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"Role of hydrogen tanks in the Life Cycle Assessment of fuel cell-based Auxiliary Power Units" Applied Energy 215 (2018) pp. 1-12;
DOI:10.1016/j.apenergy.2018.01.095

Role of hydrogen tanks in the Life Cycle Assessment of fuel cell-based Auxiliary Power Units

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Abstract

In the framework of the European project SSH2S, a solid-state hydrogen storage tank - fuel cell system was demonstrated as Auxiliary Power Unit (APU) for a light duty vehicle. In this work, we have assessed the environmental impacts and the costs of the system developed. Following an eco-design approach, we have identified the processes mostly contributing to them and we have suggested possible improvements. By performing a Life Cycle Assessment (LCA), we found that, when the electricity consumption for hydrogen gas compression is included into the analysis, a solid-state hydrogen storage tank has similar greenhouse gas emissions and primary energy demand than those of type III and IV tanks. However, the resources depletion is higher for the solid-state system, even though the inclusions of the end of life of the APU and the recycling of the materials may result in different conclusions. The costs of an APU equipped with a solid-state hydrogen storage tank are significantly higher, about 1.5 – 2 times the systems based on type III and IV tanks. However, mature technologies are compared with a prototype, which has much room for optimization. To improve both the environmental and economic performances of the APU, a reduction of structural materials for both the solid-state hydrogen tank and Balance of Plant is recommended.

Keywords: Auxiliary Power Unit, Fuel Cell, Solid-State Hydrogen Storage, Life Cycle Assessment

1. Introduction

The European Union has set ambitious climate and energy targets for 2020 in its climate and energy package [1]. These targets, known as the "20-20-20" targets, set three key objectives for 2020: i) a 20% reduction in EU greenhouse gas emissions from 1990 levels; ii) raising the share of EU energy consumption produced from renewable resources to 20%; iii) a 20% improvement in the EU's energy efficiency. These targets are considered a first step towards building a low-carbon economy.

In this climate and energy framework, heavy duty transport plays a critical role, as it is one of the most difficult to be decoupled from the use of fossil fuels. Moreover, the main Internal Combustion Engine (ICE) of the truck is used not only for traction, but also for electricity production to power the truck when it is not travelling. Due to the low efficiency of this power generation method, and to the increased number of regulations limiting the idling of the main engine, the separation the two functions, i.e. traction and power supply, is becoming attractive in order to reduce emissions of noise and pollutants [2].

Auxiliary Power Units (APUs) are considered one of the promising applications of fuel cells [3]. The main fuel cell types used for this purpose are: i) low temperature proton exchange fuel cell (LT-PEM); ii) high temperature proton exchange membrane fuel cell (HT-PEM); iii) solid oxide fuel cell (SOFC).

SOFCs have a high operating temperature range (650 - 950°C [2]), and can operate with hydrocarbon fuels, such as diesel, needing only relatively low reforming and cleaning efforts. This possibility to use the same fuel as the main ICE engine of the truck makes the SOFCs particularly suitable and, currently, the most viable alternative for applications in APUs [4,5]. The main drawback of SOFCs is the long time needed for start-up, which represents one of the challenges for system improvements [5,6].

PEM fuel cells have been found to reduce significantly the environmental impacts of vehicles, especially in terms of GHG emissions when compared to ICE vehicles [7, Bohnes F.A., Gregg J.S., Laurent A., Environmental Impacts of Future Urban Deployment of Electric Vehicles: Assessment Framework and Case Study of Copenhagen for 2016−2030, Environ. Sci. Technol. 2017, 51: 13995−14005], or APUs [8], while it was shown that climate benefits can be substantial with the use of hydrogen originating from renewable energy sources [Cox B.L., Mutel C.L., The environmental and cost performance of current and future motorcycles, Applied Energy 2018, 212: 1013–1024] [Karaaslan E., Zhao Y., Tatari O., Comparative life cycle assessment of sport utility vehicles with different fuel options, Int J Life Cycle Assess 2018, 23: 333–347]. Finally it was observed that the results can strongly depend on the type of electricity (i.e., with fossil, renewable, or nuclear origin), according to the country specific energy mix, that is used to feed the electric and hybrid vehicles [Lombardi L., Tribioli L., Cozzolino R., Bella G., Comparative environmental assessment of conventional, electric, hybrid, and fuel cell powertrains based on LCA Int J Life Cycle Assess 201,7

22:1989–2006]There is a growing interest in PEM fuel cells application as APU, thanks to their lower operating temperatures close to room conditions, and thus shorter start-up times [4]. HT-PEM fuel cells have, obviously, a higher operating temperature (160-180°C) than LT-PEMs. This has an important consequence on the hydrogen purity requirements: HT-PEMs have a higher tolerance to CO and other impurities, compared to LT-PEMs. Although some studies have been reported on optimization of fuel processing solutions to run LT-PEMs on diesels and jet fuels [9-11], HT-PEMs promise better performances, requiring a less complex system for reforming, together with a better tolerance to impurities [12].

All the above studies imply the use of a reforming process in order to obtain hydrogen from diesel (or other hydrocarbon-based fuels), which represents also the fuel of the main engine. But, in perspective, the use of innovative vehicles not running on fossil fuels, the storage of pure hydrogen on-board would be the simplest solution to avoid the combustion processes.

Storage is still one of the bottlenecks towards a sustainable carbon-free economy based on hydrogen. Among various options, hydrogen storage in the solid state has attracted much attention in recent years [13]. Many different hydride types and compositions have been investigated, as reported by studies concerning both metal and complex hydrides [14-16]. The first studies for industrial use of metal hydrides hydrogen storage were made by Daimler Benz in the eighties by combining metal hydride tanks with ICEs in mini vans [17]. By coupling a hydride tank with a PEM fuel cell, the heat recovered from the latter can be used to desorb the H_2 in the hydrides. This has been proved in the studies by Ungethüm et al. [18] and Urbanczyk et al. [19]. In both cases, an HT-PEM was coupled with a complex hydride tank. In the study by Ungethüm et al. [18] a tank filled with $CsCl_3$ - doped sodium alanate- was coupled with a 1.1 kW HT-PEM fuel cell. The authors focused in particular on heating requirements of the system during start-up: in the first 20 minutes the fuel cell was operated with H_2 from a hydrogen buffer tank while the FC-hydride tank system was heated up to the operating temperature. In the study by Urbanczyk et al. [19] a Ti-doped sodium alanate hydride tank has been coupled to a 260 W HT-PEM fuel cell. This system was run for several cycles of 3 h each, generating an electric energy of 0.66 kWh. In a study by Rizzi et al. [20], a hydride tank was filled with an intermetallic compound, $LaNi_{4.8}Al_{0.2}$, and coupled with a 1.2 kW PEM fuel cell. This system was operated for more than 6 h at an average power output of 0.76 kW. For 2 h it provided 1 kW output power, and during the running time hydrogen flow increased for the first 100 min (due to start-up) remaining constant for more than 2 h. This system is of interest for its size and because it runs at different load requirements, similarly to the aforementioned APUs.

In the framework of the European project SSH2S, it was developed a solid storage (SS) tank - fuel cell system, and its functioning was demonstrated on a real application, i.e. an APU for a light duty vehicle. As described in detail in [21], unlike the previous studies that coupled PEMs with hydride tanks containing only one type of hydride (i.e. metal hydrides [20] or complex hydrides [19]), the active material in this case is obtained coupling a mixture of complex hydrides (CxH: $2LiNH_2$ + $1.1MgH_2$ + $0.1LiBH_4$ + 3wt.% $ZrCoH_2$) with a hydride of an intermetallic compound (MeH: $LaNi_{4.3}Al_{0.4}Mn_{0.3}$). The tank is composed by several tubes, each consisting of two separate concentric cylindrical compartments containing, respectively, MeH (inner) and CxH (outer), that are separated by a filter mesh made of copper.

