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Hybrid Energy Storage and Hydrogen Supply Based on Aluminum—a Multiservice Case for Electric Mobility and Energy Storage Services

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The realization of a fully decarbonized mobility and energy system requires the availability of carbon-free electricity and fuels which can be ensured only by cost-efficient and sustainable energy storage technologies. In line with this demand, a techno-economic evaluation of aluminum as a cross-sectoral renewable energy carrier is conducted. The assessment, based on a newly developed process, involves the wet combustion of Aluminum at 700 °C resulting in heat and hydrogen (H_2) generation. The designed conversion plant enables the contemporaneous generation of electricity and on demand H_2 (up to 4 MW and 46.8 kg h^{-1}) with round-trip efficiencies as high as 40.7% and full recycling of the Al_2O_3 waste. This study, assuming the carbon-free production of Al and three different energy cost scenarios, proves the feasibility of the e-fueling station business case. The overall energy conversion including fuel production (power-to-Al), utilization (Al-to-power and Al-to-H_2), and recycling requires a capital investment of 5200 € per kW installed power without additional primary material demand. Hence, the estimated power-to-X cost for the Al-based H_2 is estimated in the range of 4.2–9.6 € kg^{-1} H_2, while wind and solar power based green H_2 production cost varies from 6.5 to 12.1 € kg^{-1} H_2.

1. Introduction

The decarbonization of energy generation and the mobility sector requires immediate action to increase the use of renewable energy technologies to cope with global warming.[1–3] At the same time, the system safe integration of renewables in the energy grid is strongly dependent on the flexibilization abilities of energy supply, transmission capacities and demand on all time scales (short term to seasonal or annual) as well as different system levels (decentralized and centralized).[4–8] This can only be achieved through the development of integrated storage and fuel systems that entail a wide set of different technologies covering different vectors (heat, fuels, and electricity).[9] Furthermore, an efficient development of cross-sectoral integration of the same is required to spur a sustainable energy transition.

Especially energy storage technologies are seen as important pillars for system flexibilization offering a high potential for sector coupling.[10] There are several technologies available as e.g. different secondary batteries (lithium-ion or redox flow batteries), mechanical energy storage (e.g. pumped hydro power or compressed air energy storage), and conversion of the renewable electricity to secondary energy carriers (i.e., power-to-H_2, power-to-methane, power-to-ammonia, etc.).[11–14] Batteries have proven to be the most suitable solution for short-term mitigation of fluctuations (excess and deficit of renewable generation) in the grid by offering a wide spectrum of grid services.[11–13] At the same time, for longer durations currently proposed energy carriers, PtX technologies are often referred as an ideal path for converting renewable and carbon-free electricity into fuels.[15] H_2 in this manner provide highest gravimetric energy density among other energy carriers but for longer storage durations its low volumetric energy density is limiting the applications, mainly due to large H_2 storage volumes and costs.[16]

In line with this, a strong market roll out of CO_2-free vehicles including battery electric vehicles (BEVs) and hydrogen-powered fuel cell electric vehicles (FCEVs—mainly for heavy-duty vehicles) is expected in the future.[17,18] In both cases

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Table 1. Gravimetric and volumetric energy density, earth abundance, and criticality of various metal and conventional energy carriers.[46,47] (☑:yes, ☒:no).

Energy carrier	Gravimetric energy density [kWh kg^{-1}]	Volumetric energy density [kWh L^{-1}]	Earth abundance[a)]	Secured supply[b)]
B	16.4	38.3	☒	☑
Li	12.7	6.8	☒	☒
Al	8.6	23.5	☑	☑
Fe	2.1	16.7	☑	☑
Na[d)]	5.9	5.7	☑	☑
Mg	7.3	12.6	☑	☒
Si	9.1	22.5	☑	☒
Ammonia (Liquified)	5.2	3.2	N/A[c)]	N/A[c)]
LNG	14.9	6.2		
H_2 (Liquified)	33.3	2.3		
H_2 (@ 700 bar)	33.3	1.4		

[a)]Source: ref. [45]; [b)]Based on EU's critical raw materials list[44]; [c)]Not available/applicable; [d)]Calculated based on Na wet combustion.[49]

(BEVs and FCEVs), an adequate charging infrastructure as well as hydrogen production and supply based on renewables is required. Besides BEV chargers in cities, shopping centers, and recreation areas, it will be necessary to equip and integrate the existing refueling stations located on highways to meet the demand of battery and fuel-cell electric vehicles. Expected substantial EV electricity demand is anticipated to cause problems (e.g., voltage instability, peak demand fluctuation, harmonic currents caused by fast charging infrastructure, transformer overheating). Also, hydrogen supply and safe on-site storage have to be assured. Hence, integration of distributed energy supply in combination with energy storage into the EV charging/refueling locations is presumed to be an appropriate solution to overcome raising issues.[19,20]

In this respect, abundant metal energy carriers (Al, Mg, Fe, etc.) are currently gaining increasing interest as alternative storage medium in the PtX context.[21–23] Mainly due to the fact that metals have significantly higher volumetric energy densities (also large H_2 storage capacities) in relation to proposed emerging energy conversion technology designs.[24–26] On that sense, supply of electricity and H_2 using metal energy carriers is thought to be an appropriate innovative concept for satisfying the demand of the developing EVs as well as FCEVs recharging/refueling infrastructure, on-site H_2 production and supply, and auxiliary grid services as well.[27] So far, besides use of metal energy carriers in batteries, slurry fuel mixtures, or as additive in propellants, several concepts have been developed which are employing pure metals as fuels in combustion systems or as anode material in secondary battery applications (i.e., aluminum-air batteries, magnesium batteries, etc.).[28–32] Among proposed concepts, designs using metals as electro-fuel (produced through power-to-metal (PtM) similar to PtX processes) mainly address the technical aspects for utilizing different metals for generating heat, electricity, and H_2.[28,29,33,34] Furthermore, conducted technical investigations imply high theoretical cycle efficiencies that makes metals appealing for further consideration.[21,35–38] Surprisingly, the economics of such conversion concepts have not been assessed in detail to provide insights about the economic feasibility and enable a comparison with other PtX technologies. Within this study, Al as an abundant and energy-dense metal is identified as a promising energy carrier for PtM applications, and the entire conversion chain (storage phase: Al production; Utilization phase: re-electrification and H_2 supply, including the recycling of the material) is techno-economically evaluated. In this respect, a sector coupling case via an industrial Al-smelting process (Hall–Héroult process), for the storage phase, and a hybrid Al-conversion unit for electricity as well as H_2 supply is analyzed. In particular, an explorative business case for grid flexibilization, BEV-load management and H_2 supply for FCEV is investigated based on different techno-economic scenarios.

2. Metal Energy Carriers

The energy storage demand needed to compensate the fluctuating and intermittent character of renewable power generation is anticipated to increase the growing demand for metals.[39–41] The availability of materials, their efficient use and recycling are key aspects when using metals to achieve the sustainable development objectives including the 2050 renewable energy transition targets.[42] Therefore, for the manufacturing of sustainable energy storage technologies assurance of the metal supply has a vital significance.[43] As a result, a sustainable supply oriented technology development is enforced by the material criticality reasons which is the main driving force of the high interest in abundant energy carrier metals.[44] In the quest for potential energy carriers, abundant metals and semi-metals are prospective candidates that match the circular economy concepts (see **Table 1**). Metals such as sodium (Na), iron (Fe), and aluminum (Al) are on research scope for the implementation of sustainable metal-based energy carriers.[45,46] Consequently, besides the earth abundance and secure supply of metals, energy density of the selected metal is also another important constraint in the energy carrier context. Of course, further aspects, such as toxicity, CO_2-emissions or social factors are relevant for the selection of suitable metals and have to be considered in future assessments.

