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Life cycle assessment of fuel cell systems for light duty vehicles, current state-of-the-art and future impacts

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ABSTRACT

Electric vehicles are a key technology for achieving a significant reduction of greenhouse gas directly emitted by the fleet of light duty vehicles. In the past, the production impacts of alternative vehicle technologies have been widely assessed within the life cycle assessment framework, with large uncertainties regarding fuel cell electric vehicles (FCEVs). FCEVs allow an almost double driving range with a single charge or refill of the tank and more than ten times the fueling/charging rate compared to batteries. However, FCEVs currently suffer from two major issues, lack of widespread fueling infrastructure coverage and high production costs, compared to the well-established Li-ion batteries. Furthermore, the data used for life cycle assessments of the technology should be constantly updated, as their technological development is rapidly moving forward. In this regard, an early detection of the likely environmental impacts from this fast development is fundamental to guide the progress of the technology.

In this study we perform a life cycle assessment of a fuel cell system for FCEV currently on the market. We found that the production of the tanks, the catalyst and the fuel cell auxiliaries are the components with lower environmental performance of the system, across all the investigated impact categories. Currently, the production of a fuel cell system with a net power output of 80 kW, and two storage tanks with a total capacity of 5 kg of H_2, generates approximately 5 ton CO_2-eq. In addition, in line with the targets set by the US Department of Energy, we performed an assessment of the prospective technological developments to identify its future impact. In the assessed prospective scenarios, we analyzed the effects of the technical improvements, and we subsequently combined them with the use of a higher share of renewable energy sources and secondary platinum. In essence, the technology shows potential reduction of the environmental footprints ranging from 25% to 70%, depending on the impact category. However, to achieve these results, the combination of renewable energy sources and a high learning rate must take place.

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1. Introduction

In 2010, light duty vehicles (LDVs) were the largest contributors to direct emissions from the transport sector, with approximately 10% of the total greenhouse gas (GHG) emitted in that year from all sectors (Edenhofer et al., 2014). As global population and well-being increase, LDV sales are also expected to grow, with some models forecasting a global stock between 2 and 3 billion vehicles in 2050 (Hao et al. 2016; Yeh et al., 2017). Given the challenges imposed by local air pollution and global warming, fully electric vehicles (EVs) rose as a key technology for decarbonizing the transport sector. The future penetration rate of EVs is still uncertain, but with many of the big markets such as China, the EU and India pushing for their market uptake, the total stock is expected to reach a significant share of the total LDVs stock by 2050 (Bunsen et al., 2019).

Within the domain of EVs, two powertrain technologies exist: fuel cell electric vehicles (FCEVs) and fully battery electric vehicles

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(BEVs). BEVs rely on Li-ion batteries, to achieve desired power and energy requirements, while FCEVs rely on fuel cell (FC) systems. There are several variants of FC systems which differ in the fuel and the electrolyte used, with consequently different performance and most-suitable application areas. Regarding electro mobility, polymer electrolyte membrane FC (PEMFCs) fueled by compressed gaseous hydrogen is the technology of choice (Mekhilef et al. 2012).

The current stock of EVs is dominated by BEVs (International Energy Agency 2018), while FCEVs, for a number of reasons, are not yet considered sufficiently mature for large-scale commercial deployment. FCEVs offer attractive features such as fueling times of approximately less than 5 min (Reddi et al., 2017) and long driving ranges (U.S. EPA 2019) in comparison to their BEV counterparts. On the other hand, disadvantages such as lack of adequate infrastructure, i.e. refueling stations, and high costs both industry- and consumer-wise (Staffell et al., 2019) are a major limitation for a market penetration of the technology. However, several original equipment manufacturers are investing in this technology and a mass scale production of FC systems would lead to a quick drop of the production costs, similarly to the effects experienced with the battery packs for BEVs since their large-scale deployment.

Research on FCEVs still aims to achieve competitive costs at high production volumes, in comparison to BEVs and internal combustion engine vehicles. To this end, the hydrogen and fuel cells program within the United Stated Department of Energy (US DOE) is focusing on targets such as reduction of platinum loading, suitable bipolar plates and satisfactory cycle life (Brian D. James et al., 2016).

While neither EV powertrain technology have significant direct tailpipe emissions, previous studies have found that the potential low impacts in the use-phase of FCEVs and BEVs (when coupled with renewable energy sources) are partly offset by the higher production impacts of EVs (Ellingsen et al., 2016a,b; Evangelisti et al., 2017; Simons and Bauer 2015). This underlines the importance of shifting the largescale production facilities from high carbon economies to low carbon economies. It also underlines the importance on following up such studies with more detailed market regional impacts – so that when the new industry starts rolling, it is as clean as can be from the transition starting points.

Several studies attempted to evaluate the impacts generated throughout the production phase of (PEM)FCEVs, with a focus on the global warming potential impact category, measured in kg CO_2-eq. Commonly, FCEVs are compared against either BEVs or conventional vehicles. However, comparative studies often rely on previously published life cycle inventories (LCIs), which are then adapted to fit a particular case study. Among the original inventories, the work performed by Notter et al. (2015), Miotti et al. (2015), Simons and Bauer (2015) and Evangelisti et al. (2017) are the most prominent and recent studies performed in the life cycle assessment (LCA) dimension with respect to FCEVs.

Notter et al. (2015) compiled a detailed LCI for a PEMFC with a peak power of 90 kW. The results stemming from the impact assessment are further compared with BEVs and conventional vehicles. The study finds that the production of FCEVs has higher impacts than both BEVs and conventional vehicles, even considering that the impacts from the production of the FCEVs' hydrogen tanks were not accounted in the study. The most relevant components, in terms of greenhouse gas emissions, were the platinum used for the catalyst and the stainless-steel bipolar plates. Simons and Bauer (2015) analyzed a FCEV with a net power output of 40 kW and compared their findings with a gasoline vehicle. On a cradle-to-gate perspective, they have found that the production of FC systems generates higher impacts than conventional vehicles, mainly due to the production of the catalyst and the auxiliary components. However, this study does not take into account the hydrogen tanks in their system (Simons and Bauer 2015). Similarly, Miotti et al. (2015) compared the potential impacts of a FCEVs with conventional vehicles and BEVs, evaluating both current and prospective impacts. Their assessment, which included also the hydrogen tank, found that the combined production of the hydrogen tanks and the auxiliaries is responsible for almost 60% of the global warming potential (GWP) impacts, making these components the biggest contributors across the entire FC system. Finally, Evangelisti et al. (2017) assessed an 80 kW PEMFC, comparing their findings with a mid-sized gasoline vehicle and a BEV. Analogously to the previous studies, the authors identified FCEVs as the most impacting technology during the manufacturing phase. Their study found that the hydrogen tanks, the gas diffusion layer, and the fuel cell auxiliaries are the main contributors.

All of the above studies highlight how the current production of FCEVs has higher impacts than conventional vehicles and BEVs. However, due to the lack of accurate primary data, adding high resolution to the components modelled requires the introduction of assumptions, which lead to a somewhat large variation in the absolute results for GWP and the main drivers of these impacts. In addition, the application of FC systems to electromobility is developing at a fast rate, requiring a constant revision of the potential impacts generated by the current state-of-the-art and by the likely future developments.

In order to increase the understanding of the current environmental impacts associated with the production of FC systems, we performed an LCA of a PEMFC system for automotive applications, combining, as main sources of data, technical specification of vehicles currently on the market (Kojima and Fukazawa 2015; Yoshida and Kojima 2015), the work done by the Department of Energy (DOE) of the United States (B. James, 2018; B. D. James et al., 2016) and the most prominent and recent studies on the research frontier. For this study, three different scenarios were evaluated. The first scenario, namely reference scenario, aims to assess the current impacts due to the production of PEMFCs. The second scenarioidentifies the likely impacts of the technology as it progresses following the development targets set by the US DOE, in combination with higher share of secondary platinum used for the catalyst. The third scenario follows the same trend of the second scenario, with further technical developments combined with cleaner energy mixes for the production of carbon fiber, and ever increasing use of secondary platinum. Furthermore, we compare our inventory and findings with the previous LCA studied performed within the same context, and perform a sensitivity analysis on the key components source of the highest environmental impacts.

