternatives. In a vast extension of previous experimental works, the reaction is conducted in isothermal conditions, however, this situation is difficult to implement in real-world applications. Therefore, in this work, an multibed adiabatic system is considered. The primary challenge lies in supplying the necessary heat for reactions occurring at 600 K for methanol reforming and 900 K for ammonia decomposition. In the alternatives proposed, a fraction of the fuel is burned with air in a combustion unit and the hot flue gases are used as heating agent looking for a standalone system. Table 1 provides details on the proportion of fuel utilized for this purpose. The ammonia alternative requires a larger amount of fuel to be burnt due to the higher temperature requirements within the decomposition reactor. It is noteworthy that the significant thermal energy requirements of the hydrogen production involve that around 20-25% of the inlet fuel must be devoted for heating purposes. Therefore, the thermal management of the hydrogen production is essential with a direct relation with the global efficiency of the

process.

Table 1: Main operating variables for power production using methanol/ammonia as hydrogen carriers

Methanol/Ammonia Hydrogen Carriers		
Variables	Methanol	Ammonia
% inlet fuel to combustion	22.5	25.5
% H_2 recovery in the membrane	78.8	96.1
Final reactor conversion (%)	97.3	97.5
kg H_2/kg fuel	0.11	0.12
Fuel cell power (kW)	1000	1000
Net power (kW)	1106	1108
Energy efficiency (%)	35.8	42.3
Energy yield (kWh/kg)	2.00	2.18
Direct CO_2 emissions (kg CO_2/kWh)	1.29	-

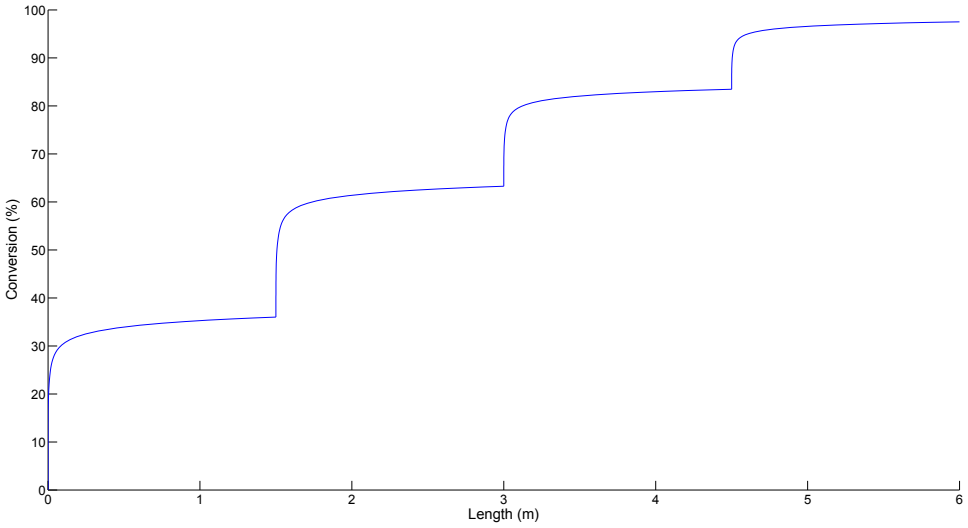

Figure 2: Conversion profile in the ammonia decomposition reactor

Under the conditions selected in ammonia decomposition (900 K and 10 bar) and in methanol reforming (600 K and 10 bar), the final reactor conversion reached a value of around 97% in both cases. This calculation was performed based on the initial flow rate of ammonia/methanol into the reaction section, taking into account the four reactors

along with their respective heat exchangers. The introduction of the four reaction sections along with the heating process among them allows for an almost full conversion of methanol and ammonia. The profile for the conversion of ammonia within the decomposition reactor is presented in Figure 2. A similar conversion pattern is followed in methanol reforming that, for the sake of brevity, is presented in the Supporting Information. The progression of the reaction conversion is mainly limited by the operating temperature. Initially, there is sufficient energy to facilitate conversion. However, as the reaction progresses along the reactor, temperature decreases, limiting the final conversion. The temperature profile is shown for the methanol reforming (as shown in Figure 3) and, for ammonia decomposition, is included in the Supporting Information. In the methanol reforming, the initial temperature is set to 600 K and, in the first reactor section, this value drops to around 450 K. Subsequently, after the heat exchanger, the temperature is raised in all reactor sections to 600 K as initial temperature. Nevertheless, due to the different reactor conversions in each section, the final temperature is different because the conversion decreases as the reactor progresses. For instance, in the last reactor section the final temperature is about 500 K. A similar pattern is obtained in the configuration for ammonia decomposition.