As functional abundant metals, Fe and Al provide almost seven to ten (respectively) times higher volumetric energy storage densities than liquified H_2. These metals are produced in large quantities and are used in a wide range of applications. Hence, they are identified as the most appropriate alternatives in comparison with other metals. Within this scope, Na is also complying the selection constraints regarding the abundance and secured supply but it is highly reactive and it delivers significantly low energy densities with respect to other carriers, anyway greater than the H_2 in terms of volumetric energy density. As a monovalent reactive metal, metallic Na is determined to be more attractive to be used in electrochemical energy conversion applications to substitute Li. In particular, due to the recent advances regarding the commercialization progress of sustainable Na-ion batteries (SiBs).[50,51]

Regarding the use of pure metals as energy carrier, Fe possesses a relatively long development history. The conventional production of Fe is realized via the common practice iron ore reduction employing blast furnaces which is a carbon-intensive process using coke. The obstacles for enabling the use of Fe as a recyclable carbon-free metal fuel is mostly related to the limited favorability of electricity integration in the reduction process and the development of a suitable combustion technology, with improvements recently published in the literature.[32,52] In order to prove the concept of solid Fe particle combustion a 20 kW Fe fuel burner was developed in Netherlands in 2018.[53] Following the positive outcomes, based on the proven concept, the developed Fe combustor was scaled-up to 100 kW power capacity in 2020 with a Technology Readiness Level (TRL) of 4–5.[54] Further developments of the iron combustor up to 1 MW power and TRL 6 are expected in the near future, boosting the availability of metal fuel-based systems.[55]

Following the previously mentioned motivating advances in the metal combustion field, Al has garnered interest as a promising reactive metal energy carrier mainly because of high energy density and the high potential for its sustainable production. The common practice Al smelting process (electrolytic reduction) is using only electricity as an energy source.[33] Use of conventional carbon anodes increases the global warming potential of the process due to direct emissions. Nevertheless, it is remarked as, with implementation of the inert and dimensionally stable non-carbon anodes being currently tested in pilot plants, a significant improvement in the process is foreseeable in a short term.[56,57] Also a significant reduction in specific energy consumption up to 15–20% is anticipated in the foreseeable future thanks to the use of the wettable drained cathode technology.[58]

Moreover, Al has also been utilized in the industrial sector for the production of energy-intensive ammonia and steel, as well as in the power generation and transport industries enabling intersectoral applications.[33,59,60] Hence, Al appears to be a very promising energy carrier. In the subsequent sections the use of Al as a metal energy carrier is discussed, including a detailed techno-economic analysis.

3. Aluminum as Energy Carrier

Al as a functional construction and energy material can aid the achievement of the sustainable technology development goals with increased circularity. Al is produced via the electrolytic reduction of alumina (Al_2O_3) (Hall–Héroult process) at elevated temperatures, which is the production step of highest interest for the PtM concept.[61] The reduction process requires only electricity, which yields significant carbon intensity reduction and higher exergetic efficiencies if electricity is generated by renewable energy sources.[62] Presently, Al demand is dominated by the transport (27%), construction (24%), and packaging (15%) sectors, but an elevated demand for transport and energy applications is anticipated. The demand for Al in the car manufacturing sector in 2050 is expected to be 55% higher with respect to 2017.[63] According to the United States Geological Survey (USGS) 2021 report, the global primary Al production in 2020 was estimated at 65.2 million tons. The largest Al producer is China, accounting for about 57% of the world production, followed by Russia (5.5%), India (5.5%), Canada (4.75%) and the United Arab Emirates (4%).[64] The International Aluminium Institute (IAI) estimates that the global recycling input rate (RIR) of Al in 2018 was 32% which contributed substantially to the net demand.[65] The production of 1 tonne primary Al takes between 4 to 5 tons of bauxite ores, which global resources are estimated to range from 55 billion to 75 billion tons, i.e., the long-term availability of bauxite (and Al) is ensured, also located in the 'so-called' politically stable countries.[64] Thus, Al is an ideal candidate due to its abundance and well-established supply chain as an energy carrier, also offering higher safety than H_2 throughout the entire chain (production, storage and use).[24]

Regarding the Al-to-power conversion, Al can be oxidized at moderate temperatures (above 700 °C), with oxygen or water via dry or wet combustion processes (see Equations 1 and 2).[66] Upon oxidation with water, 4.2 kWh $kg^{-1}{}_{Al}$ energy is released as reaction heat, which can be converted into electricity using a heat engine. Additionally, 0.111 kg of H_2 are produced, corresponding to about 4.4 kWh $kg^{-1}{}_{Al}$ (higher heating value of H_2 is 39.4 kWh kg^{-1}) of thermal energy. Besides the direct heat generation, both processes deliver approximately the same energy content.[68]

$$\text{Dry Combustion: } 4\,Al + 3O_2 \rightarrow 2Al_2O_3 (\Delta H_{@298\,K,1\,atm} = -8.6\,kWh\,kg^{-1}_{Al}) \quad (1)$$

$$\text{Wet Combustion: } 2\,Al + 3H_2O \rightarrow Al_2O_3 + 3\,H_2 (\Delta H_{@298\,K,1\,atm} = -4.2\,kWh\,kg^{-1}_{Al}) \quad (2)$$

However, the dry combustion mechanism may possibly lead to NO_x emissions when air rather than O_2 is used as the comburant while the wet combustion is an emission-free process. Depending on the Al/water stoichiometry used, $Al(OH)_3$, $AlO(OH)$ and Al_2O_3 are the potential products of the wet combustion. However, Al_2O_3 is the most convenient combustion product from the thermodynamic point of view as well as for its ease of recyclability through the Hall–Héroult process allowing for the metal recovery without additional investments.[68,69] In this sense, recovered Al can be considered as secondary Al. Hence, the wet combustion process is of particular interest for electromobility, as it yields heat to generate electricity as well as H_2 that can be used either for generating additional electricity (via a fuel cell) or re-fueling FCEVs.[59,60] (see **Figure 1**) To elaborate more on the energy carrier properties of Al, one kg of Al offers a theoretical specific energy of 8.6 kWh, which compares well or is even better than other combustibles (e.g., Li = 11.9 kWh kg^{-1}, Mg = 6.86 kWh kg^{-1}, Fe = 1.36 kWh kg^{-1},

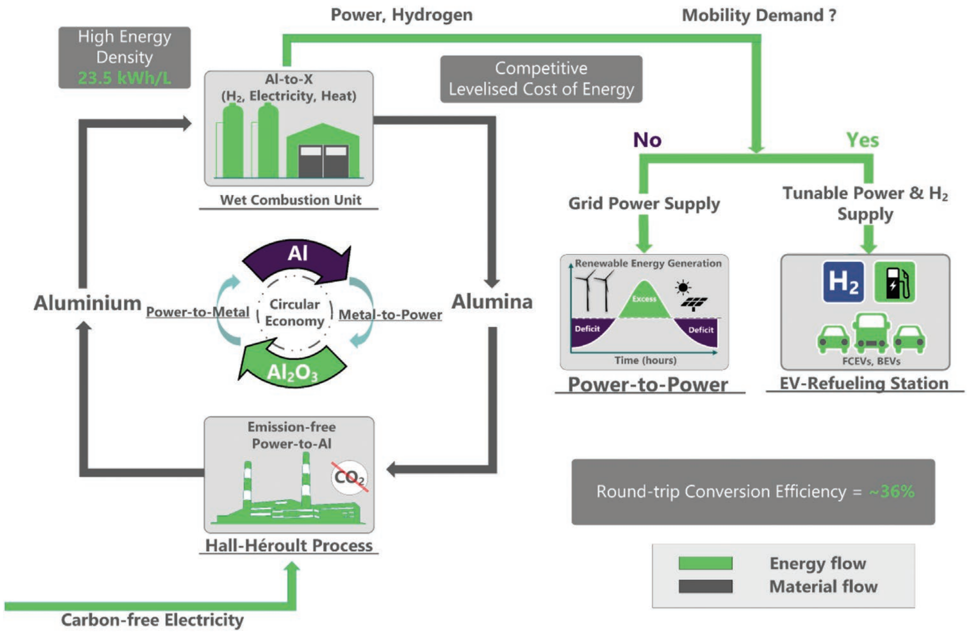

Figure 1. Aluminum as energy storage and carrier medium: circular and sectoral coupling aspects.