2. Methodology and case description

A PEMFC used in EVs is composed of three main components, that include several sub-components (Table 1). The main components are the FC stack, the fuel cell auxiliaries and the storage tanks. The FC stack accounts for all the components responsible for the generation of electrical energy due to the chemical reaction between hydrogen and oxygen. The fuel cell auxiliaries ensure that the stack operating conditions are maintained at adequate levels by managing the flows of water, hydrogen, heat, and air in and out of the system. The storage tanks contain gaseous hydrogen that is channeled to the FC stack and that reacts with oxygen to produce electrical energy, fed to the electric motor, and water and heat as by-product.

In modelling the FC system, components were sized using different key parameters (Fig. 1). The net power output of the FC system was used to size the FC system, and consequently defines the gross power output, the total area and the active area of the system. These parameters in turn defined the components masses

Table 1
Main components and sub-components included in a PEMFC for EVs. The components included in this table were used for the LCI compiled for this study.

Primary component	Sub-components	
Fuel cell stack	- Membrane	- Gaskets
	- Bipolar plates	- Membrane electrode assembly
	- Catalyst ink	- Stack compression bands
	- End plates	- Stack housing
	- Current collectors	- Coolant
H_2tanks	- Tanks (carbon fiber and resin)	- Tank auxiliaries (boss, foam, glass fiber)
Fuel cell auxiliaries	- Air management	- Fuel management
	- Water management	- Heat management
	- Electronics (controller, sensors, wiring)	

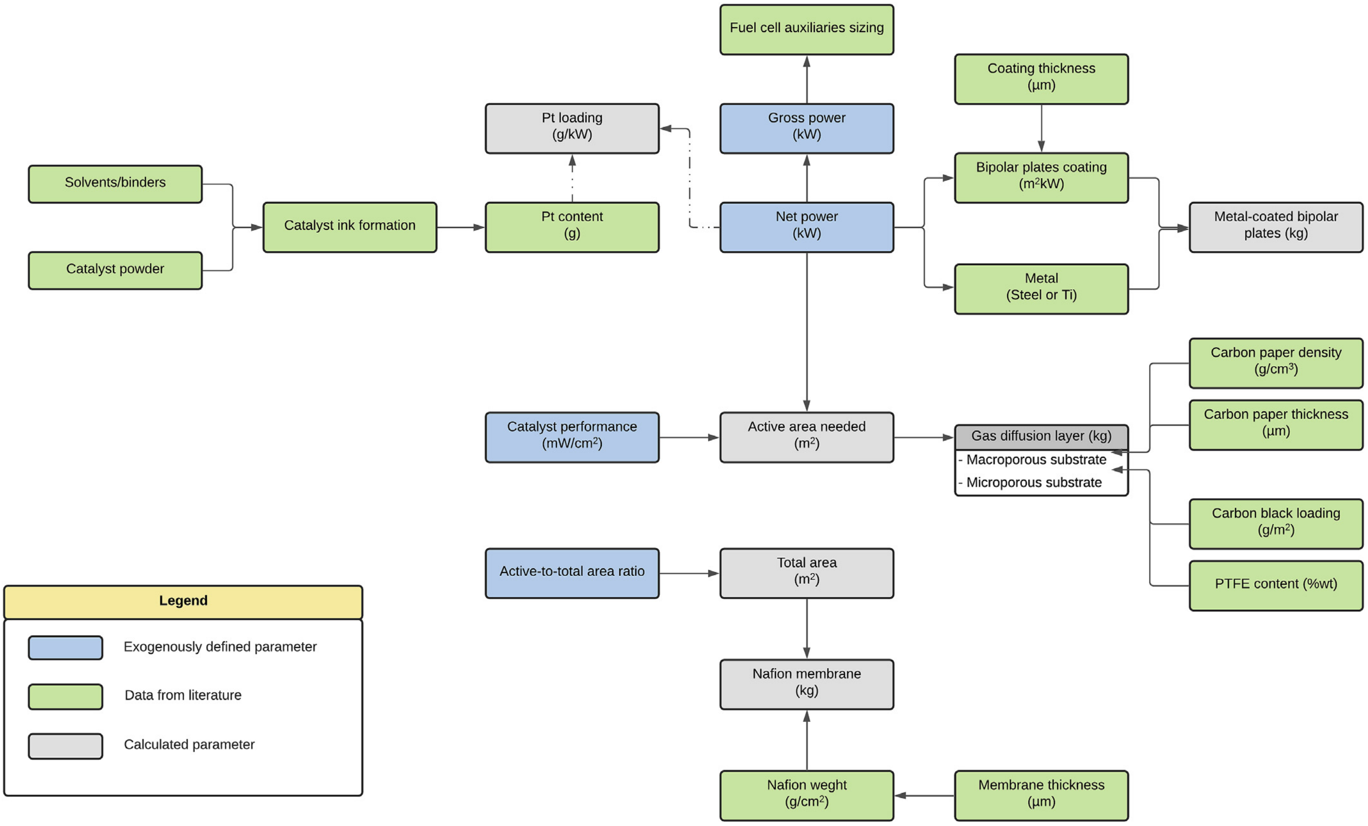

Fig. 1. This figure gives a flow chart of the system modelled in this study and the relationships between the parameters exogenously defined and the sizing of the components.

and the material requirements. To calculate the active and total areas of the system, the expected catalyst performance, measured in mW/cm^2, and the active-to-total-area ratio were used. Given the assumed performance of the catalyst, the two areas were calculated as follows:

$$Active\ area\ [m^2] = \frac{\text{Net power output of the system [kW]}}{\text{Power density of the catalyst } \left[\frac{kW}{m^2}\right]} \qquad \text{[Eq 1]}$$

$$Total\ area\ [m^2] = \frac{\text{Active area } [m^2]}{\text{Active} - \text{to} - \text{total area ratio}} \qquad \text{[Eq 2]}$$

The main parameters, once identified and calculated through Equations [1] and [2], were combined with the material requirements found in the literature for the remaining components, allowing for the completion of the life cycle inventory.

The inventory compiled for the FC system uses ecoinvent 3.6 as background database (Moreno Ruiz et al., 2019; Wernet et al., 2016) and the ReCiPe 1.13 impact assessment methodology to estimate the total contribution to the different impact categories covered by the study (Huijbregts et al., 2016). To perform the LCA, we used ARDA, a software developed in-house at the Industrial Ecology Programme at the Norwegian University of Science and Technology.

To estimate the environmental impacts of the FC system, we performed a cradle-to-gate life cycle assessment for a PEMFC system including the FC stack, the balance of plant and the hydrogen tanks. For the calculation of the impacts, the functional unit was the production of an 80 kW net power output PEMFC system, which is a power rating representative of mid-sized vehicles.

The following sections describe the assumptions and main data sources used. We then compare the results of our inventory with previous studies, to identify differences and likely sources of uncertainty in the results. To tackle the uncertainty in the data collected, we performed a sensitivity analysis on they key components, with the goal of identifying where slight variations in the weight of the component may give significant variation in the total absolute impacts. A structural path analysis was performed to identify the most environmentally intensive processes across the value chain of key materials. The full inventory can be found in the Supporting Information, together with the results stemming from the sensitivity analysis.