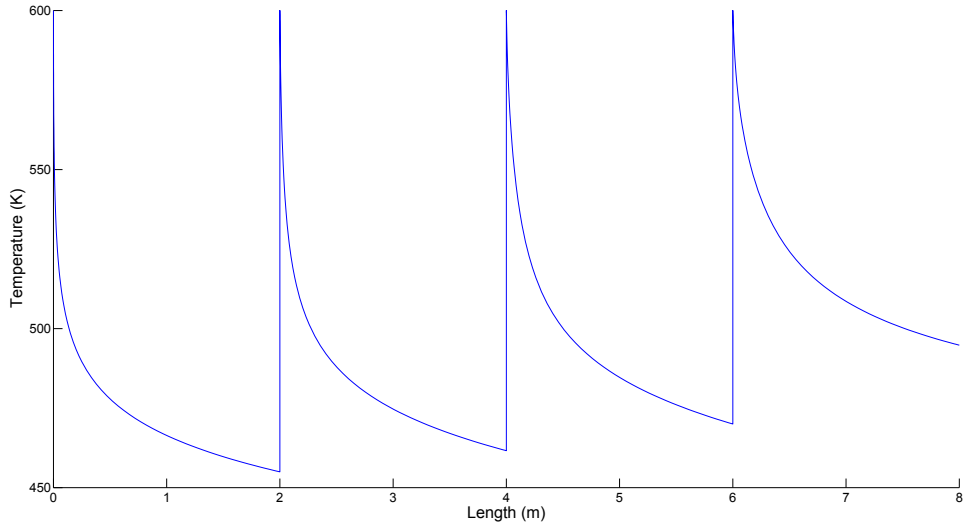

Figure 3: Temperature profile in the methanol reforming reactor

While both reactor units achieve nearly complete conversion of methanol and am-

monia, the recovery of H_2 differs significantly, as detailed in Table 1. In the case of methanol reforming, the recovery of hydrogen reaches about 79% whereas, in ammonia decomposition, the value is notably higher around 96%. This behavior is due to the different permeability properties of the species involved and the change in hydrogen partial pressures in both cases. These differences are reflected in the final yield of H_2 produced versus the total amount of fuel (methanol or ammonia) introduced in the process. In the case of methanol (via methanol reforming), the final yield stands at 0.11 kg H_2/kg methanol having a maximum potential yield (if all the hydrogen in the chemical is released and separated) of 0.19 kg/kg. On the contrary, for ammonia (using ammonia decomposition), the obtained yield is equal to 0.12 kg H_2/kg ammonia with a maximum level of 0.18 kg/kg. Therefore, the process of reaction and separation is more efficient in terms of hydrogen production in the case of ammonia (around 67% of the maximum) versus methanol (58% of the maximum). The outlet stream from the decomposition of ammonia that has not permeated through the membrane is mainly formed by nitrogen and some small amounts of hydrogen and ammonia. No additional treatment is conducted on this stream. In the case of methanol reforming, the outlet stream from the reactor is cooled down due to the easier condensation of methanol and water. The liquid phase is recycled to be mixed with the inlet stream and the gases are released.

All these results have a direct influence in the energy performance of the proposed systems. In all cases, the power capacity of the fuel cell is set to 1 MW, however, the net power production must consider also the power produced in the organic Rankine cycle (ORC) and the internal consumption within the process. This value is in both cases around a 10 % higher than the nominal capacity of the fuel cell. In terms of energy performance (considering the entire process), the thermal energy efficiency reaches a value of about 36% for methanol and 42% for ammonia. This difference is explained by the different production of H_2 between the two carriers. Ammonia decomposition (including the separation) is more efficient allowing a greater quantity of H_2 to be supplied to the fuel cell. These difference could be even larger but the higher operating temperature of ammonia decomposition involves a higher share of ammonia to combustion. Therefore, the amount of hydrogen is reduced with a lower power production rate.

Table 2: Main operating variables for power production using methanol/ammonia/hydrogen as green fuels

Methanol/Ammonia/Hydrogen Fuels			
Variables	Methanol	Ammonia	Hydrogen
Fuel cell power (kW)	1000	1000	1000
Net power (kW)	1000	885.7	1102
Energy efficiency (%)	26.5	16.8	52.9
Energy yield (kWh/kg)	1.46	0.86	17.6
Direct CO_2 emissions (kg CO_2/kWh)	0.94	-	-

In the case of the use of methanol and ammonia as green fuels (directly introduced into the fuel cell), the main results are presented in Table 2 including the comparison with the baseline case of hydrogen. Similar to the previous alternatives, all the results are based on a fuel cell with a power capacity of 1 MW. The net power varies across each alternative due to the different process configurations. In the case of methanol, the system is straightforward without additional units affecting power production or consumption. Therefore, the fuel cell power and the net power of the system are identical. In the case of ammonia, a compression step is required in the treatment of the gases from the fuel cell. Therefore, the net power is reduced versus the power produced in the fuel cell. Meanwhile, in the case of hydrogen, the inclusion of an ORC (similar to the methanol or ammonia carrier alternatives) generates additional power, thereby increasing the net power of the facility. In terms of energy efficiency, there are significant differences among the alternatives due to the different technologies selected for each fuel and the different degree of development of each one of them. In the case of methanol and ammonia, a low temperature fuel cell is included based on the previous experimental literature results. In particular, a proton exchange membrane fuel cell for methanol and an alkaline anion exchange membrane fuel cell for ammonia. These devices are still under development and the current efficiencies are low. In the case of methanol, the efficiency of the selected DMFC is equal to 23.3%, while in the case of DAFC the efficiency drops to 15.0%. This fact is reflected in the overall process energy efficiency with values of about 26% for methanol and 17% for ammonia. In the case of hydrogen, a vast extension of technologies is available with higher TRL levels. In