This work aims at assessing the environmental impacts of the APU developed in the SSH2S project with a Life Cycle Assessment (LCA) approach. Several LCA studies have been performed on PEM fuel cells [22-25], whereas an LCA on a hydride tank, to our knowledge, has not been published yet, nor a detailed inventory is available. The SSH2S hydride tank is compared with similar commercial systems (type III and IV pressure vessels), which currently represent the most widely used hydrogen storage solutions.

The goal is to assess the environmental performances of the developed system and, with an eco-design approach, to identify the processes mostly contributing to the environmental impacts and to recommend possible ways of reducing them. To complete the analysis, a simplified economic analysis was performed to understand the economic feasibility and to identify the hotspots in the cost structure.

The added value and novelties of this work can be summarised as follows:

- it is the first study on the environmental impacts of a hydrogen solid state storage system
- it compares, with a consistent and comprehensive approach, the environmental performances of solid state storage systems and pressure tanks
- it includes the energy required for hydrogen compression in the comparison of the systems
- it analyses also the costs of the different technologies considered.

2. Materials and methods

This work follows a Life Cycle Assessment (LCA) methodology to assess the environmental impacts of the APU described in detail in [21], comparing the prototype hydride tank used by the APU developed in the SSH2S project with commercial type III and a type IV compressed gas tanks. LCA is a structured and internationally standardized method aimed at quantifying all relevant emissions and resources consumed and the related environmental and health impacts and resource depletion issues that are associated with the entire life cycle of any goods or services (“products”) [26]. In this study, the LCA was performed according to ISO 14040 and 14044 standards requirements [27, 28], by using the software GaBi 6.3 from PE International [29]. Moreover, the LCA was performed according to a specific guidance document developed within the Project FC-HyGuide of the FCH JU [30]. The FC-HyGuide guidance document [31] is based on, and in line with, the International Reference Life Cycle Data System (ILCD) Handbook on LCA, coordinated by the European Commission's JRC-IES, through the European Platform on LCA. The ILCD Handbook is applicable to a wide range of different decision-contexts and sectors, and therefore needs to be translated to product-specific criteria, guidelines and simplified tools to foster LCA applications in the specific industry sectors. The FC-HyGuide project responded to this need by providing a guidance document on how to perform every step of a LCA for hydrogen (H_2) production systems and fuel cell technologies.

2.1 Goal and scope definition

The goals of the performed LCA were the following:

1. Quantify the environmental burden associated to the production of 1 unit of the SSH2S APU and the identification of the processes and material inputs that mostly contribute to it.
2. Compare the system under analysis with systems providing the same services, i.e. type III and IV compressed gas tanks associated with the same APU. This comparison was extended to the use stage of the device.
3. Provide an estimation of the environmental impacts associated with a 5 kW up-scaled SSH2S APU.
4. Analyse the impacts of extending the use of the devices to real needs (from 2 to 8 hours).

Thus, nine APUs have been modelled. To simplify readability of the text while highlighting the differences among the systems, these will be named using the (x)kW_(y)h_(ZZ) notation, where (x) represents the power output of the fuel cell, which can be 1 or 5 kW, (y) represents the operating time per cycle, i.e. 2 or 8 hours, while (ZZ) represents the type of tank, which can be a solid state hydride tank (SS), a type III (T3) or a Type IV (T4) compressed gas tank.
The differences in components among the described APUs are resumed in tab. 1.

	Stack (power)	Operating step (time)	Tank (volume or mass of hydrides)	Balance of plant
1kW_2h_SS	1 kW	2 h	6.7 kg hydrides	Original
1kW_2h_T3	1 kW	2 h	7 l	Original
1kW_2h_T4	1 kW	2 h	4 l	Original
1kW_8h_SS	1 kW	8 h	26.8 kg hydrides	Original
1kW_8h_T3	1 kW	8 h	22 l	Original
1kW_8h_T4	1 kW	8 h	11 l	Original
5kW_2h_SS	5 kW	2 h	34.2 kg hydrides	Rescaled
5kW_2h_T3	5 kW	2 h	35 l	Rescaled
5kW_2h_T4	5 kW	2 h	17.5 l	Rescaled

Tab. 1 Overview of modelled APUs.

Although for the hydride tanks only the content of hydrides is reported, it must be pointed out that also the structural part of the tank changes, as described in detail in 2.3.1.1.

To account for the different pressure level of the H_2 used by the three storage systems, the electricity required for compression during the 10 years lifespan of the different APUs was added to the analysis.

H_2 may be produced with many different sources of energy (e.g. different renewables or fossil) or exploiting excess energy from the electricity grid during periods of reduced demand by the users. The production of the H_2 was left out of the system boundaries because it cancels out in the comparison (i.e. with the same source of hydrogen and the same fuel cell efficiency it makes no difference whether the hydrogen is stored in pressure tanks or solid state), furthermore, the impacts from H_2 production would hide the differences among the systems and would likely add uncertainties.

The modelling framework is comparative attributional. The results are to be disclosed to the public and the intended audience includes scientists, industry, and policy makers. There is no multifunctionality. It was avoided the application of a specific cut-off rule; however, some inputs were considered negligible given their very limited amounts. The data quality was considered sufficient to draw the conclusions and recommendations found, given that the device is a prototype.

The relevant impact categories chosen for the impact analysis are the Global Warming Potential (GWP) and the Abiotic Depletion Potential (ADP), also called resource depletion. We have considered also, as additional technical quantity, the Primary Energy Demand (PED), renewable and non-renewable.

2.2 Life Cycle Inventory

Life Cycle Inventory (LCI) involves a systematic inventory of the input and output energy and material flows during the entire life-cycle. The data that constitute the inventory used in this work derive mostly from peer reviewed literature or from primary data from field-studies. The data used for the background processes were obtained (if not otherwise specified) from the commercial database Ecoinvent [32].

To analyse which part of the APUs contributes the most to the environmental impacts, the systems was divided in three sub systems, the Balance of Plant (BoP), the fuel cell (stack) and the hydrogen storage (tank). In addition, for gas tanks, the electricity required for H_2 compression during the 10 years lifespan of the APUs was included in the analysis.

The datasets used to model the inputs from the technosphere are taken from commercial databases (Ecoinvent and Gabi Professional). The data and information on the solid-state hydrogen storage tank using hydrides were collected by the partners of the SSH2S consortium, as result of the work performed within the project [33, 21]. The two HTPEM fuel cells considered in this study have 1 kW and 5 kW power output, differing only in the number of cells that compose the stack. The corresponding detailed inventories are shown in table SM1 of the Supplementary Material (SM). The inventories of the BoP components, for both the 1 kW and the 5 kW systems, are reported in table SM2 of SM. Some components have been neglected due to their high complexity and low contribution to the system's weight (less than 3%).

2.2.1 The solid state (SS) hydrogen storage tank

2.2.1.1 Inventory for the structural components of the SS tank

This part consists in the components necessary to confine and operate the hydrogen storage materials, as shown in fig. 1.

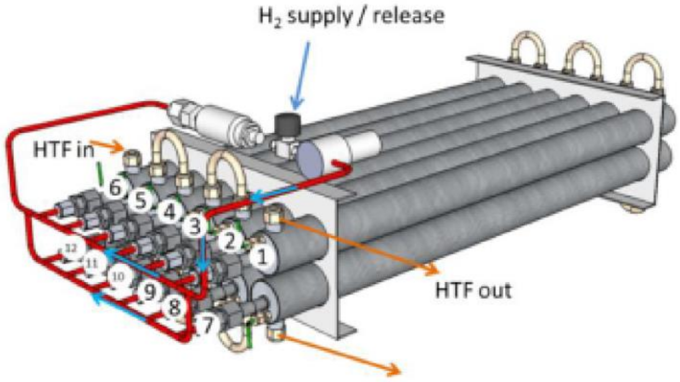

Fig. 1 Scheme of the structural part of the solid state hydrogen storage tank of the SSH2S APU .