2.1. Catalyst

Currently, platinum (Pt) alloys deposited on a porous carbon layer is the technology of choice for PEMFCs in FCEVs, providing the highest performance and, at the same time, ensuring the required operational lifetime. Based on the analysis performed by the DOE and the catalyst applied to the Toyota Mirai, we modelled the catalyst as a slurry of Pt and cobalt (Co) deposited on a high surface area carbon (HSC) layer (James 2018; Kojima and Fukazawa 2015; Yoshida and Kojima 2015). With the modelled catalyst, the stack achieves a power output of 1095 mW/cm^2 (James 2018). For the production of the catalyst, the powder preparation and the ink formation were modelled based on James and colleagues work (2018; 2016). Regarding the ink formation, a solution of water (37.5 wt %), methanol (37.5 wt %) and Nafion (10 wt %) mixed with the PtCo/HSC powder (15 wt%) was assumed (O'Hayre et al., 2016). Given the performance of the catalyst and the net power of the system, the active area needed by the system was calculated. In addition, the active-to-total-area ratio provided by James (2018), was used to obtain the total area.

In a FC stack of approximately 100 kW of power output, 30 gr of Pt are used (Pollet et al. 2019). This parameter, combined with the active area previously calculated led to an estimated current Pt loading of 0.32 mg/cm$^2_{\text{active}}$, or 0.29 g/kW$_{\text{net}}$. For the reference scenario we assumed that 30% of the Pt used in the catalyst is secondary, in line with the current market flows published by Johnson Matthey for the year 2018 (Bloxham et al., 2019).

2.2. Membrane

A PEMFC requires the use of membranes capable to allow ionic transport, be gas impermeable, and minimize the resistance through reduced thickness (O'Hayre et al., 2016). In this regard, perfluorinated polymers are widely used, of which Nafion® is the material of choice. Nafion is obtained by the copolymerization of tetrafluoroethylene (TFE) and perfluoroalkyl sulfonyl fluoride (Connolly et al. 1966). As with the Pt loading, different membrane thicknesses are assumed in the literature. Evangelisti et al. (2017), who based their work on Carlson (2005), assumed a dispersion cast membrane 50 μm thick in their baseline scenario, similarly to Simons and Bauer (2015). However, Miotti et al. (2015) assumed a thickness of 12.5 μm. Kojima and Fukazawa (2015) stated that between the Toyota Mirai sold in 2008 and the newer version put on the market in 2014, a reduction by 67% of the thickness was achieved. Thus, we expect that current FC stacks have membranes with a thickness in the range of Miotti's values, or below. Indeed, James (2018) assumed a thickness of 14 μm using Nafion 850 EW, supported on expanded polytetrafluoroethylene (ePTFE). For our study, we assumed a thickness of 25.4 μm and Nafion 850 EW supported on ePTFE (10 wt%) in the baseline scenario.

2.3. Gas diffusion layer

The gas diffusion layer (GDL) serves several functions in a PEMFC: (1) allow the gas reactants to reach the catalyst layers, (2) provide permeability for water removal, (3) conduct electricity from the bipolar plates to the catalyst layers, among others (Mathias et al., 2010), and remove heat. The gas diffusion layer consists of a porous layer and a microporous layer (MPL) (Park et al., 2012). The GDL employs graphitized carbon fibers in the form of carbon cloth, carbon felt or carbon paper. The different materials offer different advantages and disadvantages, but carbon papers seem preferred over carbon cloth due to lower thickness and lower ohmic loss (Jayakumar and Sethu 2015). For this study, we modelled a carbon paper GDL produced from polyacrylonitrile (PAN), which is the most common precursor for carbon fiber (Mathias et al., 2010). Furthermore, to enhance the water management and prevent flooding, a hydrophobic treatment is commonly applied to the GDL, dipping the material into an aqueous solution of PTFE (O'Hayre et al., 2016). To size the GDL, we assumed a thickness of 210 μm (Kocha 2013) and a density of 0.45 g/cm^3 (O'Hayre et al., 2016).

The MPL is a solution of carbon black and PTFE, and enhances water removal and electronic conductivity (Popov, Park, and Lee 2017). Regarding the carbon black, Jayakumar and Sethu (2015) performed an extensive literature review and identified a large variation of loadings (mg/cm^2) used. Thus, we took the average corresponding to approximately 3 mg/cm$^2_{\text{active}}$. Finally, a PTFE content of 15 wt% on carbon paper, was identified as the best performing and average value used in the literature (Jayakumar and Sethu 2015; Popov et al., 2017).

With the parameters mentioned above, we estimated a GDL with a weight of 1 kg, both lighter and thinner than previous studies. (Evangelisti et al., 2017; Miotti et al., 2015; Simons and Bauer 2015).

2.4. Bipolar plates

The bipolar plates (BPPs) of a PEMFC are made of either high-density graphite or coated stainless steel. The former solution offers excellent corrosion resistance, high heat management and good electrical conductivity (Taherian 2014). However, graphite-based BPPs are more suited for industrial applications, while for electromobility they present several limitations, such as brittleness and high volume and weight (Wang and Turner 2010). Hence, for automotive applications the use of metallic BPPs is common, with Toyota opting for this solution in its FC vehicle (Yoshida and Kojima 2015). The main strengths of metal BPPs over their composite-based equivalent are low cost, low thickness, high conductivity and chemical stability (Wang and Turner 2010). To avoid potential contaminations to the active section of the stack, a coating layer preventing its corrosion is applied (Taherian 2014). Toyota, in its 1st generation Toyota Mirai used gold-coated stainless steel BPPs (Kojima and Fukazawa 2015), while in the 2nd generation, lighter BPPs made of titanium coated with amorphous carbon were used (Yoshida and Kojima 2015). Regarding its sizing, we based our calculations on the DOE, that we further reduced since the 0.4 kg/kW$_{\text{net}}$ target for 2015 was achieved (Wang and Turner 2010). Regarding the coating, we used carbon black as proxy for amorphous carbon since it is its main constituent (Schwarz and Langenhove 2013). Finally, based on Yoon et al. (2008), we assumed a thickness of 10 μm for the estimation of the coating material needed.

2.5. Other stack components

Apart from the components described above, a FC stack consists of the membrane electrode assembly (MEA) auxiliaries, the end plates, the current collectors, the end-gaskets and the stack housing with compression bands. The data was mainly derived from James et al. work (2016) and Shah et al. (2004).

For the MEA, we accounted for plastic (polyethylene) required for the sub-gaskets, the resin for the seal between the BPP and the sub-gasket, and the energy required to hot press the catalyzed membrane to the GDL (Shah et al., 2004). To size the sub-gaskets of each cell, we based our model on the geometric size covered by the gaskets for each cell and assuming that the gaskets are made of silicone rubber (Evangelisti et al., 2017). The endplates, based on the system modelled in James et al. (2016), are made of a compression-molded composite called LYTEX® 9063, which consists of 63% glass fiber and 37% epoxy resin (Quantum Composites 2013). In total, 2.13 kg of LYTEX 9063 were assumed for the two endplates, and 7 kWh of electricity for the compression molding and the quality control processes. The current collectors of the system fit within the end plate, and are made of a copper foil and two copper studs, all of which weights a total of 0.5 kg, as described by James et al. (Brian D. James et al., 2016). The end gaskets were based on the data from Shah et al. (2004), where they assumed a resin for their manufacturing. Due to the lack of the same material in our database, we used epoxy as a proxy. Finally, the model accounts for the stack compression bands, made of stainless steel, and the stack housing that was modelled as 0.5 mm thick polypropylene composite.

2.6. Fuel cell auxiliaries

In addition to the stack itself, a FC system also has auxiliaries, which can be divided into four main components: heat management, fuel management, air management and water management. Note that in this paper we refer to the auxiliaries as what in the literature is often called balance of plant. The data available on these components is mainly based upon the work performed by Carlson et al. (2005), which, likewise Miotti et al. (2015) and Evangelisti et al. (2017), was the basis for most of our inventory. However, where possible we updated the bill-of-materials (BOM) and the components used based on the information provided by James et al. (2016). Due to the outdated data points used, and their similarity to the previous LCA performed on this topic, the results may be aligned with the literature. However, their robustness is uncertain, since no data is available regarding the current state-of-the-art in terms of materials and components used for the FC auxiliaries. Thus, we performed a sensitivity analysis on each component of the FC auxiliaries.