and CH_4 = 15.4 kWh kg^{-1}). Even though the gravimetric energy density of Al is lower than that of H_2 (but higher than most of the reactive metals, methanol, and ammonia), its high volumetric energy density allows to answer the long-term energy storage demand in a full decarbonized scenario. Moreover, its easy storability in open ambient conditions further contributes to long-term energy storage.[33,35]

Regarding the energy conversion efficiency, the Power-to-Metal path for the reduction of Al_2O_3 presently requires 14.2 kWh kg$^{-1}{}_{Al}$ of energy which represents the global average (China = 13.5 kWh kg$^{-1}{}_{Al}$).[70] However, the ongoing development of the new APXe and HAL4e Ultra electrolysis cells heralds an energy intensity ($\dot{E}_{Al}$) reduction to approximately 12 kWh kg$^{-1}{}_{Al}$ due to reduced current intensities.[71] Actually, the Norwegian Hydro Aluminium company reports an energy intensity of 11.5–11.8 kWh kg$^{-1}{}_{Al}$ as already achieved in a pilot industrial-scale Al smelting plant employing the HAL4e Ultra electrolysis cells.[72,73] In the Power-to-Al context, this value is a very significant metric influencing the conversion efficiency (i.e., round-trip efficiency).

Allowing for the circumstances, the system implementation is first modeled and evaluated for a typical highway refueling station, but it can be easily extended to large parking lots as well as commercial centers and large residential complexes. Most importantly, the overall approach can support full decarbonization of EVs and/or FCEVs operation using renewable energy surplus once the already decarbonization goals are achieved. In addition, the idle power capacity outside the recharge/refueling peak hours can be utilized to support the grid. Globally, this breakthrough concept offers a multifaceted bridging solution to solve the projected grid stability and EV refueling load management issues toward the renewable energy transition.[24]

The overall concept will benefit from the prospective improvements in the conventional Hall–Héroult production process, with the most important being the use of inert anodes, already regulated by the EU to achieve CO_2- and GHG-free Al production, and the implementation of the wettable drained cathode technology to reduce the anode–cathode distance and related process inefficiencies.[57,58,71,74] Nonetheless, thanks to the full recyclability of Al_2O_3, after first use energy and carbon intensity contribution from the bauxite mining, including the Bayer refining process will not be in question.[75] In a nutshell, the foreseen implementation of the emission-free Hall–Héroult process is the missing element of a carbon-free Al energy conversion cycle. Also, the proposed concept complies very well with the European framework of circular economy considering the whole life cycle of the material, because there is no need for intermediate transformation in the mentioned power-to-power (PtP) and PtX energy conversion pathways.[47]

Regarding the current state-of-the-art of Al combustion, several theoretical and early-stage experimental studies are available. The experimental studies mostly focus on the Al solid particle combustion and utilization of H_2 and steam mixture in expansion turbines.[26,30,59,60,76] In addition, conceptual combustion designs for co-generation employing Al as an energy carrier are also available in the literature.[36,77] Also, a Metal-to-Power system employing wet combustion, to exploit the full conversion of H_2 and thermal energy has been modeled by the authors with encouraging results.[33] The above mentioned Al-fueled system was demonstrated to offer a higher round trip efficiency (η_{RTE}) and superior energy storage performance with respect to a H_2-based PtP conversion system employing the PEM electrolyzer and fuel cell combination.[33] Herein a slightly modified system is presented consisting of a steam turbine, a solid-oxide fuel cell (SOFC) and a gas turbine with heat recovery sections installed downstream as presented later in Section 4.1. The techno-economic evaluation results of the proposed system is presented in order to enable a comparison with the other technologies employing renewable energy carriers along with the potential business case implications.

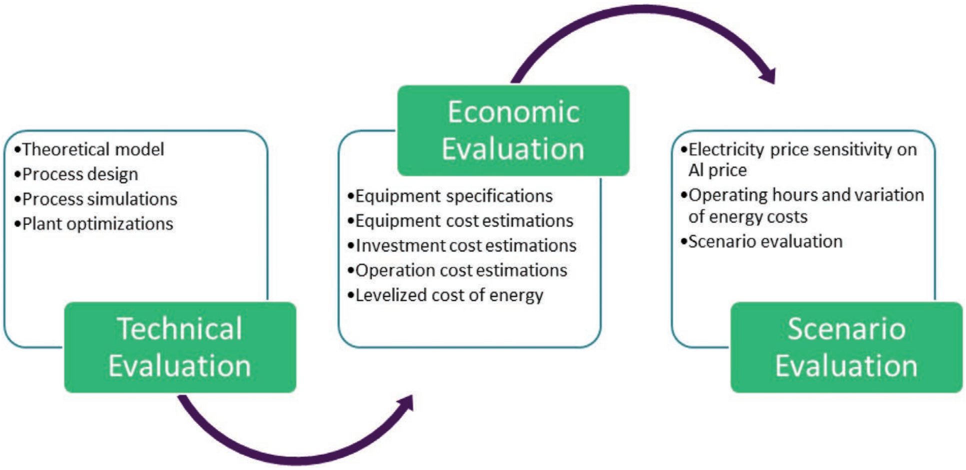

Figure 2. Methodology overview.

4. Methods: System Design and Techno-Economic Evaluation Methodology

First, a previously developed 0D process simulation model is optimized to assess different levels of H_2 production to meet the operational requirements of a refueling station operation.[33] Following this technical evaluation, an economic model is proposed to estimate the capital and operational expenditures of a conventional plant, using deterministic and probabilistic economic evaluation approaches as illustrated in **Figure 2**.