As showed in fig. 1, for the 1kW_2h_SS, the hydrides are contained in 12 stainless steel tubes (2 rows of 6 each). The tanks considered for the 1kW_8h_SS and 5kW_2h_SS systems are similar to that shown in fig. 1, although rescaled and consisting of 30 tubes (2 rows of 15 each) and 36 tubes (2 rows of 18 each), respectively. Part of this storage systems, not shown in fig. 1, is a small aluminium tank used to store the gaseous hydrogen necessary for the system's start up, as described in [22]. This is a commercial type I pressure vessel.

The SS tank prototype has been assembled with commercial materials (i.e. stainless steel sheets, glass fiber etc.) and various components, as reported in tab. 2.

Material	**Amount (kg/unit of product)**		
	1kW_2h_SS	1kW_8h_SS	5kW_2h_SS
Stainless steel	44	105	126
Aluminium alloy 6061	3.7	3.7	17.2
Glass fiber	3	5.1	5.6
Copper	2.5	6.1	7.4
Aluminium foil	1.0	2.5	3.0
Acetone	1.0	1.0	1.0

Tab. 2 Inventories for the SS tanks.

The amount of 6061 aluminium alloy reported in tab. 2 represents the type I tanks used for the start-up of the systems.

2.2.1.2 Inventory for the hydrogen storage materials

This sub-unit consists on one side in the active hydrogen storage materials. The 1kW_2h_SS system contains 3.4 kg of a widely-used intermetallic compound, $LaNi_{4.3}Al_{0.4}Mn_{0.3}$ (MeH), and of a similar amount of complex hydrides mixture $2LiNH_2 + 1.1MgH_2 + 0.1LiBH_4$ + 3wt%$ZrCoH_3$ (CxH). The contribution of the $ZrCoH_3$ and of the $LiBH_4$ was neglected because their element contribution results in few hundredths of kg (for example Zr is only 0.06 kg) and there are no data in literature and commercial dataset with sufficient quality.

The individual elements contributions are given in tab. 3. The relative amount of elements in hydrogen storage materials is maintained also for the other hydride tanks.

$LaNi_{4.3}Al_{0.4}Mn_{0.3}$	3.4 kg
La	1.13 kg
Ni	2.05 kg
Al	0.09 kg
Mn	0.13 kg

$2LiNH_2 + 1.1MgH_2$	3.3 kg
N	1.2 kg
Li	0.6 kg
Mg	1.2 kg
H	0.3 kg

Tab. 3 Individual contributions of elements for the 1kW_2h_SS tank.

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A considerable attention has been dedicated to the process route for the hydrogen storage materials. The detailed process path considering all the production and treatment steps is sketched in fig. 2. There is little known on the individual steps of this path to be able to perform a detailed analysis on it. To our knowledge, in fact, no LCA studies have been published on this type of materials. The processing route for each material will be then detailed in the following sections.

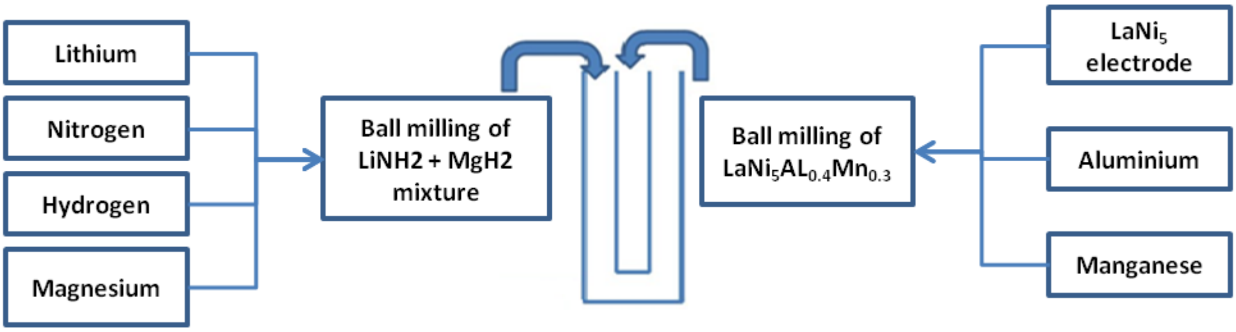

Fig. 2 Detailed process paths for the production of the hydrogen storage materials. Left: complex hydride (CxH); right: (MeH). A single tube, consisting of two separate concentric cylindrical compartments containing, respectively, MeH (inner) and CxH (outer), is shown in the middle.

The inventory for CxH is reported in tab. 4.

Material	**Amount (kg/unit of product)**		
	1kW_2h_SS	1kW_8h_SS	5kW_2h_SS
Total	**3.3**	**13.4**	**17.2**

Lithium	0.60	2.43	6.25
Magnesium	1.20	4.87	3.12
Hydrogen	0.30	1.22	6.25
Nitrogen	1.20	4.87	1.56
Ball milling	**kWh/unit of product**		
Electricity (EU-27 grid mix)	161	654	839

Tab. 4 Inventory for CxH production.

$LiNH_2$ and MgH_2 were mixed and ball milled together to form a pulverized complex hydride mixture. The data relative to the energy involved in the process are primary data, i.e. 2.1 kW maximum ball miller power, with 70-75% normally consumed, i.e. about 1.5 kW, for a total milling time of 106 hours. The inventory for MeH is reported in tab. 5.

Material	**Amount (kg/unit of product)**		
	1kW_2h_SS	1kW_8h_SS	5kW_2h_SS
Total	**3.4**	**13.5**	**17.2**
$LaNi_5$ (electrode material)	3.20	12.6	16.1
Manganese	0.13	0.55	0.66
Aluminum (primary)	0.09	0.35	0.45
Ball milling	**kWh/unit of product**		
Electricity (EU-27 grid mix)	161	654	839

Tab. 5 Inventory for MeH production.

For $LaNi_5$, the production of electrode material was considered, as it is reasonable to suppose that similar levels of purity of the material will be required for both electrode and hydrogen storage applications. As energy input for the ball milling process of MeH does not differ significantly from that of CxH, the same values have been considered.

2.2.2 Inventory for type III tanks

The commercial type III tanks are considered to operate at a pressure of 350 bar. The size of the tanks, constituting materials and relative amounts are reported in tab. 6.

		1kW_2h_T3	1kW_8h_T3	5kW_2h_T3
Size of the tank (l)		**7**	**22**	**35**
Component	**Material**	**Amount (kg/unit of product)**		
Liner	Aluminium	2.9	9.8	15
Overwrap	Carbon fiber	2.4	8.2	12.6
	Epoxy resin	1.6	5.5	8.4
Bosses	Stainless steel 316	0.5	0.5	0.5

Tab. 6 Inventory for type III tank.

2.2.3 Inventory for type IV tanks

The commercial type IV tanks operate at 700 bar. The size of the tanks, constituting materials and relative amounts are reported in tab. 7.

	1kW_2h_T4	1kW_8h_T4	5kW_2h_T4

Size of the tank (l)		**4**	**11**	**17.5**
Component	**Material**	**Amount (kg/unit of product)**		
Liner	High Density Polyethylene (HDPE)	1.6	5.6	7.4
Overwrap	Carbon fiber	1.2	4.2	5.5
	Epoxy resin	0.6	2.0	2.6
Outer layer	Glass fiber	0.1	0.4	0.6
Bosses	Stainless steel 316	0.5	0.5	0.5

Tab. 7 Inventory for type IV tanks.