The air management system is made of four components, with the compression expansion motor being the heaviest piece, with a weight of 5.7 kg. Regarding its material composition, the work of Sinha et al. (2008) was taken as a reference given the highly detailed BOM. The other components, namely the air filter, air ducting and mass flow sensor, were entirely based on the BOM in Carlson et al. (2005).

Regarding the water management, we accounted for the humidifier, the demister, the air pre-cooler and the Nafion tubing. Regarding the humidifier, the data contained in Sinha et al. (2008) and Carlson et al. (2005) were used. Furthermore, a humidifier uses a honeycomb material, commonly cordierite. Polyurethane was used as a proxy, since it can be used as precursor for the production of cordierite (Carty and Lednor 1996). James et al. (2016) modelled the air pre-cooler as 100% aluminum, from which we calculated its weight using the prices of the component and of the aluminum.

To model the heat management, we combined the data in Carlson et al. (2005) and in Sinha et al. (2008), where the weight and material composition of the antifreeze liquid, the fan and the radiators are described. The work from Sinha et al. (2008) was also the reference for modelling the material composition of the fuel management, i.e., blowers, ejectors and pipes. Finally, we considered the electronics required for the sensors, the wiring and the mounting frames and fasteners used for the correct placement and communication of the system with the electronic control unit of the vehicle.

2.7. Hydrogen tanks

To store hydrogen at high pressures, Type IV 700-bar storage systems are the current state-of-the-art (Hua et al., 2017). The composite material used in these tanks is modelled using 60% wt. Toray T700 carbon fiber (TORAYCA 2019) and 40% wt. epoxy resin (Hua et al., 2017). Moreover, in combination with the composite, we accounted for auxiliary components of the tank, which consist of bosses, the plastic tank liner, valves and a regulator (Ahluwalia et al., 2016; Hua et al., 2017). The main materials for these components are foam, glass fiber, high density polyethylene, aluminum and chromium steel. We modelled a two-tank system yielding a capacity of 5 kg of hydrogen, which parallels the tank solution equipped in the Toyota Mirai, and ensures a range above 500 km with current fuel efficiencies. Ahluwalia et al. (2016) developed a model on ABAQUS to analyze the tanks equipped by Toyota in its FCEV, providing the BOM that guided the inventory compilation of this study. Regarding the energy required for manufacturing the tank, we based our assumption on Miotti et al. (2015) that estimates a conservative value of 20 kWh. Evangelisti et al. (2017) assumed 4.5 kWh, while Simons and Bauer (2015) do not include the tank in their system.

For the production of carbon fiber, the production steps described by James et al. (2016) were taken as a reference due to the high level of details. Carbon fibers are prepared using polyacrylonitrile (PAN) as precursor, which is a polymer manufactured through the polymerization and electrospinning of acrylonitrile (95% wt.) and methacrylate (5% wt.), with a synthesis yield of 58% (James et al., 2016; Meng et al., 2017). Once the electrospun roll is prepared, oxidation and carbonization processes occur, which lead to a total consumption of heat and electricity of approximately 118 MJ and 41 kWh per kg of carbon fiber produced, respectively (James et al., 2016).

2.8. Assessed scenarios

The data and assumptions described in the previous sections represent the reference scenario, which assesses the production impacts from current FCEVs. However, the technology is developing at a fast rate, with consequential improvements regarding the materials used per stack and the energy required to produce the components.

Regarding the technical developments of the technology, we modelled two scenarios aligned with the DOE targets for FCEVs in 2020 and 2025 (James 2018). Table 2 shows the modelling parameters used for both the reference scenario and the future scenarios assessed. The datapoints marked with an asterisk were taken from the work presented by James (2018). We focused on membrane thickness, stack power density, Pt loading, metal-coated BPPs. In addition to the US DOE targets, other likely developments such as less carbon-intensive energy sources for the production of the tanks, coupled with a more efficient production, and a constantly increasing share of secondary platinum were evaluated. Thus, as the scenarios assessed are including both technical targets and likely improvements, we differentiate these two aspects by referring to them as d*evelopment stage 1* and d*evelopment stage* 2. For the BPPs, following the DOE targets, we assumed that titanium will be replaced with cheaper stainless steel coated with gold c. In addition, we assumed that their thickness will decrease over time, based on the developments observed in the previous years (Argonne National Laboratory 2017), assuming material requirements of 0.25 kg/kW$_{net}$ and 0.2 kg/kW$_{net}$ for the development stages 1 and 2, respectively. Previous studies have found that the production of the carbon fiber tanks is a major hotspot of emissions, due to the high energy requirements. Hence,

Table 2
Main modelling parameters and component weights for all scenarios analyzed.

Technical specifications	Unit	Reference scenario	Development stage 1	Development stage 2
Net power	*kW*	**80**	**80**	**80**
Stack power density	*mW/cm^2*	**1095***	**1250***	**1500***
Active area	*m^2*	**7.31**	**6.4**	**5.33**
Total-to-active area ratio		**0.625***	**0.625***	**0.65***
Total area	*m^2*	**11.7**	**10.2**	**8.2**
Pt loading	*mg/cm$^2_{active}$*	**0.32***	**0.19***	**0.135***
Share of secondary Pt	*%*	**30****	**50****	**75****
Nafion membrane thickness	*µm*	**25.4***	**10***	**10***
Storable H_2	*kg*	**5**	**5**	**5**
Subsystem weights				
Tank system	*kg*	**105**	**105**	**105**
Fuel cell auxiliaries	*kg*	**59**	**59**	**59**
FC stack	*kg*	**45**	**38**	**33**
Total weight of FC system	*kg*	**208**	**205**	**205**

The * indicates a parameter part of the US DOE targets (James, 2018). The ** indicates our own assumption. The remaining datapoints, except for the storable H_2, are parameters calculated based on *.

we assumed that the energy intensity associated to the production of carbon fiber will constantly decrease, with a 15% reduction in development stage 1 and a 30% reduction occurring within the development stage 2. Given the likely developments of the European electricity mix, aligned with the Sustainable Development Scenario (IEA 2018), we applied a future mix with a 50% share of renewable energy sources. This cleaner electricity mix was used to test the sensitivity of the carbon fiber production for the hydrogen tanks and the GDL. Furthermore, the sensitivity cases assume higher share of secondary Pt used for the production of the catalyst, with 50% and 75% in the development stage 1 and 2 scenarios, respectively.

3. Results and discussion

3.1. Global warming potential (GWP)

The production of the tank and the catalyst combined contribute to 53% of the GWP. Other notable sources of GHG emissions are the auxiliaries (17% of total impact), and the titanium-made BPPs (14%). In total, 5 tons CO2-eq are generated throughout the manufacturing phase of the FC system (Fig. 2).

The production of the storage tanks was identified as the main contributor to the GWP impacts due to the highly energy intensive manufacturing of carbon fiber. The 2 tons CO2-eq generated by the tank's production make up 40% to the total GWP impacts. Particularly, the production of 60 kg of carbon fiber was identified as source of 36% of the total impacts, due to total utilities requirements summing up to 2.5 GWh of electricity and 7.3 GJ of heat for its production. For the production of carbon fiber, we assumed an average European electricity mix and heat produced from natural gas. With these assumptions, we have found that 75% of the impacts are stemming from the electricity generation, while the remaining 25% being generated from the heat production.

The bipolar plates, which is the heaviest component, is the third largest contributor to GWP by a single component. The high carbon intensity associated with titanium mining and refining were identified as main driver of impacts., whilethe coating applied to the BPP has a negligible impact.

The preparation of the catalyst is responsible for 24% of the total GWP impacts (1.1 ton CO_2-eq). The majority of the catalyst impact is due to the mining activities of platinum and the high amount of electricity used in this phase. The remaining impact is driven by the preparation of the catalyst powder, which involved the use of various acids, the disposal of spent solvents, and the treatment of spent catalysts for the recovery of secondary platinum.