The cost of the provided energy (electricity and H_2) depends on the operating conditions of the refueling/recharging station shaped by the EV's electricity and FCEV´s H_2 demand. Also, the provision of services to the grid in case of low demand from mobility can impact the cost. The Al-to-P system must operate flexible to meet the varying demands for H_2 and electricity (either for charging or the grid). Considering the present EV market forecast, Al-to-H_2 conversion factors of 0, 10% and 17.5% are assumed depending on the SOFC load partition from 100% (only electricity is generated), to 80% and 65% (diverting partial H_2 flow to the storage), respectively.[18,78,79] The process economics and cost sensitivity for these three operating conditions have been evaluated also taking in consideration based on three different energy scenarios:

- *Scenario-I High Energy Intensity*: Al is purchased at the standard commodity prices, but excluding the carbon electrode costs. Under this assumption, the Al cost is calculated using the 2020 global average energy requirement of 14.25 kWh per kg of Al and the electricity price to be 50 € MWh_e^{-1}.
- *Scenario-II High Efficiency and Low Electricity Price*: Energy efficient smelting process with prospective lower energy intensity ($\dot{E}_{Al}$ = 11 kWh kg^{-1}_{Al}) are assumed together with a lower electricity price (30 € MWh_e^{-1}) due to higher penetration of renewables in the grid.
- *Scenario-III Zero Electricity Price*: This scenario assumes the frequent availability of zero or even negative electricity spot market prices stemming from large renewable generation surplus and a specific energy requirement of 11 kWh kg^{-1}_{Al}. Although, the frequency of negative electricity prices is anticipated to increase in the future, for this first exploration it is assumed that the energy for Al production can be purchased free of charge while negative electricity prices are not considered. Negative spot market prices, represent a very narrow timeframe (97–211 hours with negative prices observed in Germany within 2016–2019, +123% increase in 2019 in a six-month period consecutive negative price observations).[80]

4.1. Technical Evaluation

The performance of the energy storage system fed by Al (delivered by the smelting process at 900 °C or as in situ grinded solid particles) has already been evaluated employing the wet combustion reaction.[33] Section 5.1 presents the results obtained in the Al-powder feeding case, hence this is easier to implement in EV fueling stations due to operational reasons. The operation strategy hinges on the delivery of inactive Al to the plant, and on-site pulverization to ensure the maximum safety. Noting that the Al powder is highly reactive therefore explosive when exposed to air, and molten Al transportation is also a more complex operation. The investigated system consists of an Al combustor, a high temperature fuel cell (SOFC), a steam turbine (ST) and gas turbine (GT) bottom cycles (see **Figure 3**). The wet combustion products are H_2 (utilized in a SOFC) and heat, which is removed to control the combustor temperature. A secondary water-cooling circuit is implemented to produce superheated steam exploitable in the ST for the power generation. With respect to the previous work, part of the H_2 can be also exploited to refuel FCEVs, for which a five-stage H_2 compression section is also included.[33] However, most of the H_2 (from 65% to 100%) is used in the SOFC to produce electricity. The exhausts from the SOFC (unreacted H_2) are burned and sent to a GT for power recovery. Two heat recovery sections, thermally integrated to the Al combustor cooling and the SOFC cathode feeding circuits, respectively, are installed downstream the GT.

The technical performance of the system is evaluated in terms of metal conversion and round-trip efficiency (RTE), i.e., the efficiency of the global PtX process considering also the electrical consumption required for Al production. In

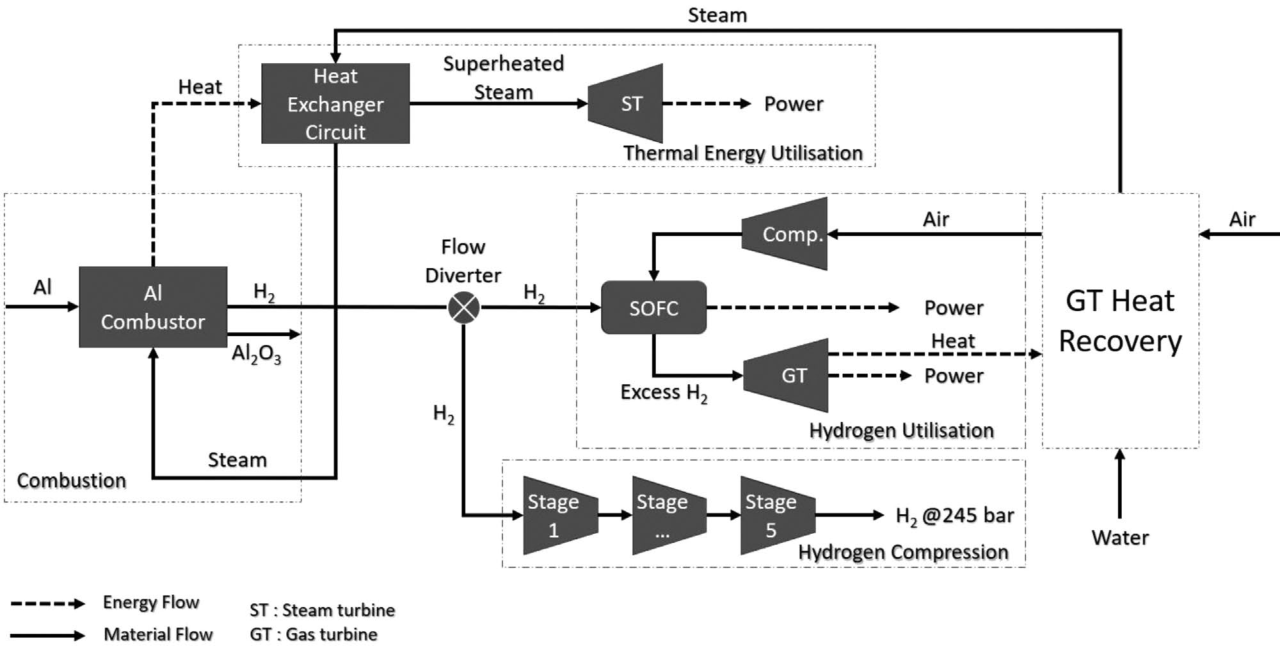

Figure 3. Simplified plant layout showing energy and material flows.

this regard, 11 kWh kg^{-1}_{Al} (39.63 kJ g^{-1}_{Al}) is considered as the electricity consumption for Al production. Such a value corresponds to a reasonable 15% reduction of the specific consumption exhibited by current best practice of Hall–Héroult electrolysis cells (ca. 13 kWh kg^{-1}_{Al}) resulting from the implementation of the wettable drained cathode technology, as remarked by Moya et al. in 2015.[81] Moreover, further improvements are expected to result from the implementation of inert and dimensionally stable noncarbon anodes, as previously mentioned. Inert anodes have been implemented in pilot plants and commercial inert anodes for retrofitting the existing electrolysis cells, which are set to be made available by 2024.[57,82] Finally, it is remarked that the assumed specific energy consumption for Al production (11 kWh kg^{-1}_{Al}) is only slightly lower than the one already achieved in Norway.[72] Based on the given assumptions, the modified system is evaluated from a technical perspective.

4.2. Economic Evaluation

In addition to the technical performance evaluation, a business case for the purchase of Al from the producers is envisaged to conduct an economic evaluation of the capital and operational expenditures as well as the levelized cost of energy (LCoE, and LCoH). The business case grounds on the purchase of Al from the producers and transportation to the hybrid refueling stations while the combustion product Al_2O_3 is returned to Al producers for recycling. Here the multifaceted point is the fuel production and purchase agreement between Al producers and plant operators. Hence, different considerations are made for the purchase price of Al, commodity value of Al_2O_3 and the electricity price. Since the energy storage system aims the residual load compensation, excess renewable energy surplus with low spot market prices creates a business case for the Al production, which are expected to occur more frequently in future.[83]

Based on this business strategy, the economic evaluation of the proposed system is carried out using deterministic and stochastic approaches to address the uncertainty in the results. In fact, the development of the technology is still at the research level and such a business case does not exist. One of the major problems is that the system is known in general terms based on the simulation model and the variation of the design causes an uncertainty in the capital investment estimations. However, using standard cost estimation methods it is possible to estimate the capital investment requirements with a ±30% uncertainty.[84] Moreover, taking into account the system equipment specifications based on the process design, the required investment for the equipment is estimated using learning curves for each specific process equipment (i.e., steam turbine, gas turbine, SOFC, heat exchangers, pumps, separators, and others).[84,85] The learning curve estimates are rescaled according to the equipment specifications via scale exponents provided in the literature.[86] Following the equipment cost estimations, installation factors are used to estimate the installed equipment cost, and introduced in the model[84] (see Table S2, Supporting Information). The obtained cost estimations yield cost intervals for each equipment accounting for the uncertainty using the given methods. These intervals form the bandgap of the deterministic inputs, later converted into probability distributions via the developed stochastic model. Finally, other capital investment costs such as engineering, procurement and construction (EPC) and 15% working capital (WC) is assumed to quantify the total depreciable CAPEX (Equation S1, Supporting Information) consisting of direct and indirect CAPEX.