this work, a SOFC is selected for comparative purposes with respect to the H_2 carriers' schemes (previously presented). Due to the more mature status (with a fuel cell efficiency of 60%), the process energy efficiency rises to around 53%, two or three-fold the values of DAFCs or DMFCs. The efficiencies of the use of methanol or ammonia as green fuels are significant lower compared to the previous alternatives as H_2 carriers. In spite of the more complex structure (with the associated energy losses) in the use of these chemicals as hydrogen carriers, the current lower efficiencies of DMFCs or DAFCs make this alternative competitive. In terms of energy yield, the values of methanol and ammonia as green fuels show reductions of even more than 50% compared to the previous alternatives. Hydrogen has a particularly high gravimetric energy efficiency, therefore, the yield value is in a different order of magnitude. However, the flip side is the low volumetric energy density which complicates the adoption of hydrogen in certain applications.

Another crucial factor in power generation using alternative fuels is the level of carbon emissions. When considering fuels like ammonia (directly or via hydrogen as an intermediate) or hydrogen itself, no direct CO_2 emissions are anticipated. However, in the case of methanol, during reforming or within the fuel cell, CO_2 emissions are associated with this chemical. Typically, these emissions amount to around 1 kg of CO_2 per kWh of generated electricity, as outlined in Tables 1 and 2. Variations among different methanol technologies are due to differences in fuel cell efficiency and the combustion of a fraction of the fuel during methanol reforming. Following power generation, it is essential to address these CO_2 emissions through carbon capture technologies to enable a circular approach to carbon dioxide utilization.

3.2. Economic evaluation

In this section, a systematic economic evaluation of the different alternatives proposed has been performed. As baseline scenario, the costs of methanol, ammonia, and hydrogen have been taken from the ranges outlined by Nemmour et al. [73]. For methanol, a range of 800-1600 €/t has been proposed, with an average value set at 1200 €/t. Similarly, ammonia exhibits a proposed range of 720-1400 €/t, with an average cost of 1060 €/t. As for hydrogen, the cost ranges from 4000-6000 €/t, with an average cost

of 5000 €/t. In Figure 4, a summary of the capital expenditure (CAPEX) for the different alternatives is presented.

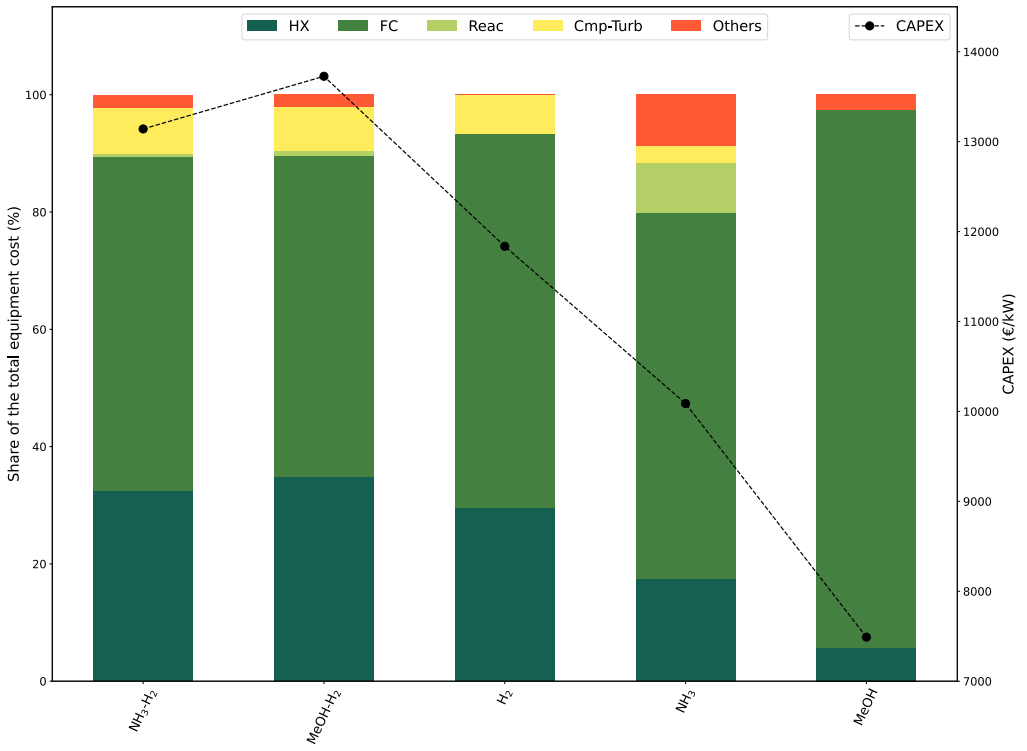

Figure 4: Total CAPEX and its distribution for the different power production pathways