Carbon intensive materials are used in the auxiliary FC components: aluminum, TFE, the production of the magnet for the compression expansion motor and the electronics. The fuel cell auxiliaries are responsible for 17% of the total GWP impacts. In total, 0.8 tons of CO2-eq are generated for the production of the auxiliary components. Our results align with previous studies (Evangelisti et al., 2017; Miotti et al., 2015) basing their main assumptions on the work done by Carlson et al. (2005) like this study. However, our findings are significantly lower than Simons and Bauer (2015), which accounts for about 1.7 tons CO2-eq and based their inventory on the work performed by Notter et al. (2015) and the US DOE in 2007.

The water management system and the control units are the components for which it was not possible to find more accurate data and we therefore estimated the same BOM as Evangelisti et al. (2017) and Miotti et al. (2015). For the fuel and air management we combined the data from Carlson et al. (2005) with the work performed by Sinha et al. (2008). The water management recovery system has the largest impact contribution from the auxiliaries, primarily due to the Nafion tubes and the aluminum pre-cooler. Following, the electronics were identified as the second main source of impacts, due to the production of the wiring cables and the aluminum.

The Nafion membrane contributes to less than 2% of the total GWP impacts, mainly due to the production pathway of PTFE. The low amount of material employed led to comparatively negligible impact than the rest of the system, although the material proves to be particularly carbon intensive (approximately 200 $kgCO_2$-eq/kg of Nafion produced, based on our inventory). Regarding the other auxiliary components such as gaskets, housing and current collectors, together they add up to the remaining 4% of the impacts for GWP, not indicating any noteworthy carbon intensive materials and processes, with the given amounts employed.

3.2. Other impact categories

Other impact categories investigated (Fig. 3) follow the same trend as GWP, having the production of the catalyst, the auxiliaries and the tanks as the main drivers of emission, while the membrane, the BPPs and the rest of the stack do not contribute as significantly, and consistently, to the total impacts. Regarding Pt, the mining is a major contributor to particulate matter formation (PMFP), metal depletion (MDP) and terrestrial acidification (TAP). On the other hand, the sulfidic tailings and their off-site treatment were found

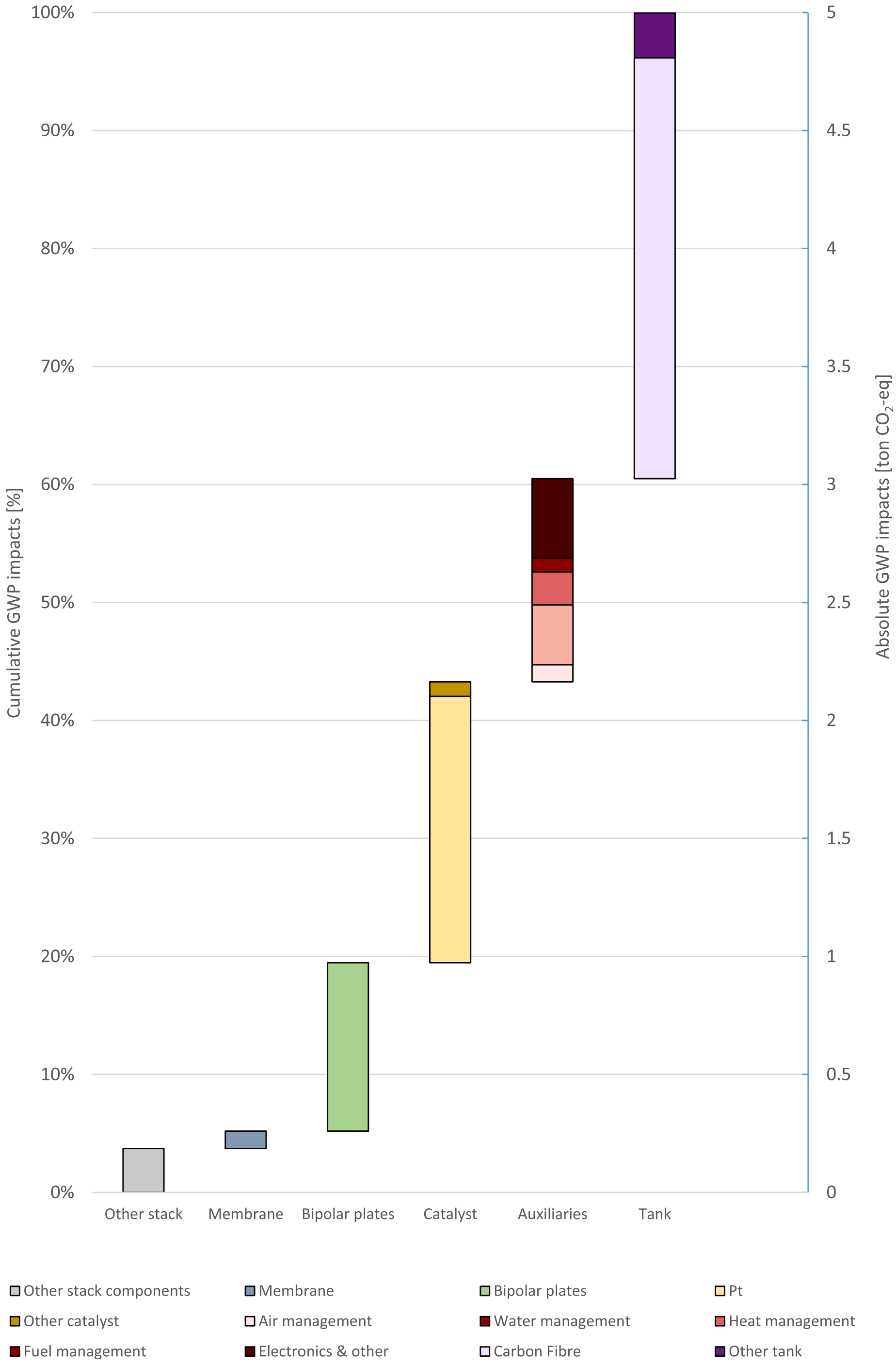

Fig. 2. Cumulative (left axis) and absolute (right axis) contribution to the GWP impact category of the 80 kW_{net} FC system in the reference scenario, split into fuel cell system components. Where possible, the component's impact was broken down to highlight the materials or components giving the highest contribution.

particularly relevant for human toxicity potential (HTP) and freshwater ecotoxicity potential (FETP). Furthermore, a large share of Pt is currently mined in South Africa, which strongly relies on coal. Given the high energy inputs required for mining Pt, the mine operations from coal were identified as a strong contributor to impact categories such as HTP, marine eutrophication (MEP), fossil depletion (FDP) and TAP.

The impacts generated by the production of the tanks are mainly driven by the production pathway of carbon fiber. More precisely, the Sohio process and the polypropylene precursor used for the production of acrylonitrile were found significant contributors for PMFP, MEP, FDP and TAP. Regarding the impacts stemming from the auxiliaries, the processing of aluminum and copper were found particularly relevant for the HTP and FETP, which are the impact categories where the auxiliaries impacts standout the most in terms of relative contribution.