The estimated depreciable total CAPEX is then used for estimating the cost of fixed assets and the salvage value of the assets using the Modified Accelerated Cost Recovery System (MACRS) rates for the given investment period.[87] As a rule of thumb, assuming an investment period of 20 years, the system economics are modeled for determining the operational expenditures (OPEX) coming out of different operation modes (0%, 10%, and 17.5% Al to H_2 mass conversion rates corresponding to 100%, 80%, and 65% SOFC loads expressed as percentage of the rated power) and varying capacity factors (or full load hours (FLHs)). The OPEX is a composition of fixed and variable operational expenditures (see Equations S2 and S3, Supporting Information). Therefore, to estimate the fixed operational expenditures ($OPEX_{fix}$), the fixed cost components such as fixed operation and maintenance (O&M) cost, fuel cost (in this case Al), and insurance expenses are considered[88,89] (see Table S4 for the OPEX estimations, Supporting Information).

For the Al-based system, a revenue from the sale of combustion product (Al_2O_3) is estimated and deducted from the Al cost, as the Al_2O_3 has an economic value being the primary material used for Al production. In the 2014–2019 timeframe the Al commodity price ranged between 1.2 and 1.95 € kg^{-1}[90] (see Table S3 and Figure S3, Supporting Information). For the quantification of different price components, 43% for Al_2O_3, 24% for energy, 17% for the carbon anode, 11% for the fixed costs and 5% for other production related costs are assumed as reported by the Norwegian aluminum producer, Norsk Hydro.[91] Thus, the full recyclability of Al_2O_3 brings along benefits from both the environmental and economic point of views.[92] The economic value of Al_2O_3 ranges between 0.5 and 0.8 € kg^{-1} of Al equivalent (1.89 kg Al_2O_3 are needed to produce 1 kg Al).[62] Thus, the recycling activity is taken into consideration as an income that can yield approx. 43% reduction in the Al price. In addition to that, since the case is based on a carbon-free Al, implementation of carbon-free inert anodes in the Hall–Héroult process may enable up to 17% cost reduction, as the carbon demand is eliminated. Hence, a 43% cost reduction of Al can be taken as granted, while a further prospective 17% reduction can be considered for the prognosis due to use of inert anodes, thus a total of up to 60% reduction of the Al price. Considering the second cost voice, i.e., energy, its share is usually reported in the range of 23–45% of the Al price and, specifically, equal to 24% in the Norwegian producer's case.[91,93,94] Noting that the energy intensity is assumed 11 kWh kg^{-1}_{Al} in this study. Al production in Norway also takes advantage of the high share of cheap hydropower electricity resulting in an electricity price of about 30 € MWh_e^{-1}[95] (see Table S5, Supporting Information). Assumption of an average electricity price of 30–50 € MWh_e^{-1} for the strategic industries is in line with the forecasted German scenarios for 2050.[96] The system equipment is classified under three categories as thermal, fuel cell and heat recovery. Hence, the fixed OPEX is estimated using the capital weighted cost allocation method and generation share of each conversion section. Instead, the variable OPEX is added based on the selected equivalent FLHs, and annual H_2 and electricity generation in the reference year. For the overall system life these costs are assumed for each year based on the introduced cost inflation rates (see Equation S2 explanation, Supporting Information). As a measure of the economic performance of the storage cycle, the levelized cost of energy method is used. Since the system outputs are electricity and H_2, the final cost of the supplied energy is represented with the levelized cost of energy (LCoE) in € kWh_e^{-1}, and levelized cost of H_2 (LCoH) in € kg^{-1}_{H2}. The general definition of the levelized cost of energy connotes the ratio of total net present value (NPV) of expenditures and NPV of supplied energy.[97] The NPV is obtained discounting the sum of all the expenditures with a rate of 2.3%.[98] Consequently, using all the given assumption and methods, the LCoE and LCoH estimation results for the given scenarios are presented in Section 5.2 addressing the baseline costs, 10th and 90th percentiles of the probabilistic estimation results.

5. Results

5.1. Technical Evaluation Results

In this section the technical performance aspects of the system are introduced utilizing the specifications indicated in the methods section. The Al-powder fueled power plant is developed on the basis of the system layout previously designed by the authors.[33] The modified system design allows simulations under different SOFC part load conditions to enable H_2 production for a refueling station. According to the electricity and H_2 demands, the SOFC partition allows part of the H_2 stream to be diverted to a 245-bar storage tank typically employed in refueling stations. Specifically, a maximum SOFC stack partial load operation down to 65% of the nominal power (2000 kW) is considered, which has been proven to be feasible.[99] In fact, aiming to reduce thermal stresses, it is possible to maintain a constant SOFC operating temperature within the 65–100% partition range, via a suitable management of the cathode air stream as successfully implemented in a hybrid SOFC/GT power plant.[100] The fuel cell working parameters under partial operating conditions have been set as detailed below based on the above-mentioned studies. For a preliminary analysis, two partial load operation points of the SOFC have been considered: 65% and 80% of the 2 MW rated power. Also, a new compression section has been added for the storage of H_2. For a detailed understanding regarding the process streams, the readers can refer to the Supporting Information. In **Table 2**, the simulation results for the three

Table 2. Plant technical specifications and performance.

SOFC part load	SOFC power [kW]	GT power [kW]	ST power [kW]	Compression required power [kW]	Total power [MW]	H_2 flow rate [kg h^{-1}]	η_{M-P}	η_{M-X}	η_{P-X}
100%	2000	906	1064	0.0	3.9	–	81%	81%	35.6%
80%	1600	683	916	53.2	3.1	28	65%	88%	38.8%
65%	1300	520	884	89.0	2.6	46.8	54%	93%	40.7%

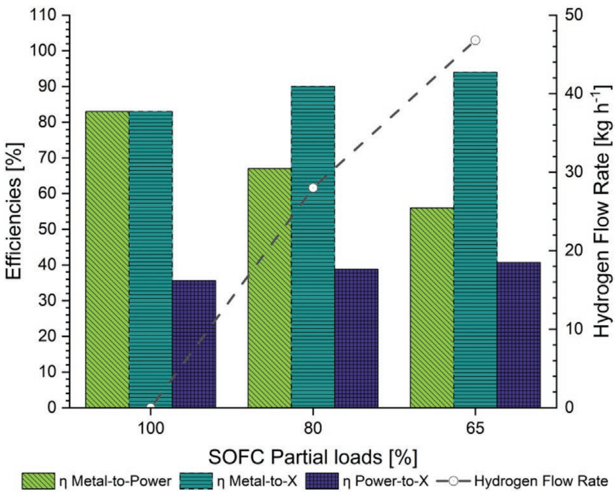

Figure 4. Efficiencies and H_2 stored flow rate versus SOFC partition.

SOFC operating points (65%, 80%, and 100% nominal power) are presented.