2.2.4 Use phase

The expected lifespan of commercial type III and IV tanks, as well as the type I hydrogen buffer, is assumed to be 10 years. During this period, the tanks should be periodically inspected at least once every five years. This inspection, however, normally does not imply substitutions of components. The estimation of the lifespan of the hydride tank is not straightforward, because it is a prototype. Two contributions to the life span can be identified: the hydrides and the structural part of the tank. As an example of cycling stability of hydrides, studies are available in literature for $LaNi_5$ [34] and MgH_2 [35]. According to the results of these studies, in 10 years (3000 cycles) 80% of the initial capacity of $LaNi_5$ is preserved, while MgH_2 after 1000 cycles preserves 70% of the initial capacity, but its behaviour after a higher number of cycles is not reported in the study.

Reversibility of hydrogen sorption reactions has been demonstrated for the selected system up to 10 cycles [36]. Further studies extended the analysis up to 30 cycles, showing partial reduction of the hydrogen gravimetric density [37]. In 10 years, thus, a slight performance reduction of the SS tank is expected, but it was neglected given the uncertainty associated to this novel technology. No fatigue strength tests have been carried out on the structural parts of the tank, it is thus difficult to give an estimation of its lifespan. It is however reasonable to think that the lifespan of the whole tank could be 10 years.

Similarly, the fuel cell requires few maintenance operations during its life [38] and, for this reason, it was not considered a limiting factor. As regards the BoP, few plastic and metal components might be changed once or twice in 10 years of operation (e.g. check valves, H_2 sensor). However, the impact of these components on the use phase, seen their low weight, is considered negligible and thus it has not been considered.

A considerable work on impact assessment of various hydrogen production methods has been performed in previous studies [39-41]. In this study, when comparing APUs with the same features and differing only by the type of tank, given a hydrogen source, the amount of hydrogen required in this case is the same for all similar APUs, thus this impact is cancelled out in the comparison. On the other hands, adding to LCA the same amount of GHG emissions and resources depletion for hydrogen production to the three systems would just dilute the differences among them.

When changing type of tank within the same type of APU, on the other hand, the operating pressure of the tank changes, it is thus interesting to evaluate also the impact of the energy involved in the compression of the gas up to the operating pressures of the tank. The life scenario is supposed to be consisting of one charge/discharge cycle per day for 300 days per year, with a total life of the device of 10 years. The energy required for hydrogen compression to 70, 200, 350 and 700 bar has been estimated to be 2.6, 3.3, 3.7 and 4.1 kWh/kg H_2, respectively [43] corresponding to about 8, 10, 11 and 12 % of the energy content of the hydrogen (LHV). These values are in line with those found in literature for similar compression processes [44]. The electricity required for compression during the 10 years lifespan of the different APUs is reported in tab. 8. The electricity mix chosen is the EU-27.

		1 kW_2h	1kW_8h	5kW_2h
Tank		**Energy input (kWh)**		
SS - Prototype tank	Hydride part (70 bar)	1350	5370	6750
	Buffer (200 bar)	570	570	2670
T3 - Type III tank (350 bar)		1950	7770	9480
T4 - Type IV tank (700 bar)		2130	8490	10380

Tab. 8 Electrical energy inputs necessary for hydrogen compression to be filled in various tanks.

It is necessary to point out that the energy input for the compression of the hydrogen to 200 bar in table 8, is equal for the 1 kW systems considered, since in both 2 and 8 hour cycle cases, the start-up of the APU is supposed to occur only once.

In conclusions, the use phase includes only the energy necessary for the compression of hydrogen up to the pressures required by various tanks. Neither the contribution of the hydrogen production itself, nor that of the maintenance of the components of the APU were considered. The latter, in fact, requires little maintenance.

2.3 Life cycle impact assessment

The materials and energy flows identified in the life cycle inventory phase have been categorized and assigned to the relevant impact categories using the commercial software GaBi [29]. The impact assessment methods used are those recommended by the ILCD [45]. The numerical results for all the impact categories analysed are reported in table SM3 in SM with a contribution analysis on the subsystems, while in table SM4 in SM the analysis is based on the materials most contributing to the impacts generated by the tanks production.

2.3.1 Climate change

Fig. 3 shows the results for the climate change impacts of producing the fuel cells and tanks in term of greenhouse gas (GHG) emissions. Data are shown comparing systems providing the same power and operating for the same time, but considering different tanks. For the impact category climate change the metric used in this work is GWP100, calculated according to IPCC AR5 [46], and the corresponding unit used is kg of CO2eq. .

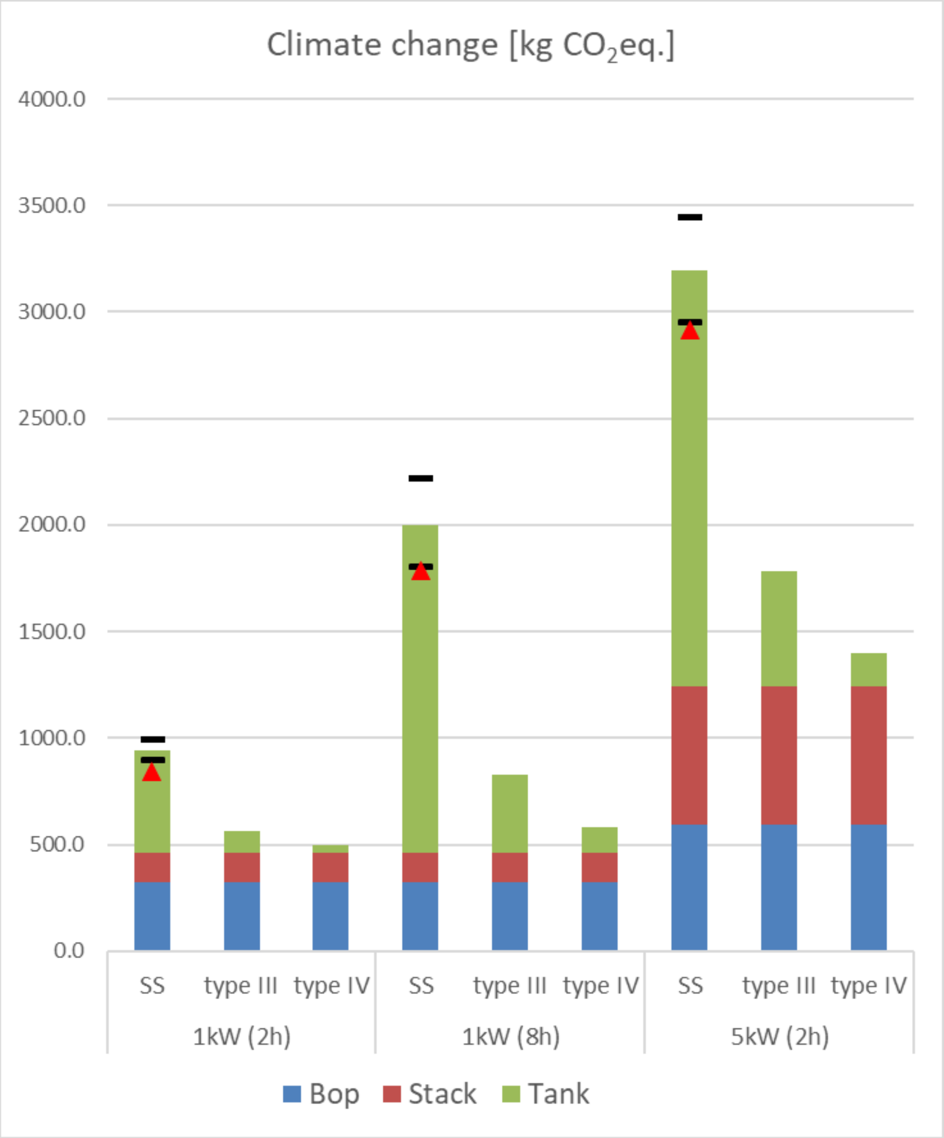

Fig. 3 GHG emissions of the manufacturing phase of one unit of systems analysed, with the sub-systems relative contribution. The black dashes show the total emission occurring when the specific GHG emissions per kg of MH and CxH are increased or decreased by 20%. The red triangle represents the total emissions occurring by reducing the total amount of steel used by 50%.