The capital cost associated with the various analyzed ranges from 7,000 to 14,000 €/kW. As a general trend, the use of methanol/ammonia as hydrogen carriers involves a higher investment compared to direct devices. The introduction of the ammonia decomposition or the methanol reforming substantially rises the capital cost of these systems. Direct methanol fuel cell is the alternative with the lowest capital costs. This system operates in a more straightforward manner, incorporating only minor additions to the fuel cell unit. It is important to highlight that the alternatives with lower CAPEX tend to exhibit lower energy efficiencies, as presented in the previous section. Therefore, this trade-off is analyzed and clarified in the analysis of the operating costs (presented below). Regarding the distribution of the CAPEX, the fuel cell constitutes the central unit with a share of the total investment ranging from 55% to 92 %. This is especially

evident in direct alternatives, where methanol and ammonia are used as green fuels. Heat exchangers also represent a significant fraction of the total capital cost. These units play a crucial role in the decomposition alternatives to provide the necessary thermal energy in methanol reforming or ammonia decomposition. In addition, the systems that involve a hydrogen fuel cell (using the SOFC technology) include an associated ORC, increasing also the capital cost related to heat transfer processes.

These values of capital expenditure can be compared with other power production technologies. For example, with different renewable generation alternatives such as PV panels (810 €/kW), onshore wind turbines (1178 €/kW), biomass-based (1998 €/kW) or concentrated solar power (3950 €/kW) [74]. Or with traditional options such as natural gas combined cycle (924-1386 €/kW) or nuclear power generation plants (6470-7394 €/kW) [75]. Therefore, the capital cost associated to power production using methanol/ammonia with fuel cell units is significantly higher. Two primary reasons contribute to the higher capital costs associated with methanol/ammonia fuel cell systems. Firstly, the high capital cost related to fuel cells, which are still undergoing development, with anticipated cost reductions in the future. Secondly, the scale of operation plays a significant role. The current values for methanol/ammonia have been computed for 1 MW. For this small production capacities, the economies of scale have a relatively minor contribution (for example, in the hydrogen production using methanol reforming or ammonia decomposition). The CAPEX results obtained can also be compared with the use of methanol or ammonia for power production using a thermochemical pathway. This route is based on the combustion of the fuel using a gas turbine. Blanco et al. [7] analyzed this alternative and yielded a CAPEX value of 2250 €/kW for methanol and 3700 €/kW for ammonia. These values have been calculated for a power facility with a nominal capacity of 100 MW. The capital costs of fuel cell systems are higher than the ones obtained for thermochemical alternatives due to the high capital cost of the fuel cell system and the different capacity levels.

Figure 5 illustrates the Operational Expenditures (OPEX) for the various processes analyzed in this study. In contrast to the CAPEX, the operating cost of the alternatives based on the use of methanol/ammonia as hydrogen carriers is notably lower compared to the direct ammonia or direct methanol fuel cells due to the tradeoff between the ef-