The H_2 flow rate sent to the storage tank corresponds to 28 and 46.8 kg h^{-1} for the utilization of the SOFC at 80% and 65% of the nominal power, respectively. For the sake of completeness, the values of the electric power absorbed by the compression stages and the system efficiencies are also provided. Specifically, the MtP ($\eta_{M\text{-}P}$) and MtX ($\eta_{M\text{-}X}$) efficiencies (**Figure 4**), the latter also including the produced H_2, are calculated. Moreover, a different trend between $\eta_{M\text{-}P}$ and $\eta_{M\text{-}X}$ arises as the partition increases. This behavior is explained by the different impact of the energy, stored as H_2 or generated as electricity in the SOFC, on the overall efficiency calculation.

Globally, based on the MtX efficiency, also the PtX ($\eta_{P\text{-}X}$) has been assessed considering the specific energy consumption ($\dot{E}_{Al}$) of 11 kWh kg^{-1}_{Al} according to the details provided in Section 6.

5.2. Economic Evaluation Results

Based on the technical evaluation results and design specifications, a detailed economic evaluation is conducted to estimate capital expenditures (CAPEX), operational expenditures (OPEX), and the levelized cost of provided energy and hydrogen (LCoE and LCoH) using stochastic approaches. Considering the direct and indirect capital requirements (equipment cost, installation, engineering, and procurement, and working capital), the estimated total cost of capital expenditures corresponds to 21 M€ (baseline) following the assumptions provided in Section 4 and Supporting Information. Considering the uncertainties in the equipment and other capital related costs to make the plant operational, this cost is in the range of 16–25 M€, i.e., 4200–6200 € (Figure S2 and Table S1, Supporting Information) per installed kW power capacity, stemming from the equipment purchase and installation.

As the OPEX is highly dependent on the selected business case and annual equivalent operation hours, operation related costs are considered separately based on different SOFC part loads. As explained in Section 4.2, the capital weighted cost allocation method is used to estimate the OPEX more precisely. Thus, system components are classified for power generation and H_2 supply separately, and the weightings are estimated for the thermal system (steam turbine, heat exchangers, pump, combustor, and solid separator), H_2 supply (SOFC, heat exchangers, air and H_2 compressor) and heat recovery (HR) (heat exchangers, gas turbine, and air compressor). Some of the synchronous equipment (air compressor, combustor, etc.) costs are apportioned according to power generation share of the specific system sections. Detailed equipment specifications and estimated costs are given in the Supporting Information. The operation strategy of the design stipulates a minimum SOFC partial load of 65% due to the reasons explained in the technical evaluation results. To interpret this technical design criterion into economics, it should be considered that even though there is no electricity demand on a specific time, 65% of the generated H_2 must be utilized for electricity generation. Hence, the operational costs arising from the electricity generation via fuel cell and HR are allocated to the H_2 cost based on the cost allocation weightings (**Table 3**).

The thermal system supplies only power, but the given 1% and 2% weightings at 80% and 65% SOFC utilization rates correspond to the expenses due to deficiency of the thermal system as a result of the partial operation of the fuel cell and H_2 supply system, and HR section. Consequently, the allocated fixed and variable costs are considered to determine the cost of the supplied energy. The required parameters for the allocation of the capital and operational expenses are integrated into the economic model.

A first estimation of the LCoE and LCoH is conducted for a base case assuming 4000 full load hours (FLHs) of operation and using the carbon-free-adjusted Al price ranging from 1.28–1.7 (μ = 1.48) € kg_{Al}^{-1} (based on $\dot{E}_{Al}$ = 11 kWh kg^{-1}, electricity price: 50 € MWh_e^{-1}). The 4000 FLHs value is selected for comparison with other PtX technologies as reported data is often referring to this value and it corresponds to the medium flexibility.[101–105] Based on this assumption, LCoE and LCoH (see **Figure 5**) are estimated for different part loads.

Table 3. Estimated cost allocation weightings for different SOFC part load rates for the electricity generation.

SOFC part load		100%	80%		65%	
Power system sections	System investment	Power	Power	H_2	Power	H_2
Thermal system	€ 2 836 183	21%	20%	1%	19%	2%
Fuel cell & H2	€ 7 931 623	59%	41%	18%	28%	31%
Heat recovery (HR)	€ 2 601 938	19%	16%	4%	13%	7%
TOTAL	100%	77%	23%	60%	40%	

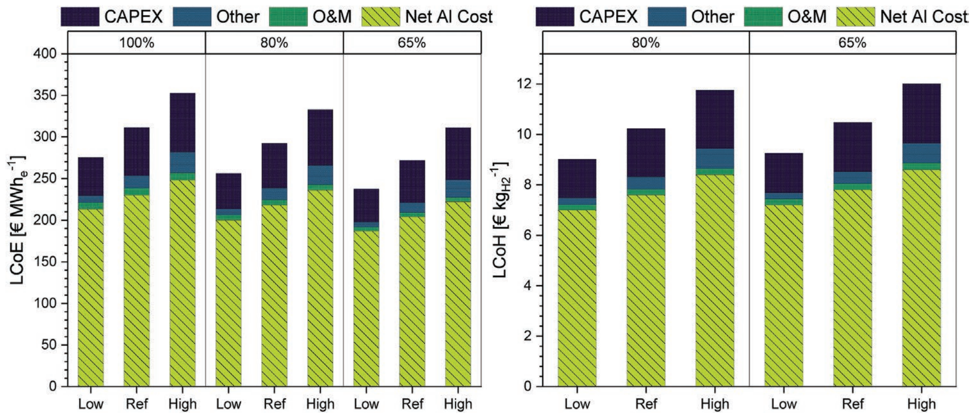

Figure 5. LCoE and LCoH estimations for the base case, i.e., 4000 FLHs and electricity price of 50 € MWh_e^{-1}. (Low: lower end, Ref: reference, and High: higher end.) The H_2 kWh equivalent is estimated based on the higher heating value of 39.4 kWh kg^{-1}.

The estimated LCoE varies between 238 and 353 € MWh_e^{-1}, while across given SOFC partial loads, the estimated LCoH accounts for 9–12 € kg^{-1} of H_2 produced. However, the net green H_2 fuel production costs based on the wind and solar power is reported as 6.5–12 € kg^{-1} H_2, including the H_2 import scenarios from Middle East and Northern Africa (MENA) regions.[101] Estimated LCoH for a base case, i.e., without any sophisticated business case consideration, provides a positive impression regarding the competitivity of the Al-based H_2, especially considering its small-scale, but on site production. The cost breakdown indicates that the highest share of the energy cost is the Al cost. Therefore, in search of a fair business case between Al producers and plant operators, the sensitivity of the Al price is investigated (**Figure 6**).

The variation of Al price between lower and higher ends results in ±23% change in the LCoE (Figure 6a) and LCoH (Figure 6b) if an energy intensity of 11 kWh $kg_{Al}{}^{-1}$ and 30 € MWh^{-1} electricity price is considered. Also, the variation of the Al_2O_3 economic value has an impact of ±16% change on the LCoE and LCoH. To elaborate on the impact of the Al price, three different energy price scenarios are considered as previously introduced: *High energy intensity scenario* (Scenario-I), *high efficiency and low electricity price scenario* (Scenario-II), and *zero electricity price scenario* (Scenario-III). The specific parameters for the above introduced scenarios are summarized in **Table 4**. Scenario-I and -II are addressing the present and near future carbon-free Al prices based on the given energy intensity and electricity prices. Scenario-III, namely '*Zero electricity price scenario*' aims to identify the techno-economic potential toward 2050 for the different availability of excess renewable energy production.