In general, the mining activities required for the extraction and production of primary materials, e.g. Pt, Al and Cu, are the main drivers of impacts across several impact categories. Nevertheless, other impacts are associated with the indirect electricity

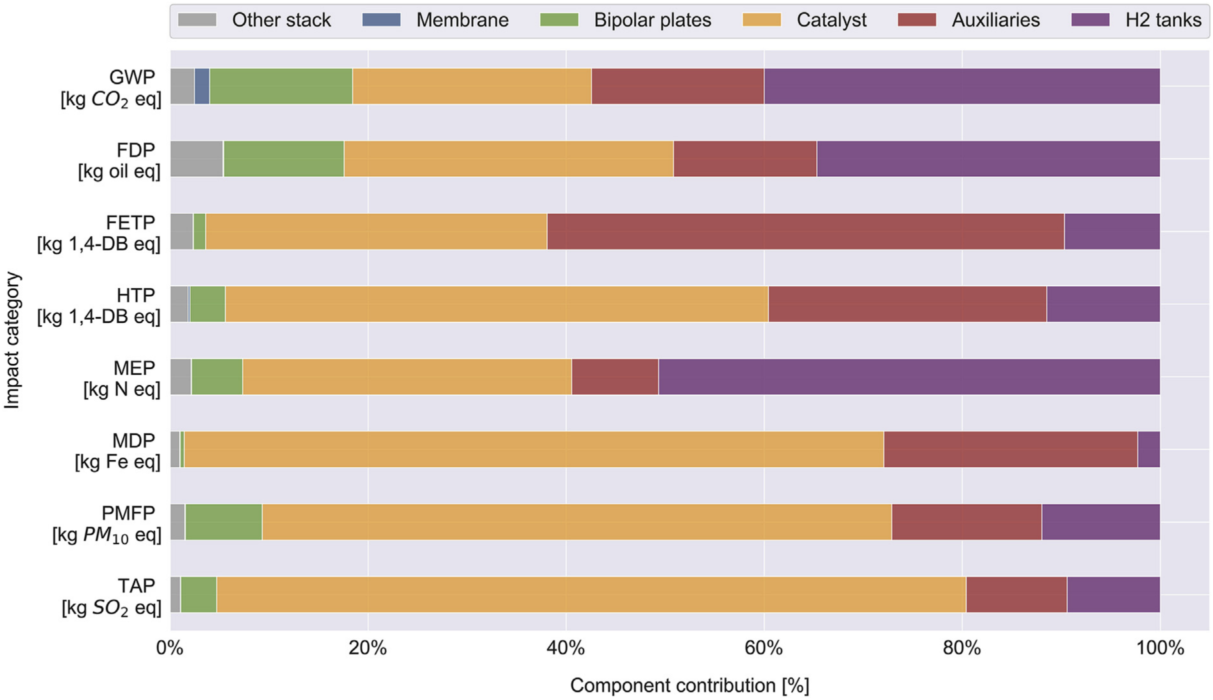

Fig. 3. Advanced contribution analysis for the impact categories investigated by the reference scenario. The acronyms used for the impact categories are: GWP – Global Warming Potential, FDP – Fossil Depletion Potential, FETP – Freshwater Ecotoxicity Potential, HTP – Human Toxicity Potential, MEP – Marine Eutrophication Potential, MDP – Metal Depletion Potential, PMFP – Particulate Matter Formation Potential, TAP – Terrestrial Acidification Potential.

requirements and the processing of raw materials. These findings suggest that significant impact reductions could be achieved by employing secondary materials, which would require lower energy inputs and can avoid the generation of the wastes linked with mining activities.

3.3. Comparison to previous literature

Fig. 4 compares our GWP results with those from literature, using a cradle-to-gate approach. For the reference scenario, our results lie between the findings from Miotti et al. (2015) and Evangelisti et al. (2017). The light-green frame in the figure includes the state-of-the-art LCA studies. The studies have been published on a timeframe of five years, thus some discrepancy in the results is expected, as more data on the technology became available over time. The largest differences are found in the production intensity of specific sub-components in the stack and the hydrogen tanks. Regarding the production impacts of the stack, the findings from Evangelisti et al. (2017) stand out far from this study and the work by Miotti et al. (2015). This is attributable to the graphite and carbon fiber requirements (in kg/kW_{net}) assumed for the production of the BPPs and the GDL, respectively. While the findings from Simons and Bauer (2015), for the other stack components, seem comparable to the findings by Evangelisti et al. (2017), the former's results were aggregated. In fact, their *Other stack* results include also the catalyst and membrane, that were aggregated due to lower resolution of the results presented in the paper, thus they cannot be directly compared due to lack of resolution.

Another major difference between studies is found in the production of the storage tanks. Results range from 18 kg CO_2-eq/kW_{net} (Miotti et al., 2015) to 51 kg CO_2-eq/kW_{net} (Evangelisti et al., 2017). Despite all the models use PAN as precursor for carbon fiber, the impacts vary due to (i) the energy intensity of carbon fiber production, ranging from 34 to 70 kWh/kg of carbon fiber, and (ii) to a lesser extent due to the different assumptions on the mass of the tanks. Evangelisti et al. (2017) modelled a single tank with 71.9 kg of carbon fiber and a total weight of 117 kg, while Miotti et al. (2015) assume 66.8 kg of carbon fiber and a total weight of the tank of 101.2 kg.

A modest variation was found for the Nafion membrane, where two thicknesses were used, 50 μm (Evangelisti et al., 2017) and 25 μm (this study, Miotti et al., 2015). The membrane modelled in our reference scenario gives the lowest impacts compared to previous studies. However, these difference is mainly due to the variation of impact factors in the backgorund database, ecoinvent, for TFE. In both literature and patents (Cells 1925; Connolly et al., 1966; Laird 1997), the fragmented data available on the polymer's preparation leads to modelling uncertainty, particularly regarding the yield at high production volumes, the energy use and the preparation of the precursors.

Regarding the catalyst, the results range from approximately 9 kg CO_2-eq/kW_{net} (Evangelisti et al., 2017) to approximately 14.6 kg CO_2-eq/kW_{net} (this study), with the work performed by Miotti et al. (2015) yielding 6.5 kg CO_2-eq/kW_{net}, per fuel cell stack. The Pt loading span from a minimum of 0.15 mg/cm^2 (Simons and Bauer 2015) to a maximum of 0.6 mg/cm^2 (Evangelisti et al., 2017). The results from this study are the highest compared to the literature, although total platinum loading lower compared to Miotti et al. (2015) and Evangelisti et al. (2017). The different results are driven by both different Pt loadings and preparation steps and also by the changes in the impact factors across the backgorund databases used.

The BPPs, due to the different technology assessed in this study, cannot be directly compared with previous studies. However, it can be noted that coated stainless steel bipolar plates proved to generate lower production impacts (Miotti et al., 2015) than their graphitic counterparts. Furthermore, material intensities (kg/kW) for metallic BPPs are being constantly reduced (Yoshida and Kojima