Fig.3 clearly shows that the construction of the solid-state storage tank has much higher emissions than that of the pressurized tanks type III and IV, while the contribution of the BoP and stack are independent from the used tank.

The BoP and Stack subsystems provide the largest contribution to the GHG emissions in the case of T3 and T4, if only the construction phase is considered, while the SS tank has emissions similar to the sum of Bop and stack only in the case of the 1 kW system operated for 2 hours. The GHG emissions of the SS tank are much higher than the BoP and stack for the bigger system (5 kW) or the system operated for more hours (8 hours).

Given the relevance of the hydrides and steel production on the total impacts, we have carried out a sensitivity analysis to understand how these results are affected by a change in the input data.

In the case of the hydrides, considering the intrinsic uncertainty of the Ecoinvent data and the fact that $LiBH_4$ was neglected, we have calculated what would be the impact of increasing or decreasing the environmental impact of MH and CxH production by 20%. As shown in Fig. 3, the total GHG emissions would change significantly (about + or - 5, 10 and 8 % respectively for the 1kW(2h), 1kW(8h) and 5kW(2h). However, the emissions from the manufacturing of the SS system would still remain much higher that the pressure vessels.

The SS system analysed is a prototype, with some room for improvement with regard to the consumption of materials. In particular, the vessels used, within the project, for CxH and MH containment, can withstand a pressure of 200 bars, in order to have an adequate tool for performing further experiments. Given the lower pressure used and the potential optimisation in commercial applications, we have analysed what would be the impact of halving the amount of steel used. The results are shown as red triangles in Fig.3. Again, the GHG emissions are reduced substantially (about 11.6, 12.0 and 9.8 % respectively for the 1kW(2h), 1kW(8h) and 5kW(2h)), but the emissions of the SS system remain much higher than the pressure vessel systems.

As shown in Fig.4, when the GHG emissions due to the H_2 compression during the use phase are added, the three systems of storing hydrogen have basically the same emissions per kWh of electric power produced. In fact, the higher emissions due the construction of the SS tank are balanced out by the additional demand of energy for the hydrogen compression in T3 and T4 systems.

The results would be different if the electricity used for the hydrogen compression would come from an electricity grid with a different energy mix, such as with a high penetration of renewables. To understand what would be the impact of lower GHG emissions from the electricity system on the total emissions per kWh, we have performed a sensitivity analysis by reducing by 50% the emissions of the electricity mix. As the red dashes in Fig. 4 show, the GHG emissions from the SS systems would result higher than the type III and type IV systems (about 16, 20 and 24 % respectively for the 1kW(2h), 1kW(8h) and 5kW(2h)).

We note also a clear economy of scale for both the longer daily operation (i.e 8 hours) and the larger FC system (i.e. 5 kW), with the system operated for 8 hours a day with the lowest GHG emissions per kWh.

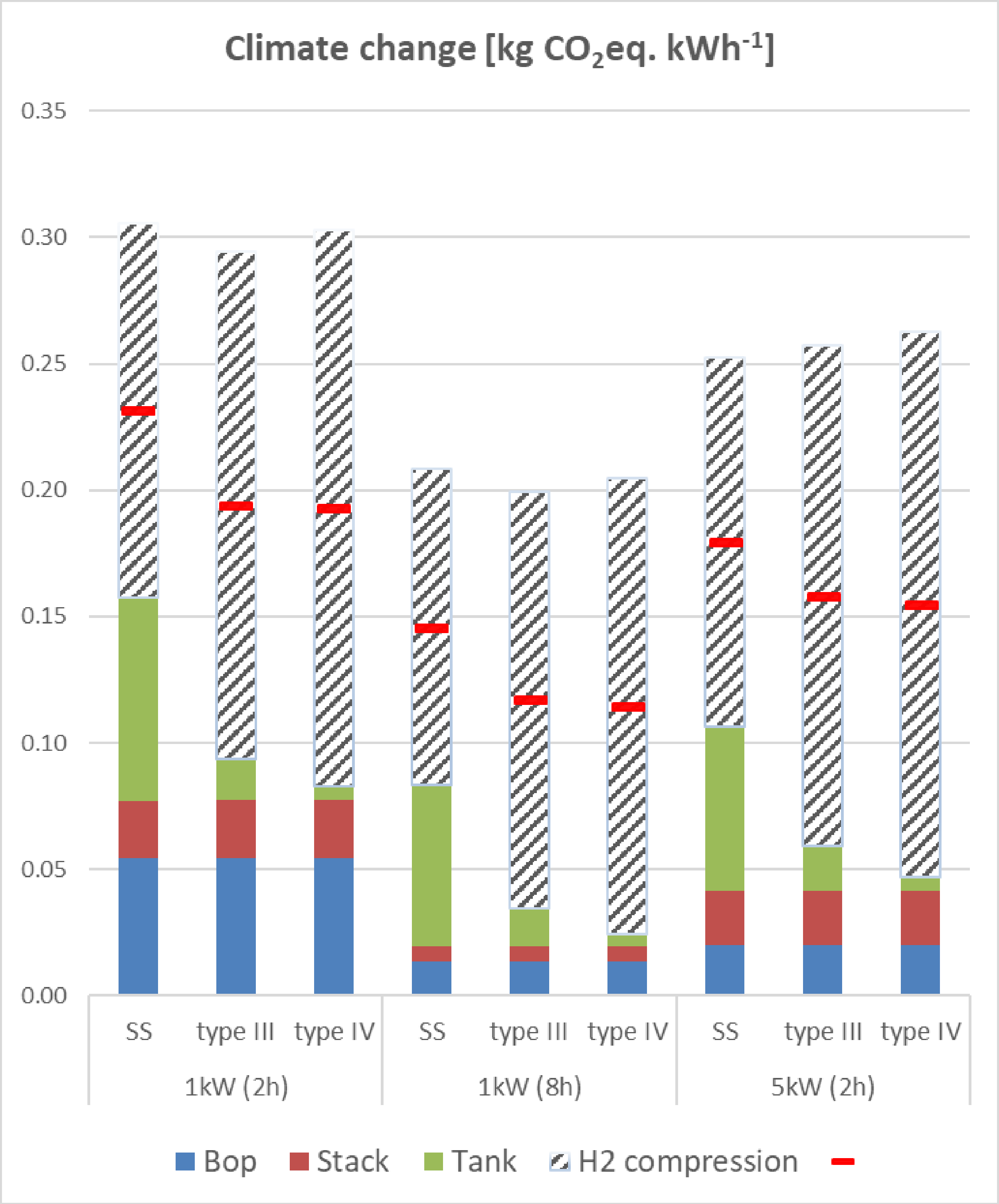

Fig. 4 GHG emissions per kWh of power produced, with the sub-systems relative contribution and including hydrogen compression with the current Italian Electricity Mix. The red dash represents the total GHG emissions of the different systems when the electricity used for hydrogen compression comes from a future energy system where the emissions are halved.

The impacts of the tanks have been analysed more in detail. The contribution to the GHG emissions among the constituting materials of the tanks is shown in fig. 5.