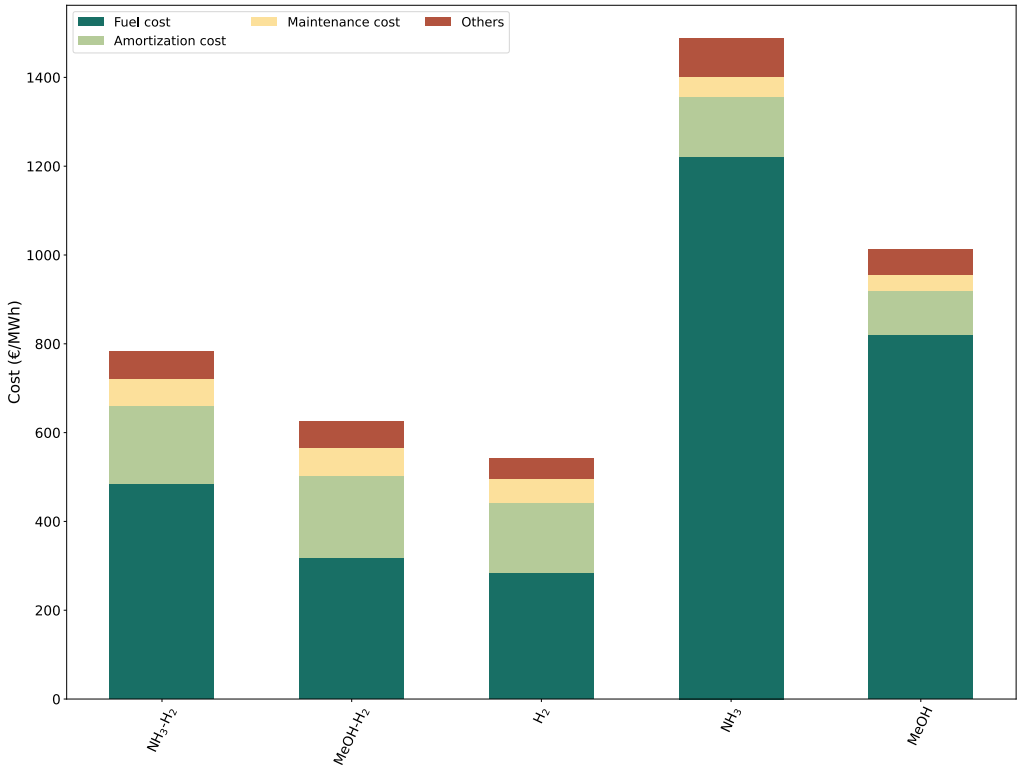

Figure 5: OPEX of the different alternatives with its breakdown

ficiency and the process complexity. For alternatives employing methanol/ammonia as hydrogen carriers, the cost is around 500-800 €/MWh and, for the green fuels direct options, around 1000-1500 €/MWh. Despite having a higher investment, the better energy performance of the hydrogen carrier options contributes to reduced operating costs for these alternatives. The most favorable alternative is the direct use of hydrogen (baseline scenario). This option avoids methanol reforming or ammonia decomposition, which reduce the energy efficiency of the system and increase the CAPEX of these alternatives. However, the main limitation about the use of hydrogen is its storage and transport conditions. Therefore, it is crucial to contextualize these results within the overall cost implications of utilizing different fuels.

As depicted in Figure 5, the main contributor to the OPEX is the fuel cost. This factor holds particular significance in the alternatives where methanol/ammonia are directly used as green fuels in fuel cells. Due to the reduced investment of these options, the fuel

cost emerges as the principal item accounting for more than 80% of the total cost. Conversely, in alternatives involving hydrogen, where the capital cost is higher, the amortization cost becomes the second-largest significant contributor. In these alternatives, the fuel cost represents a share of around 50-60%, and the amortization cost about 20-30%. Therefore, the reduction in the production cost of green methanol/ammonia will be the main driver toward a full implementation of these power generation alternatives as energy storage or energy carrier options.

Comparing the OPEX of these alternatives with the current cost of electricity from various power generation technologies provides valuable insights. For example, the current cost of electricity for some main renewable systems are: 60-100 €/MWh (solar PV), 40-80 €/MWh (onshore wind) or 90-170 €/MWh (biogas) [76]. Or, for the traditional generation sources: 120-200 €/MWh (coal) or 80-280 €/MWh (natural gas) [76]. As expected, the cost of these technologies is lower than the use of methanol/ammonia. These chemicals are employed as energy storage/carriers, hence, electricity is firstly produced from renewable sources, this electricity is devoted to chemical production and, as required, methanol/ammonia are transformed into electricity again. Consequently, the complete cycle incurs higher costs and lower efficiencies. However, these systems will be crucial to ensure the stability and robustness of a renewable energy system with high penetration of renewable sources. Schmidt et al. [77] analyzed the cost of electricity for different storage alternatives. These values are more comparable with the current proposed systems. Pumped-hydro has an associated cost equal to 185-278 €/MWh for short-terms applications, however, this value rises to 2778-3704 €/MWh for seasonal storage. Batteries show a storage cost of 370-556 €/MWh for short-term applications dramatically increasing to levels of 37037-41667 €/MWh for seasonal storage. In addition, Blanco et al. [7] evaluated the conversion of methanol and ammonia into power using a thermochemical route. For this alternative, a power cost of around 200-400 €/MWh is expected. The reduced cost is explained by the higher energy efficiency of these systems (around 30-35%) and the higher power production capacities, exploring the benefits of the economics of scale. Therefore, in this context, the use of methanol/ammonia emerges as attractive alternative, particularly, for seasonal storage.

3.3. *Sensitivity analysis*

As mentioned in the previous section, two parameters have a paramount importance in the profitability of the use of methanol and ammonia: the energy efficiency of the fuel cell and the price of methanol/ammonia. In this section, a sensitivity analysis about this two parameters in presented for the different alternatives analyzed in this study.