Therefore, it is considered for investigating the theoretical minimum or maximum limit LCoE and LCoH values. As shown in **Figure 7**, the 23% reduction of the energy consumption for Al production and the 40% reduction of the electricity price yields around 30% reduction on the LCoE. In particular, the LCoE for the *high energy intensity scenario* decreases to 334 € MWh_e^{-1} and for the *high efficiency scenario* to 235 € MWh_e^{-1} assuming 4000 FLHs. As for the *zero-cost electricity scenario*, the LCoE estimate further diminishes to 162 € MWh_e^{-1}. The same applies for the LCoH, with the reduced electricity price the reference estimates are 11.8, 8.4 (–29%), and 5.4 (–54%) € $kg^{-1}{}_{H2}$, respectively, for Scenario-I, -II, and -III. Besides the competitive energy storage costs, LCoH estimations imply promising economics for all scenarios, noting that H_2 is provided only as an auxiliary service. In other words, this indicates a high economic viability potential of Al-based H_2 produced because of wet combustion process, if other process equipment necessary for the power conversion are excluded.

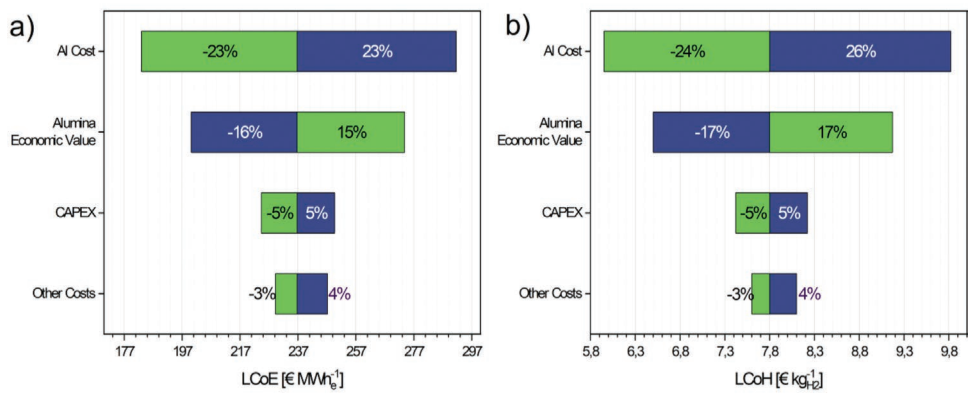

Figure 6. a) LCoE and b) LCoH cost sensitivities based on Scenario-II. The green and blue sections of the floating bars refer to decrease and increase on the selected cost components, respectively while the percentages refer to the change in LCoE or LCoH.

Table 4. Al price and energy intensity scenarios and scenario-specific parameters.

Scenario	Al price [€ kg_{Al}^{-1}]	Al_2O_3 price [€ kg_{Al}^{-1} eq.]	Energy intensity [kWh kg_{Al}^{-1}]	Electricity price [€ MWh_e^{-1}]
Scenario-I (High energy intensity)	1.44–1.86 ($\mu = 1.65$)	0.52–0.83 ($\mu = 0.67$)	14.25	50
Scenario-II (High efficiency and low electricity price)	1.06–1.48 ($\mu = 1.26$)	0.52–0.83 ($\mu = 0.67$)	11	30
Scenario-III (Zero electricity price)	0.73–1.15 ($\mu = 0.93$)	0.52–0.83 ($\mu = 0.67$)	11	0

Compared to other PtX technologies (such as power-to-methane, -methanol, -kerosene, etc.) H_2 produced via the recovered Al does not only offer better volumetric energy density, but other advantages such as no GHG emissions (with the implementation of inert anodes and 100% carbon-free electricity supply), on-site production and consumption, recyclability, and energy form versatility among others.

6. Comparison with Other Flexibilization Alternatives

The preliminary results indicate that the on-site hybrid generation of H_2 and electricity holds potential for developing business cases to use Al as a metal energy carrier. Here, the fuel production cost of the Al is estimated considering different energy intensities and introduced prospective energy intensity improvements. Thereafter, the same operational conditions (equal FLHs) are assumed to enable a comparison with the other Power-to-X technologies. It can be seen in **Figure 8**a that Al-based H_2 displays high fuel cost competitivity against P-to-methane, P-to-methanol, and P-to-kerosene based on the obtained fuel production costs even with the current electricity prices *(Scenario-I: High energy intensity scenario)*. Scenario-II and -III are anticipating the trends toward 2050, assuming 30 € MWh_e^{-1} and 0 € MWh_e^{-1} electricity prices during regular and excess renewable energy availability, respectively. Furthermore, toward Scenario-III, carbon-free Al-based H_2 production costs

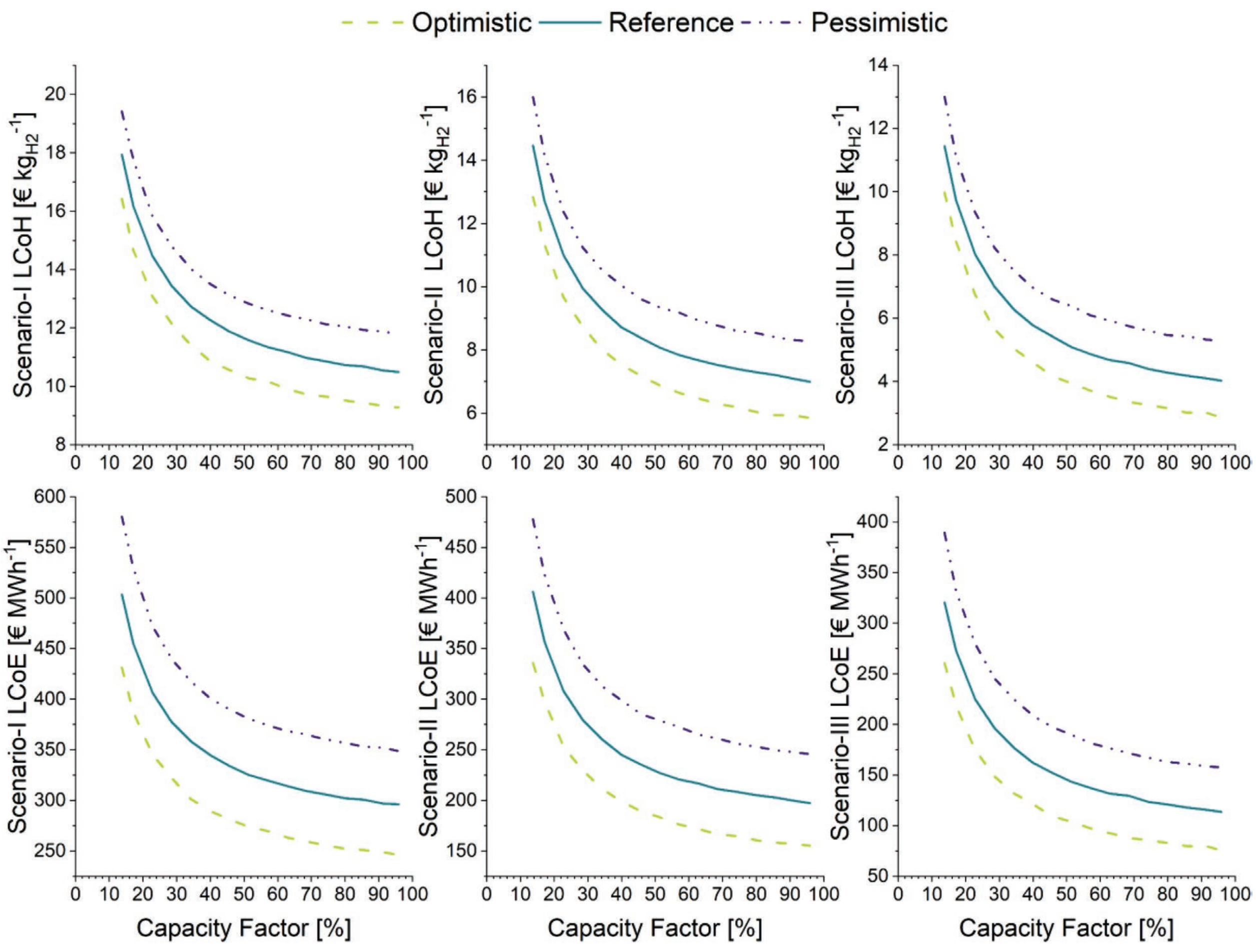

Figure 7. Variation of LCoE (PtP conversion) and LCoH (net fuel production cost) for the selected scenarios across all annual equivalent full load hour operations.