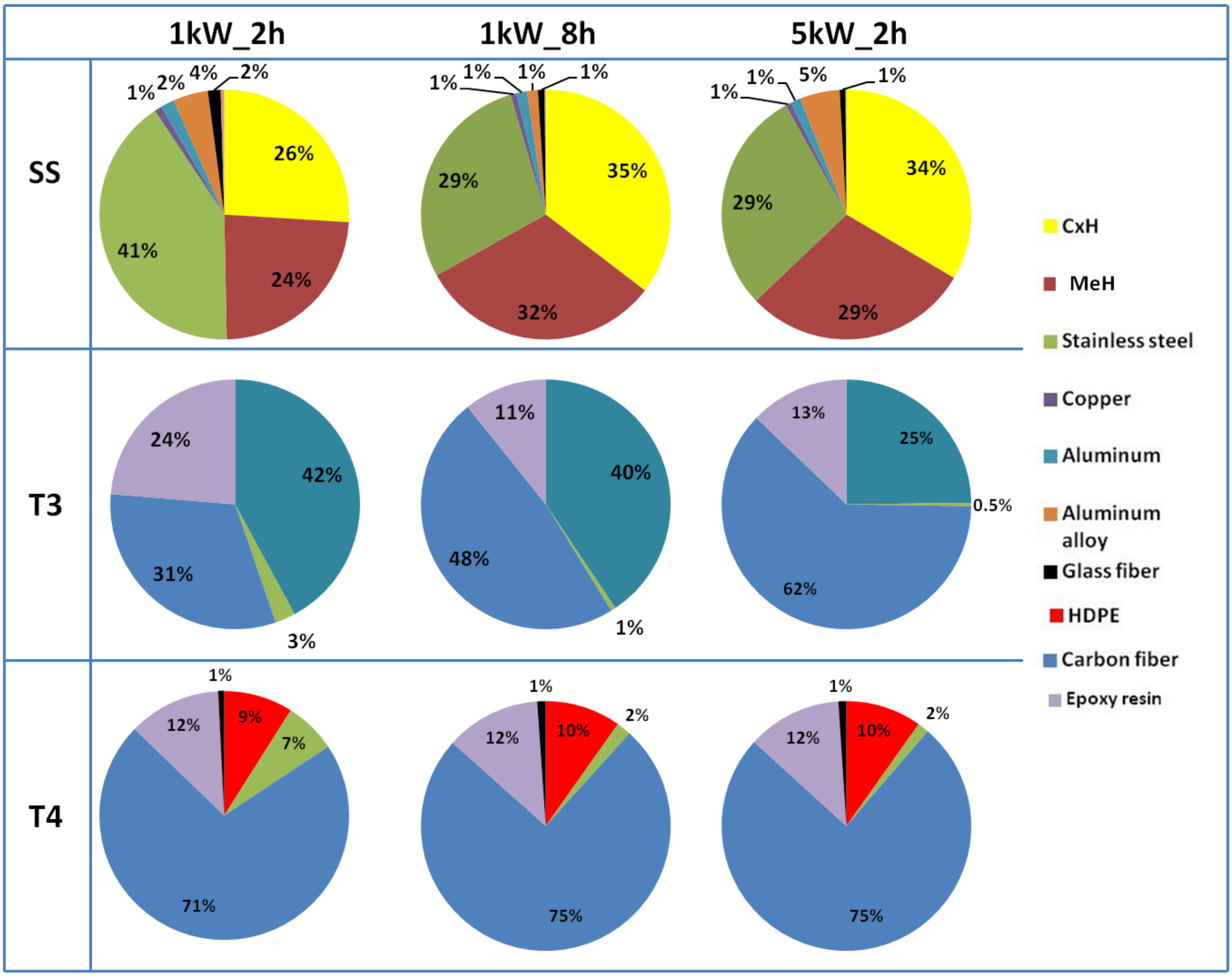

Fig. 5 GWP distribution among the constituting materials of the tanks.

For the SS tank, it is evident that the impact is mostly shared between stainless steel and the hydrides, while the contribution of the other structural materials is less than 10% in all cases. The small differences in impact distribution among the different SS tanks are related to the slight differences in their design, as explained in paragraph 2.2.1.1.The impact of CxH on GWP comes mainly from magnesium production, based on the electrolysis of molten $MgCl_2$, which is a very energy-intensive process [47]. Another significant impact is given by ball milling step in the production process. It is, however, to be pointed out that the ball milling process has been carried out with a lab scale equipment, thus it is reasonable to consider that, with the use of industrial equipment, this impact may be significantly reduced. In the case of MeH, on the other hand, almost the totality of the GWP associated with the hydride production is related to the production of $LaNi_5$.

For both type III and IV tanks the carbon fibre manufacturing plays a dominant role in GWP. This is mainly due to the energy intensive carbonization process of the polyacrylonitrile precursor [48]. Another significant contribution is given by aluminium production for the type III tank.

As far as the BoP assembly is concerned, a 30% contribution is given by the glass fibre-reinforced polyamide used for the cabin containing the APU. This impact is mainly the result of glass fibre production, which is a very energy intensive process involving silica melting [49].

The 11% contribution from aluminium is not surprising, considering the high energy needed for raw aluminium production [50]. The same holds also for the highly-alloyed steel (21%) [51]. On the contrary, it is not intuitive that 14% of the total GWP emissions are caused by the tetra-fluoro-ethylene (Teflon) contained in the seals of tubes and other components, even though its mass is rather small. However, during the production of Teflon, according to the Ecoinvent data, small amounts of HCFC 22 (chloro-difluoro-methane) and CFC 12 (dichloro-difluoro methane) are released [52]. Being the GWP of these substances 1810 and 10900 respectively [50], their contribution to the total GWP is to be regarded as a weak point. Considering the fuel cell stack contribution, the major contribution comes from platinum, as confirmed by previous studies [22, 54,55].

2.3.2 Primary Energy Demand

The amount of primary energy demands for manufacturing the APUs (i.e. BoP, stack and tank) and hydrogen compression is summarized in fig. 6.

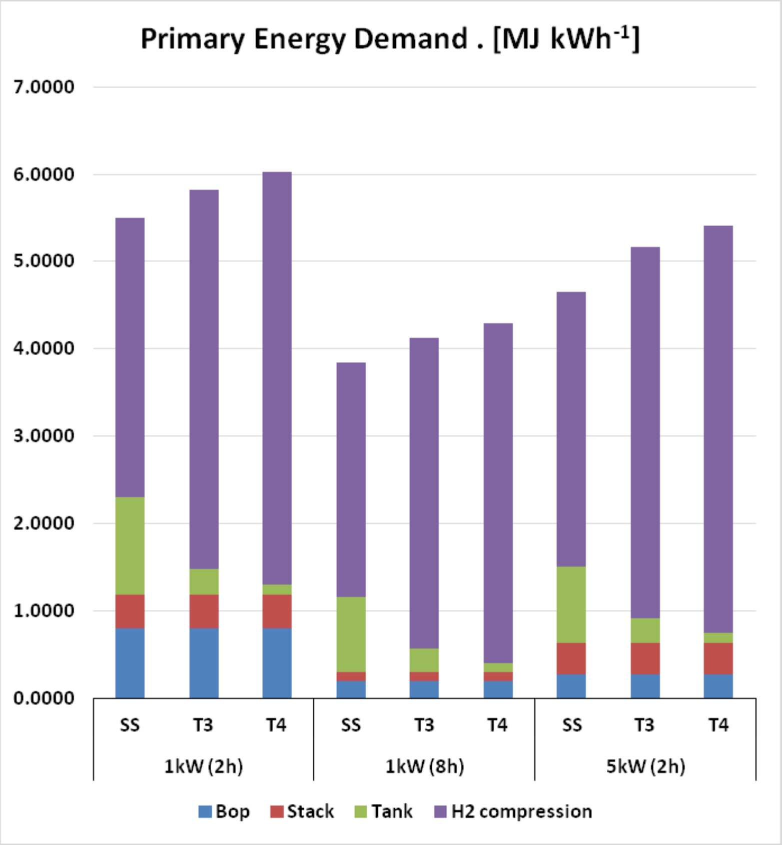

Fig. 6 Primary energy demand for the systems analysed.