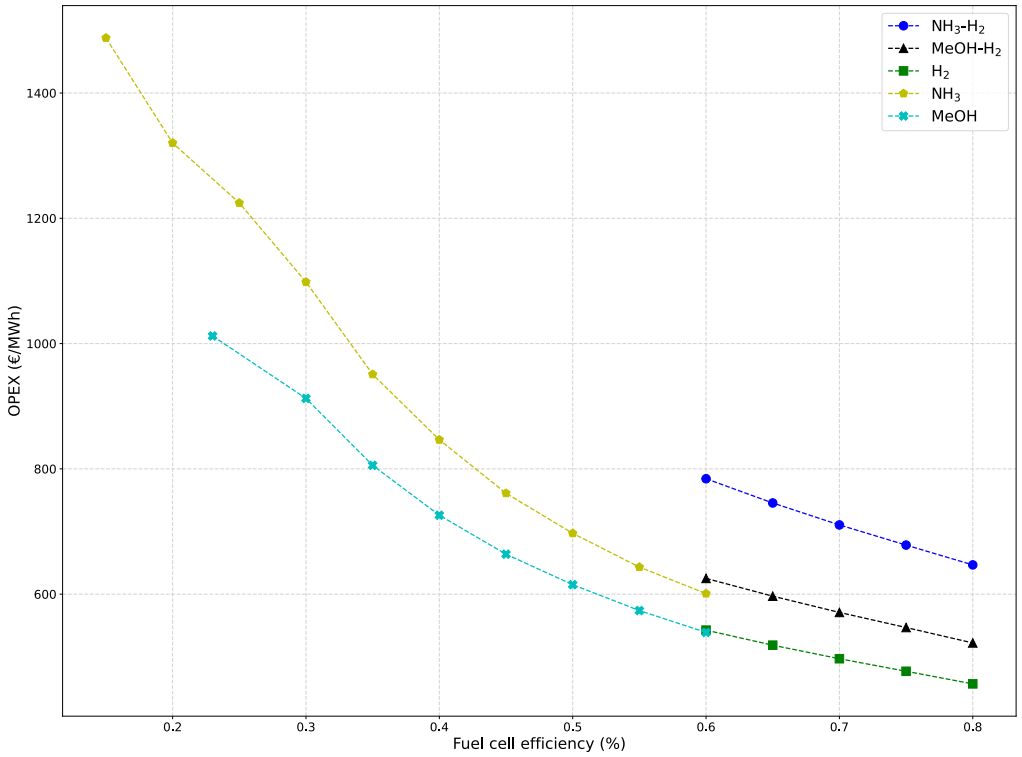

Figure 6: Operating cost of the different alternatives as a function of the fuel cell efficiency

Firstly, the assessment of the energy efficiency of the fuel cell is performed. Currently, there is a great difference between the levels of efficiency reach by the hydrogen fuel cells (around 60%) and those reach by direct ammonia (around 15%) or direct methanol (around 25%) fuel cells. Therefore, this large difference has a notably incidence in the economic performance of these alternatives. However, hydrogen fuel cells present a higher degree of development with efficiency improvements over the years. In the case of direct methanol/ammonia fuel cells, these are still in the early stages of technological development, with improvements expected in the next years. Therefore,

at this point, an evaluation of the influence of this variation in the efficiency into the operating cost of the full process is developed and presented in Figure 6. For the direct methanol/ammonia fuel cells, a range of energy efficiencies between their current levels and the efficiency of the current hydrogen fuel cell is considered. For hydrogen fuel cells, a range between 60% and 80% is selected because, due to its higher current development, lower efficiency improvements are expected. In the case of methanol, the current efficiency of direct fuel cell should reach a value of about 47% to be competitive with the scheme based on methanol reforming plus hydrogen fuel cell. In the case of ammonia this level correspond with a energy efficiency of the direct fuel cell around 43%. Therefore, current efficiencies would have to at least double to reach competitive levels. If these direct fuel cell efficiencies get the same level than their hydrogen counterpart, these alternatives show a better economic performance. Even, in the case of methanol, levels similar to the use of hydrogen as a fuel can be achieved.