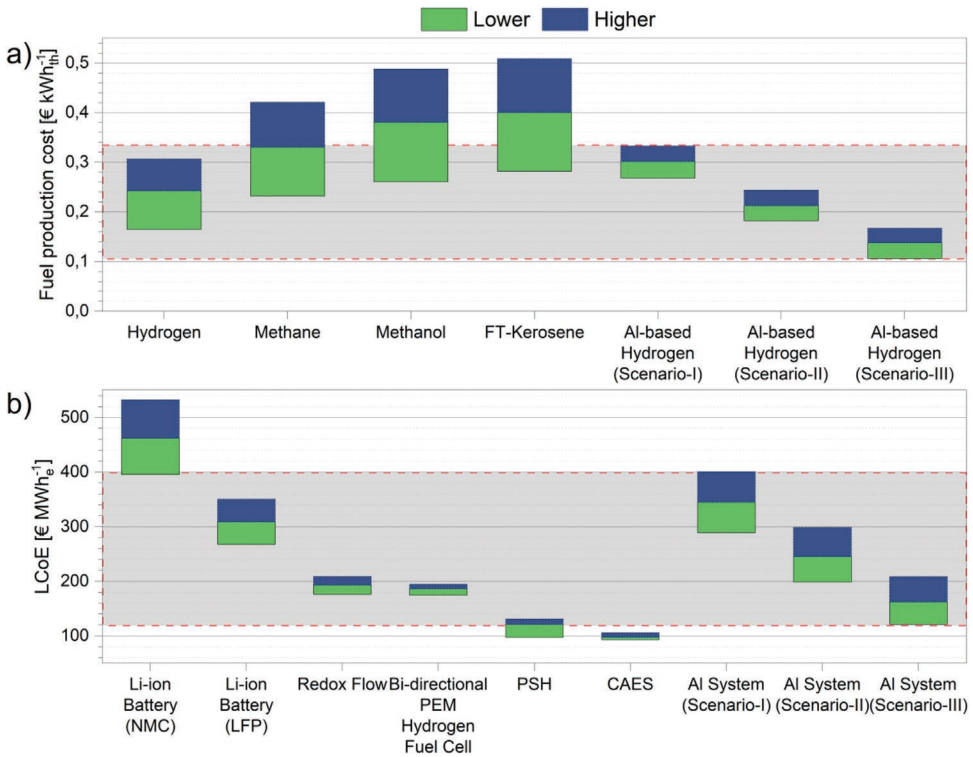

Figure 8. a) Comparison of net fuel production costs based on 4000 FLHs,[101,104] and b) LCoE of various storage technologies based on power-to-power conversion path. (The power capacity of the selected technologies for comparison is 100 MW_e and estimated LCoE values are based on the assumed 10 h of daily storage duration.)[102] (FT Kerosene: Fischer–Tropsch-synthesis kerosene, NMC: nickel–manganese–cobalt, LFP: lithium–iron–phosphate, PSH: pumped storage hydropower, CAES; compressed air energy storage.)

become more competitive depending on the potential improvements in the Hall–Héroult process and electricity prices in relation to the green H_2. Considering these anticipated electricity prices and its impact on the Al price, a reality where Al-based H_2 is more competitive than wind and solar power based green H_2 may come to fruition in the future.

As for electricity generation, the estimated LCoE values (Figure 8b) display a favorable energy storage path that is motivating for the further consideration of Al for the energy storage applications. The LCoE range of storage technologies varies on a spectrum where compressed air energy storage (CAES) represents (based on the given assumption in Mongird et al. 2020) the lower limit and Li-ion batteries the upper. It has to be mentioned, that the LCoE of CAES, are highly dependent on natural gas and CO_2 certificate prices that can vary extremely.[106,107]Although, battery applications mainly target the short-term energy storage demand, secondary electricity provided by the Al system becomes more advantageous in terms of economics. Also counting on the H_2 supply and other ancillary grid services (i.e., scheduling and dispatch, reactive power and voltage control, loss compensation, load following, system protection, and energy imbalance), it definitely asserts a suitable application for supporting the charging/refueling infrastructure with a reasonable cost. In this respect, Al-based electricity and H_2 storage system appears to hold a high potential to respond the energy supply concerns of the electric mobility.

7. Conclusion and Discussion

The conducted study provides qualitative and quantitative information on the techno-economics of Al as an energy storage carrier and medium demonstrating its validity for supporting both, the successful transition of carbon-neutral mobility and energy system. The proposed concept aims to respond several challenges (i.e., seasonal and short-term energy storage, generation-demand balance, H_2 supply, and inadequacy of refueling/recharging infrastructure) at the same time with a high techno-economic feasibility potential. In particular, ascertained power-to-X costs and high round-trip efficiency boosts the motivation for the use of Al which eliminates the costly storage demand thanks to its high volumetric energy density. Considering that a 7.5 m layer of Al stored under a football field provides ≈1 TWh of seasonal/annual electric energy storage, the volumetric energy density advantage of Al is of great importance. (see Supplementary Calculations) Additionally, one cubic meter of Al yields ≈23 MWh, which corresponds to the annual electricity demand of average six European dwellings (EU average electricity consumption: 3.7 MWh_e per dwelling).[108] The LCoE and LCoH estimations give indications regarding the economic benefits, and potential for avoiding the burdens for costly H_2 storage and transmission need. Most importantly, the comparison reveals the fact that Al has a vast techno-economic potential to discover. The LCoE is assessed very close to the one of the most cost-competitive alternative bi-directional PEM H_2 conversion system (with cavern storage), which refers to the cheapest possible concept with a complex energy conversion chain. This is a novel concept which aims to resolve one of the biggest challenges of European energy systems which include environmental sustainability, economic feasibility, safety, and self-reliance on the energy supply. In addition to the feasibility of Al-powered fueling stations for BEVs and FCEVs, this approach may be applicable in other business cases such as grid services, commercial centers, apartments, industrial complexes and households. However, a carbon-neutral Al production and availability of experimental prototype demonstrations are necessary for the realization of such a breakthrough innovation. It is important to point out that the concept can be carbon-free only with the decarbonization of one process in the chain namely the Hall–Héroult process. Even all these concerns are already addressed and clarified with planned implementation of the technology development steps, it is not possible to make clear statements about the sustainability of overall Al energy conversion chain and the proposed hybrid system without a life-cycle assessment. An equivalent assessment to the one here presented should be carried out to explore the environmental feasibility. On the whole, based on the investigated techno-economics and operational benefits Al is identified as a very suitable metal energy carrier with a high potential to respond all multi-domain aspects of the energy transition.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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aluminum, electric vehicles, energy storage, hydrogen, metal, power-to-X, renewable energy carriers

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