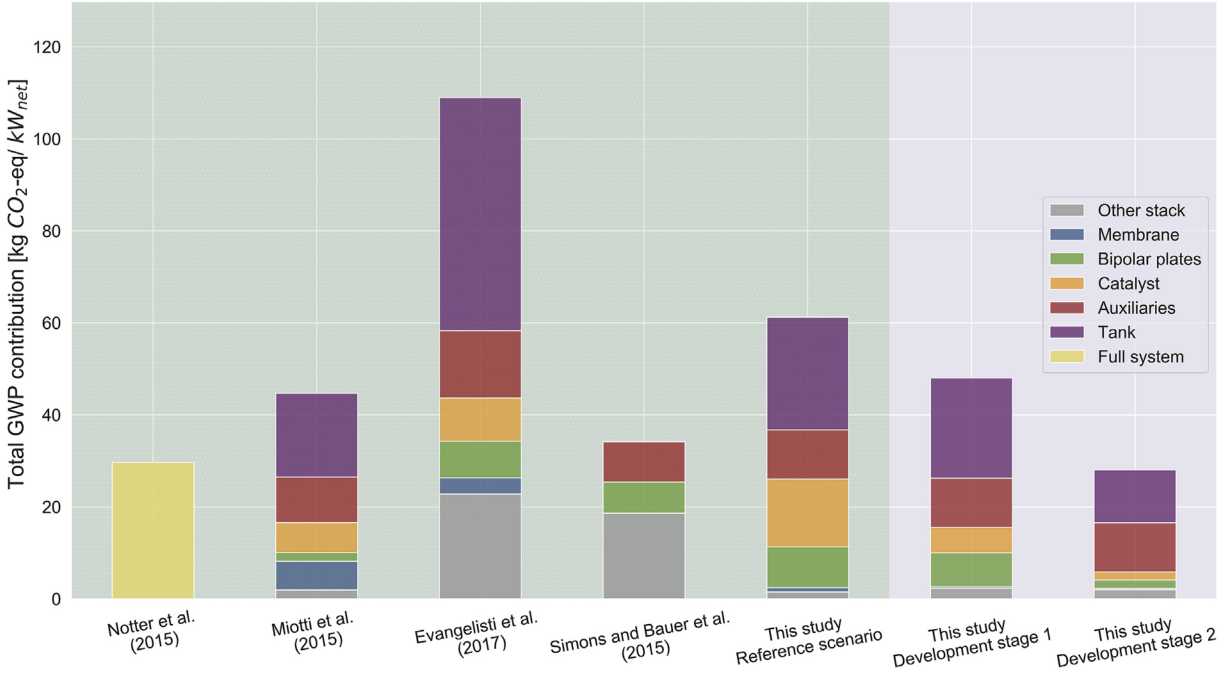

Fig. 4. Comparison of the normalized cradle-to-gate GWP impacts, in kg CO_2-eq/kW_{net}, of our study with preceding studies. The results from the other studies were aggregated, where possible, to have the same component classifications. The columns in the green area depict the impacts of current PEMFCs, while the two columns outside the green area show our assessment of the future impacts. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2015), and reduction of their impacts can be foreseen due to the DOE targets (James and Spisak 2013). The same cannot be stated about graphite-based BPPs, where lower volume and light weighting are hardly achievable (Taherian 2014). Currently, the titanium BPPs equipped in the Toyota Mirai are a significant contributor to the GWP impact category (14% of the total), but their forecasted substitution for a less carbon intensive material such as stainless steel (James 2018) can reduce the footprint of the component.

The variations described above and shown in Fig. 4 highlight the uncertainty in all of the studies. The lack of resolution on the primary data leads to assumptions that may differ from the current manufacturing state-of-the-art. Nevertheless, all the studies agree on the components contributing the most to the GWP impact category. This is, the production of carbon fiber for the storage system and the mining activities associated with platinum.

3.4. Sensitivity analysis

The analysis of the reference scenario highlighted that the FC auxiliaries are among the top contributors to the environmental impacts of a FC system for EVs. However, data and literature describing the state-of-the-art management systems are scarce. The most recent data, to our knowledge, was published by James et al. (Brian D. James et al., 2016), which presented a cost analysis of FC systems and uses the work performed by Carlson et al. (2005) to represent the material composition of some components.

Due to the high uncertainty of the available data, we concentrate on these components in the sensitivity analysis. In the sensitivity analysis, we analyzed how the total impacts would be affected by a 10% increase in the material demand for each component of the FC auxiliaries. Additionally, we performed the same analysis to other relevant components, e. g the tanks, the catalyst and the bipolar plates. Due to the linear relationship between inputs and impacts, these conclusions serve as an indication as to how robust the impact results are to uncertainty of the material inputs to the components. In the analysis of the auxiliary components, we found that increasing each component's mass would yield an increase in GWP ranging from 0.1% for the air management system, to a maximum of 0.5% for the water management system. When the mass of all of the components were increased by 10%, total GWP impacts increased by less than 2%. Therefore, a 10−20% weight variation of the auxiliary components would result in negligible differences in the absolute impacts.

In contrast, the sensitivity analysis of the H_2 tanks, the catalyst and the bipolar plates shows an increase of the total GWP impacts by 4%, 2% and 1%, respectively. Hence, the catalyst proves to the component yielding the most significant variations to the total impacts as a results of perturbations to total material requirements. Results for the other impact categories assessed in this study were similar to results for GWP. Results from the sensitivity analysis can be found in the Supplementary Information.

3.5. Assessment of the prospective technological developments

In the future, FCEVs can achieve significant improvements as the technology matures, fulfilling the ambitious technical performance goals set by the US DOE. Among the targets, the reduction of the Pt loading onto the electrodes with a simultaneous increase of the specific power density (mW/cm^2), and the application of lighter BPPs and thinner membranes seem the main goals (Argonne National Laboratory 2017; James 2018). In our two forecasting scenarios, we aimed to assess the likely future impacts of PEMFCs for EVs in line with these potential developments.

In total, a reduction of the GWP impacts by 21% and 54% can be achieved (Fig. 5), leading emission reductions of 1.0 and 2.6 tons CO_2-eq (Fig. 4), from the reference scenario. Between the reference scenario and the development stage 1 scenario, the largest impact reduction is achieved through the reduction of the platinum loading onto the electrodes; with a total potential impact reduction

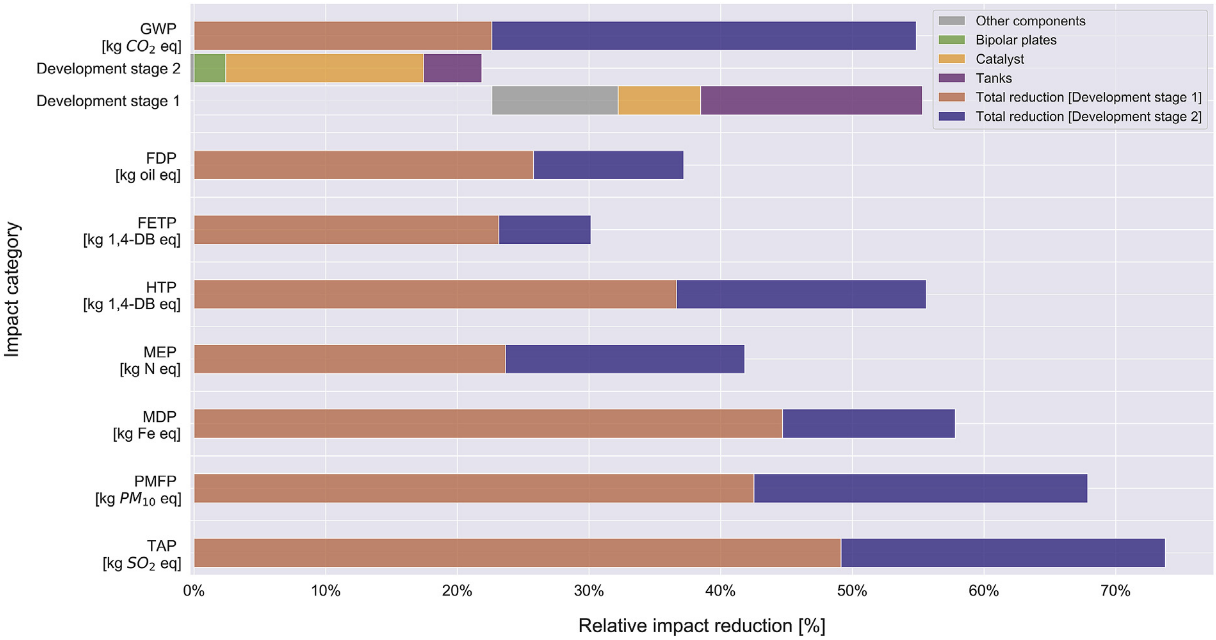

Fig. 5. Potential reduction of the impacts across all the impact categories investigated from the reference scenario. The light orange bars show the impact reduction achieved in the form the reference scenario to the development stage 1 scenario. The blue bars illustrate the total reduction from the reference scenario to development stage 2 scenario. The two bars belor the GWP impact category show a decomposition analysis of the main drivers of reduction. Here, with ‘other components’ we mean all the components not explicitly mentioned in the bars shown in the figure. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

of 15%. Other significant reductions are brought by the replacement of the current carbon-coated titanium plates by gold-coated steel bipolar plates, and the lower energy requirements assumed for the production of the tanks. These two improvements, on the BPPs and the tanks, together would avoid 0.35 ton CO_2-eq (7%), in the development stage 1 scenario, and 1.6 ton CO_2-eq in the development stage 2 scenario. Thus, showing that as the technology develops, significant reduction in the footprint can be achieved.