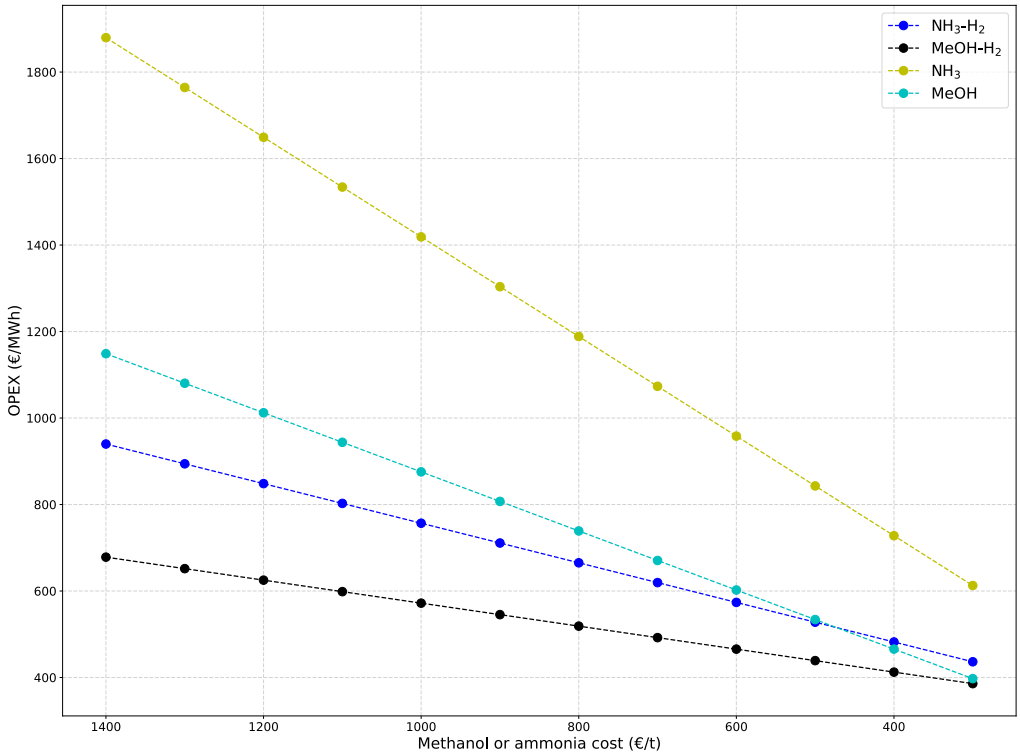

Figure 7: Operating cost of the different alternatives as a function of the fuel price

Secondly, the influence of the methanol/ammonia price in the final cost of electricity of the facility is analyzed. As presented in Figure 5, the fuel cost has a significant influence in the final cost of electricity. The cost of green methanol/ammonia are highly fluctuating with expected improvements in the next years [73]. The production route is a key factor determining the cost of green methanol/ammonia. For example, for methanol, Moioli and Schildhauer [78] calculated the cost of green methanol from biogas to be around 900 €/t. For Power-to-methanol, Pratschner et al. [79] computed a cost of 785 €/t. Methanol produced from biomass gasification has a lower cost, around 400 €/t [80]. In the case of ammonia, production costs vary based on scale and production technologies for Power-to-Ammonia, ranging from 800 to 1400 €/t [81, 82]. In addition, biomass-based alternatives reduce these levels to around 400-600 €/t [83]. Therefore in this analysis, a range between 1400 €/t and 300 €/t is established to cover the current ranges and the possible improvements in the future. For example, by 2050, a price of green methanol of 231-583 €/t is expected [84] and 285-350 €/t for green ammonia [85]. The results of this analysis are presented in Figure 7 using the current levels of energy efficiency. If fuel cost forecasts become true, the cost of electricity from methanol and ammonia using fuel cells will be significantly reduced. The lower the fuel price, the smaller the difference between direct and hydrogen-based alternatives. Even the direct methanol alternative can become more competitive in economic terms than those using H_2 as an intermediate. If the ammonia/methanol price is reduced below 400 €/t, the final cost of electricity can range between 400 and 600 €/MWh.

3.4. Transportation and power generation

In the previous sections, the results are circumscribed to the process itself. At this point, hydrogen as such shows the best economic performance with the lowest operating cost. However, the main limitations about the use of hydrogen come from its storage and transport conditions. Therefore, in this section, an integrated assessment of the transportation of these chemicals and their subsequent transformation into power (via fuel cells) is performed. The holistic assessment is based on a fuel cell with a power capacity of 1000 kW, as in the previous sections. According to the process evaluation, the amount of fuel required is calculated and this chemical is transported for a given distance (between 0 and 3000 km) using the different transport alternatives. To compute

the transportation cost for each chemical, an average value is taken from the literature for the different fuels. For hydrogen, the transportation cost is set to 6.8 €/t km for truck (as gas), 6.36 €/t km for truck (as liquid), and 0.66 €/t km for pipeline [86]. For methanol, the transportation cost is equal to 0.286 €/t km for truck, 0.025 €/t km for rail, 0.028 €/t km for pipeline, and 0.0015 €/t km for ship [87]. Finally, for ammonia, the cost is established at 0.33 €/t km for truck, 0.055 €/t km for rail, 0.026 €/t km for pipeline, and 0.0044 €/t km for ship [87]. Based on this transportation costs along with the production costs computed in this work, an overall cost in calculated for different transportation distances (as shown in Figure 8).