The energy demand for the construction of the SS system is clearly much higher than the energy demand for the tanks T3 and T4. However, when the energy for compressing the hydrogen is included in the analysis, the SS system shows a lower total energy demand per kWh of energy produced. Similarly to the GHG emissions, the economy of scale is higher for the system operated for 8 hours a day.

2.3.3 Resource depletion

The resource depletion measures the consumption of natural resources for the production and use of the system analysed. It is based on scarcity and the characterisation factors are named Abiotic Depletion Potentials (ADP) and it is expressed in kg of antimony equivalent, which is the adopted reference element. The resources depletion of the systems analysed is reported in fig. 7.

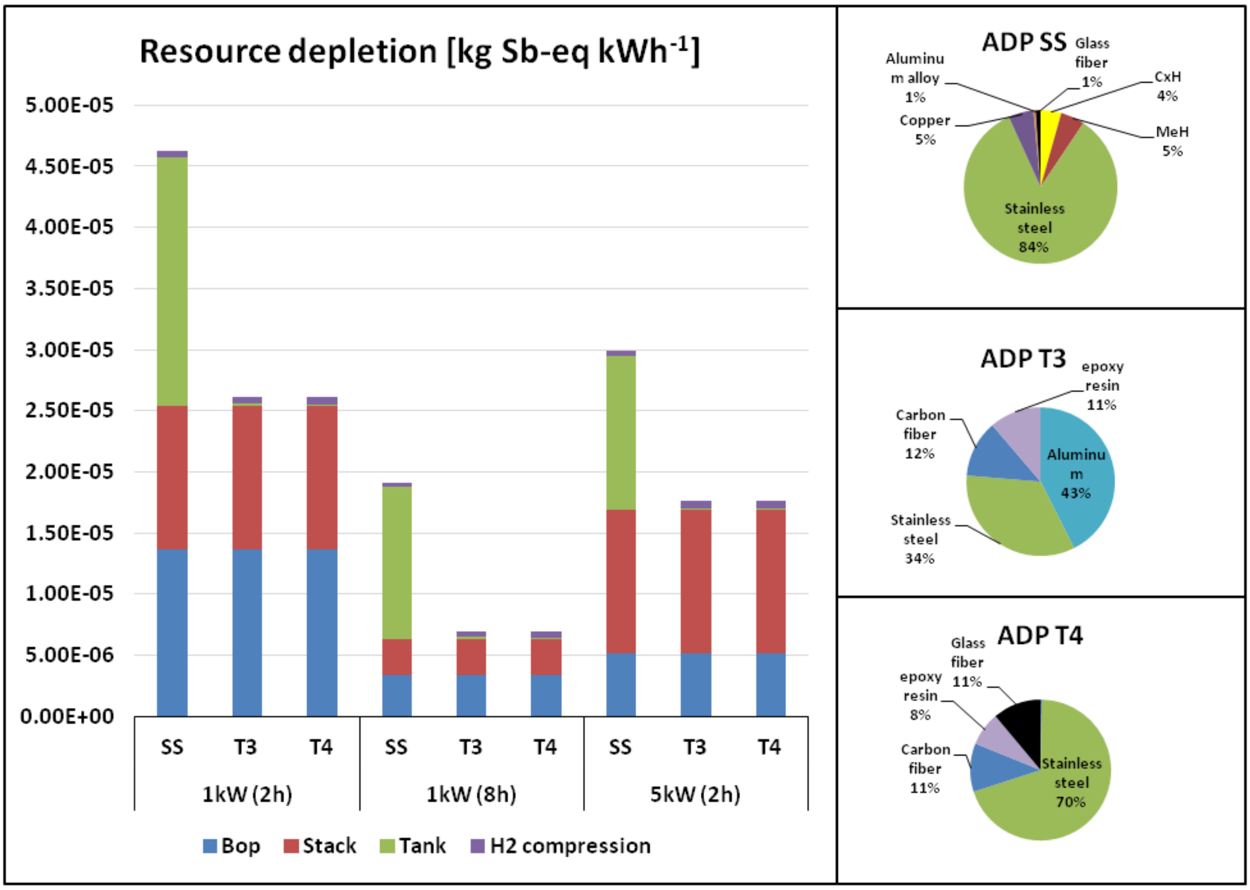

Fig. 7 Abiotic Depletion Potentials for the systems analysed. Left: contribution of the subsystems; right: materials contribution for the tanks of 1 kW systems running for 2 hours.

For this impact category, the contribution of the operation of the system (i.e. the hydrogen compression) is almost negligible. In fact, the construction of the systems represents basically the totality of the ADP. About the differences among the considered storage tanks, the SS system is by far more resource consuming than the T3 and T4 systems, where the tank production has a negligible impact compared to the stack and BoP construction.

Surprisingly, for the SS tank, the highest contribution to ADP is not due to the metals used for the hydrides, but rather to the structural materials (mostly steel). It should be noted that the analysis does not include the end of life of various systems, therefore the possible recycling of the materials is not accounted for.

2.4 Economic analysis

A cost analysis has been performed to evaluate the economic aspects of the APU based on the prototype hydrogen tank, in comparison with solutions already available in the market. Since, apart from the prototype tank and BoP, the other components (fuel cell, hydrides and buffer) are commercial products, the starting point for the cost analysis is the production of 1 APU unit. The perspective costs for a large-scale production of 10000 APU were then estimated getting quotations for large quantities. It must be pointed out that the prices of commercial type III and IV tanks are average values obtained from different quotations: the prices of these devices in fact can vary significantly according to manufacturer, operating pressures and number of tanks to be purchased. The detailed costs are reported in table SM5. The cost distribution among the main components of considered APUs is reported in fig 8.