The Development Stage 1 considers only the technical developments foreseen in the near-term, coupled with 15% lower energy requirements for the production of carbon fiber. Besides these developments, the future production of the components may occur in combination with the use of energy produced by renewable sources. Since a large portion of the parameters used in the development stage 2 scenario are based upon the US DOE scenario for 2025, thus looking at a future condition, we coupled the material requirements with the Sustainable Development Scenario for the European electricity generation in 2025 (IEA 2018), to simulate a hypothetical future cleaner energy mix. We used the future mix for the direct energy inputs associated to the production of the carbon fiber used for the GDL and the tanks. Fig. 5 shows the further impacts reduction achievable with the combination of technical improvements and a likely future European energy mix. Precisely, from the reference scenario to the development stage 2 scenario, the absolute GWP impacts stemming from the tanks would be halved, while the catalyst and the bipolar plates would be seven to ten times less carbon-intensive than their actual environmental performance, for the BPPs and the catalyst respectively. However, such a significant improvement requires a steep learning curve for the technology, and the deployment of renewable energy sources aligned with the 2 °C target goal, coupled with the use of secondary Pt for the catalyst formulation. Thus, the future potential impact reductions might fall somewhere in the greyscale area in Figure 6, setting the maximum value to the current reference scenario and its minimum to the development stage 2 scenario.

Fig. 5 shows the potential impacts reduction for the development scenarios compared to the reference scenario, across all the impact categories assessed in this study. n scenario 1, the first drop in the impacts is mainly caused by perturbations such as the reduction of the Pt loading onto the catalyst from the current 0.32 mg/cm^2 to 0.125 mg/cm^2 (-60%), a higher share of secondary Pt, the assumed reduction in the energy demand for the carbon fiber production, and to a smaller extent by the substitution of the titanium bipolar plates with stainless steel. Although these changes contribute significantly to the reduction in the GWP, other impact categories also benefit from these variations, namely HTP, FDP, MDP, PMFP and TAP. The avoided impacts for the mining of Pt, with all its upstream activities included, are the main cause of the significant impact reduction observable in Fig. 5. On the other hand, the substitution of Ti on the BPPs is mainly affecting the FDP, with an almost negligible contribution to the reduction of the impact across other impact categories.

The further technological development of fuel cell systems, modelled in the development stage 2 scenario, is bringing more reductions to the environmental impacts, with a wide variety of reduction across different impact categories The reduced energy requirements and a higher share of renewable energy sources, compared to the reference scenario, were found as the most effective measure for reducing the manufacturing impacts of the technology.

The technical goals set for the technology, yet ambitious and with potential for impact reductions, have the highest potential from the reference scenario to the development scenario 1. Here, a major progress is foreseen regarding the catalyst, for which mining of primary Pt proves to be the greatest driver of impacts. Nevertheless, from this point on, to further reduce consistently the environmental impacts of FC systems, technical developments must be coupled with cleaner production schemes.

In this study, we focused on the use of secondary platinum, decreased energy requirements for the production of carbon fiber, and cleaner energy sources for the production of the latter. Moreover, this study investigated the technical developments foreseen in the short term, while in the medium-long term the technology might haver significant variations from the inventory compiled for this study. For instance, the catalyst might either employ Pt-free alloys or various other formulations (Ellingsen et al., 2016a,b). The same applies to components such as the fuel tanks, the auxiliaries and the bipolar plates. Thus, while we aimed to highlight the potential impact trajectory of the technology, a constant update of the data used in LCAs is necessary. From an environmental perspective, the key developments appear to be the reduction of the Pt loading onto the catalyst and the use of coated-stainless steel BPPs. However, the reductions observed by the consideration of cleaner energy sources and secondary Pt suggest that a cleaner production is required, in addition to technical improvements. With these elements together, FC systems can reduce their current production impacts up to 50% across several impact categories. The hydrogen tanks, although highly relevant for the current environmental impacts of the technology, are technically constrained by the storage and operating conditions of current FCEVs, namely the driving range of around 500 km that requires 5 kg of H_2. To achieve these conditions, the only existing solution is the application of a storage pressure of 700 bar, which leads to the application of carbon fiber, due to its mechanical strength (James et al., 2016). However, future FCEVs might equip recycled carbon fiber which, as shown in previous studies, generates lower production impacts (Meng et al., 2017). Nevertheless, currently, recycled carbon fiber does not offer the desired strength required for storing hydrogen at high pressure, and therefore its future use in this application is uncertain.

4. Conclusion

Our study aimed to update the representing the state-of-the-art LCA inventories for the FC technology applied to electric vehicles. The results stemming from the life cycle impact assessment were compared to previous relevant studies with high resolution inventories. We found that the technology is not mature enough yet, with disagreements regarding the components' design and thus, life cycle impacts across different studies. Due to the low market readiness level of PEMFCs and the low number of vehicles produced equipping this technology, capturing the impacts is challenging and based on fragmented data. Despite these uncertainties, we find that the production of the fuel tanks and the cathode catalyst are critical components for the emissions generated in PEMFC manufacture, which is in agreement with the previous studies. However, our study, like previous ones in the literature, base some of the assumptions on data that is outdated, such in the case of the auxiliaries. These components play a key role in both the performance and the environmental impacts of the technology, and more robust and recent data is required to identify possible improvement pathways. Furthermore, the developments assessed in this paper are based on both technical targets and on assumptions regarding the employment of secondary materials, technological improvements and use of cleaner energy sources. However, future PEMFCs for FCEVs may differ significantly, which will require novel inventories and analyses.

There is, however, significant potential to improve manufacturing emissions, since as the technology develops, it will see lower material intensity and cleaner production pathways. Potential solutions to mitigate the high production impacts might come from the use of secondary platinum from spent catalytic converters and recycled carbon fiber for the storage tanks.

The current higher impacts and production price in combination with a sparsely populated fueling network, compared to BEV, put the FC technology on a disadvantageous starting position. Nevertheless, high production volumes could significantly contribute to an improved condition, enabling the technology to find its niche in the market, due to several advantages over BEVs. The LCA framework can systematically contribute to the understanding of the current environmental performance of the technology and, if coupled with accurate data, can precisely pinpoint the main areas of concern. At the moment, no LCA studies base their inventories on primary data, which raises several challenges in modelling the production pathway of components such as the catalysts, which involve various steps and several chemicals. Currently, the production of FC systems is currently expensive and environmentally intensive, although with a clean H_2 production, the overall life cycle impacts may be lower than current internal combustion engine vehicles. Thus, it is expected that in the future the technology will play a key role in the decarbonization of the transport sector, if an environmentally sound approach is kept throughout its entire life cycle phases.

CRediT authorship contribution statement

Lorenzo Usai: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing - original draft. **Christine Roxanne Hung:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing - review & editing. **Felipe Vásquez:** Formal analysis, Investigation, Writing - review & editing. **Max Windsheimer:** Formal analysis, Investigation, Writing - review & editing. **Odne Stokke Burheim:** Writing - review & editing. **Anders Hammer Strømman:** Conceptualization, Methodology, Software, Writing - review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2020.125086.

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Glossary

BEV: battery electric vehicle
BOM: bill of materials
BPP: bipolar plates
Co: Cobalt
EV: electric vehicles
FC: fuel cell
FCEV: Fuel cell electric vehicle
FDP: Fossil depletion potential
FETP: Freshwater ecotoxicity potential
GDL: Gas diffusion layer
GHG: Greenhouse gas
GWP: global warming potential
HSC: high surface area carbon
HTP: human toxicity potential
LDV: light duty vehicles
MDP: Metal depletion potential
MEA: membrane electrode assembly
MEP: Marine eutrophication potential
MPL: microporous layer
PAN: polyacrylonitrile
PEMFC: polymer electrolyte membrane fuel cell
PMFP: Particulate matter formation potential
Pt: Platinum
PTFE: Polytetrafluoroethylene
TAP: Terrestrial acidification potential
TFE: Tetrafluoroethylene
US DOE: United States Department of Energy