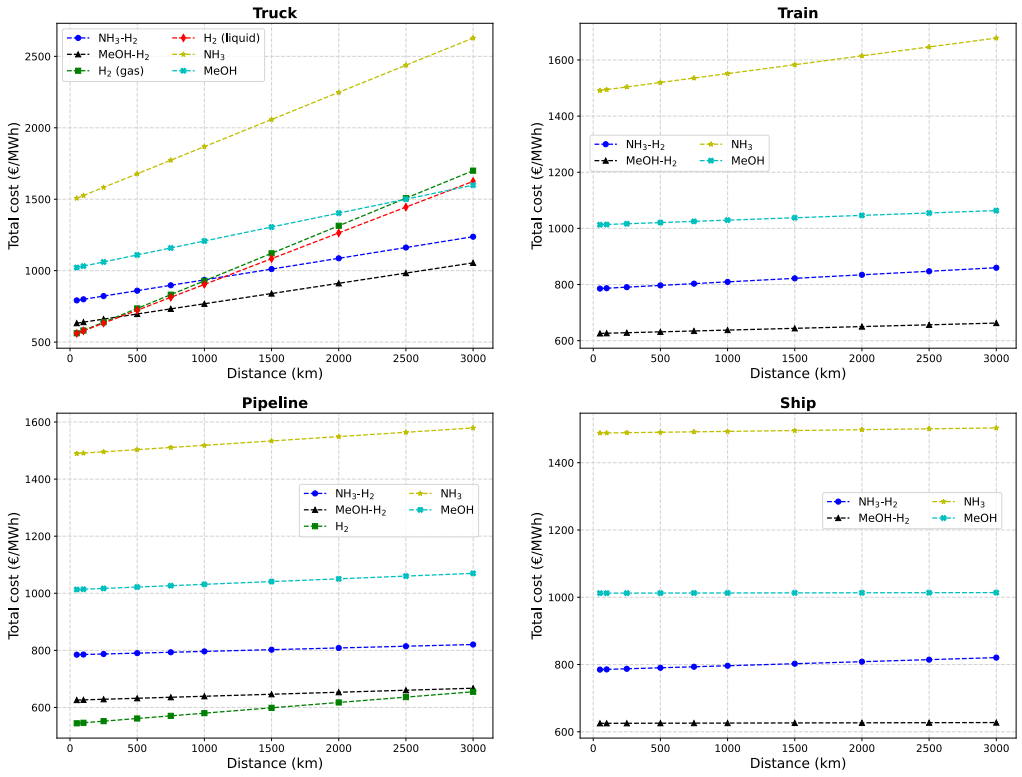

Figure 8: Total cost of transportation and production for different distances

For shorter distances, the percentage of the transportation cost in the total cost remains low in all cases. Therefore, as hydrogen shows the best operating cost in the transformation stage, this is the technology with the best performance. In the case of rail transport or shipping, the hydrogen transport is not considered due to the difficul-

ties associated and the lack of data for these alternatives. As distance increased, the transport costs associated with hydrogen experiment a notably increase, higher than in the case of methanol or ammonia. For example, when transported by truck over 3000 km, the transportation cost represents around 70% of the total cost for hydrogen. Consequently, the total cost of the system exceeds the cost values associated with ammonia and methanol. In the case of truck transport, the transition point is around 400 km, where the use of methanol as hydrogen carrier is recommended. Regarding pipelines, the hydrogen transportation cost is significantly lower than the case of truck, shifting the transition point to around 3000 km. Hence, methanol and ammonia demonstrate the great potential in applications where transportation and/or storage is critical. The use of pipeline is the alternative where the transportation cost is tractable in the use of hydrogen. However, the construction of this infrastructure involves a significant investment. For instance, a European initiative aims to connect Spain and Portugal to Germany via France for transporting green hydrogen. The Spanish Government estimates an investment of 2,500 M€ for the section (455 km) between Barcelona (Spain) and Marseilla (France). Therefore, 5.5 M€/km are required for this infrastructure [88]. Given the distances between Spain/Portugal and Germany, which is around 2500 km, this infrastructure could offer favorable operating costs, although at the initial stages of the transition region, aligning with the results of this study.

4. Conclusions

A comprehensive comparison between the utilization of methanol and ammonia as green fuels or as hydrogen carriers has been systematically conducted. The final goal in both cases is to produce electricity using the fuel cell technology. This comparison encompasses technical and economic evaluations of the various alternatives. The use of ammonia and methanol as hydrogen carriers shows a better technical and economic performance. The current H_2 fuel cell devices reach a notably high efficiency value compared to direct methanol or ammonia fuel cell. Despite of the introduction of the reforming/decomposition scheme (with its associated energy losses), the overall energy efficiency of these alternatives is about 35-40% versus the direct alternatives with values around 15-25%. This better efficiency is reflected in a lower operating cost (about 600-

800 €/MWh). The expected improvement in the values of efficiencies for the direct alternatives hold the potential to reduce production costs. However, the use of hydrogen as such is superior to the use of methanol or ammonia when only the power production process is considered. If the transportation cost are included, the use of methanol or ammonia emerges as a powerful alternative. Consequently, methanol and ammonia are poised to play a pivotal role, especially in scenarios where long-term storage and transportation are necessary.

Nomenclature

a_i	Activity of component i
$C_{S_i}^T$	Total surface concentration of each site (1, 1a, 2 and 2a)
E_a	Apparent activation energy for membrane
J_{H_2}	Flux of hydrogen
k_j	Rate constant for reaction j
K_j^{eq}	Equilibrium constant of reaction j
K^*	Adsorption coefficient
k_{reac}	Kinetic constant
K_p	Equilibrium constant
p_i	Partial pressure of component i
Pe_0	Permeability coefficient
r_i	Rate of the reaction i
$r_{H_2}^p$	Hydrogen permeation rate
R	Gas constant
T	Temperature
δ	Membrane thickness
α	Kinetic parameter
Φ	Effectiveness factor
Ω	Catalytic activity

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