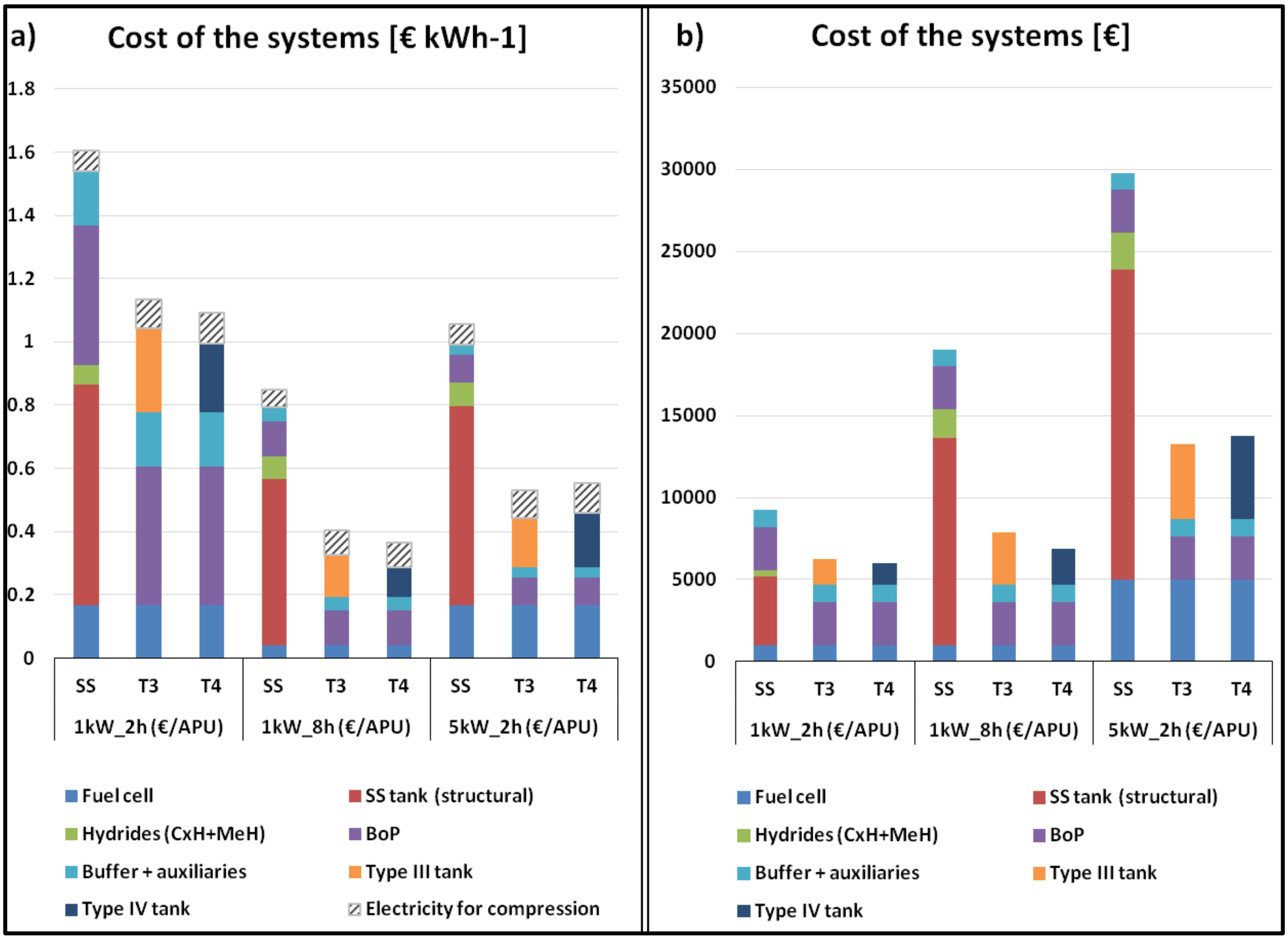

Fig. 8 Costs of the systems modelled with analysis of the contribution of the subsystems: a)cost per kWh including the electricity for hydrogen compression , b) total cost.

Fig. 8 shows the dominant role of the tank on the total costs of the SS systems, followed by the BoP and the stack, while, for the T3 and T4 systems, the part mostly contributing to the cost is the BoP. However, as clearly shown in fig. 8, the contribution of the hydrides represents only a small part (less than 10% for all SS systems) with respect to the structural part (from 45 to 66%).

The cost of the T3 and T4 systems is significantly lower than that of the SS tanks, as expected. Another significant contribution is given by the BoP (from 9 to 54%). Similarly to the tank, in fact, the BoP of SS systems is a prototype, made by assembling commercial components, that in some cases are not even optimized for the specific application.

The inclusion of the cost of the electricity for the hydrogen compression does not change significantly the results. In facts, even if the average European cost per kWh for households (0.211 € per kWh, [56]) is added to economic analysis, the lower electricity consumption of the SS system does not compensate for the higher costs for investment (see fig. 8). There is, however, a clear economy of scale in using larger energy storage systems and smaller fuel cells.

4. Discussion

With this study, we have analysed nine different APUs configurations based on HT-PEM fuel cells, considering both the environmental impacts of production and operation for a life time of 10 years. As expected, and shown by the material inventories, a prototype tank based on solid-state hydrogen storage materials requires a significantly higher amount of materials for its construction, with respect to pressurized gas type III and IV tanks commercially available. On the other hands, it requires an operating pressure (70 bar) much lower than type III and IV tanks (350 and 700 bar, respectively), and therefore a significantly lower amount of electricity for hydrogen compression is necessary in the use phase.

About climate change, the energy saved along the operation of the SS systems practically pays back the additional emissions during the construction phase with the current electricity mix. With a potential future electricity mix, less relying on fossil fuels, the GHG saved during the operation are, instead, not enough to payback the higher construction emissions. In addition, when the electricity for compression is included, the primary energy consumption is even slightly lower for SS systems than for the T3 and T4 tanks. The SS systems may result even more efficient if a longer lifespan would be assumed. There are, therefore, trade-offs between climate change and energy efficiency. The ADP is instead always much higher for the systems adopting solid-state hydrogen storage. However, the results may be different if the end of life of the systems, and therefore the recycling of the materials, would be considered. It may be speculated that the recycling of metals, of which it is mostly made in the solid-state tank, is easier than the recycling of fibres, which have a higher share in the pressure tanks T3 and T4.

For all the analysed impacts (i.e. GWP, ADP and PED) we have found, for the SS systems, a high contribution from the structural materials used for the BoP, mostly steel.

The impacts due to the hydrides in the tank appear instead rather limited. These values, not yet available in any LCA inventory, have been estimated for the first time, considering the whole processes of production and ball milling. Dealing with a prototype tank, the estimation has been carried out as an extrapolation of lab-scale activity. In case of a scale-up, including a better industrial management of the whole process, these contributions might be further reduced.

Similar arguments can be drawn for the cost analysis. The costs of hydrides are much lower than the cost of the structural materials and the BoP. Even in this case, these high costs are probably because, although we have estimated the costs for mass production, the system analysed is a prototype, far from being commercially optimized. Considering costs per kWh of energy production from the APU, the total cost of SS systems, although non-optimized, is about 1.5 – 2.0 time the cost of the T3 and T4 ones. It should be also noted that the SS systems have the added value of being considered safer [57,58], because of the lower hydrogen pressures involved in the management.

5. Conclusions

To conclude, by accounting for the electricity consumption for hydrogen compression, APU systems using tanks based solid-state hydrogen storage materials have similar environmental performances than tank III and IV compressed gas systems. However, the ADP is higher, but the inclusions in LCA of the end of life of the APUs and the recycling of the materials may result in different conclusions for this impact category. The costs of SS APUs are significantly higher with respect to T3 and T4 APUs, however we are comparing mature technologies with a prototype which has much room for optimization. To improve both the environmental and economic performances of SS systems, an important reduction of the structural materials of both the tank and the BoP is recommended.

In short, the conclusions of this work can be summarised as follows:

- The manufacturing of SS hydrogen storage systems generates GHG emissions much higher than pressure vessels, and, while pressure vessels GHG emissions are a fraction of the manufacturing of the BoP and Stack together, in the case of SS systems they are similar to twice the combined emissions of BoP and Stack. This is due to the much larger use of materials.
- The consumption of resources, Abiotic Depletion, is, similarly to the GHG emissions, much higher for SS systems, again because of the larger use of materials (tough most materials can be recycled, the recycling is not included in this study).
- The Primary Energy Demand is, instead, higher for pressure vessels, mostly because of the higher requirement of electricity for compression.

- The costs of construction and operation are much higher for SS systems than pressure vessels.

Future and further works should be aimed at including in the analysis the end of life of the structures, other impact categories and other aspects such as safety.

Obtained results fill a knowledge gap in the assessment of hydrogen storage technologies. In fact, economic and environmental performances in real applications for APU have been identified for solid-state hydrogen storage tanks in the current state of development.

Acknowledgments

The research leading to these results has received funding from the European Union's Seventh Framework Programme (FP7/2007–2013) for the Fuel Cells and Hydrogen Joint Technology Initiative under SSH2S project, grant agreement n. 256653.

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