Manuscript Details

Manuscript number	COELEC_2019_14
Title	Current Status of Automotive Fuel Cells for Sustainable Transport
Short title	Automotive Fuel Cells for Sustainable Transport
Article type	Review article

Abstract

Automotive Proton Exchange Membrane Fuel Cells (PEMFCs) have finally reached a state of technological readiness where several major automotive companies are commercially leasing and selling Fuel Cell Electric Vehicles (FCEVs), including Toyota, Honda and Hyundai. These FCEVs claim vehicle speed and acceleration, refueling time, driving range and durability that rivals conventional Internal Combustion Engines (ICEs), and in most cases outperforms Battery Electric Vehicles (BEVs). The residual challenges and great improvements for PEMFCs that need to be resolved over the next decade are performance at high current density, durability and cost. These are expected to be resolved over the coming decade during which time a hydrogen infrastructure needs to become widely available. Here, we briefly discuss the status of automotive PEMFCs, misconceptions about the barriers that platinum usage creates, and the remaining hurdles for the technology to become broadly accepted and implemented.

Keywords	Fuel Cell Vehicles; PEM Fuel Cells; Platinum; Non-Precious Metal Catalysts; Hydrogen
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Conflict of interest document.docx [Conflict of Interest]

Current Status of Automotive Fuel Cells for Sustainable Transport

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Automotive Proton Exchange Membrane Fuel Cells (PEMFCs) have finally reached a state of technological readiness where several major automotive companies are commercially leasing and selling Fuel Cell Electric Vehicles (FCEVs), including Toyota, Honda and Hyundai. These FCEVs claim vehicle speed and acceleration, refueling time, driving range and durability that rivals conventional Internal Combustion Engines (ICEs), and in most cases outperforms Battery Electric Vehicles (BEVs). The residual challenges and great improvements for PEMFCs that need to be resolved over the next decade are performance at high current density, durability and cost. These are expected to be resolved over the coming decade during which time a hydrogen infrastructure needs to become widely available. Here, we briefly discuss the status of automotive PEMFCs, misconceptions about the barriers that platinum usage creates, and the remaining hurdles for the technology to become broadly accepted and implemented.

Keywords: Fuel Cell Vehicles; PEM Fuel Cells; Platinum; Non-Precious Metal Catalysts; Hydrogen

Introduction

Weaning the transport sector off hydrocarbons has been a major challenge for the automotive industry since the 1970s oil crash, intensifying in the last decade with pressure to decarbonize [1,2]. Around 1/5th of global CO_2 emissions originate from ICEs burning fossil fuels [3], and air pollution caused by particulates, NO_x, SO_2 and CO cause 9million premature deaths per year worldwide, more than are attributed to tobacco smoking [4]. These problems are only set to worsen, as the global passenger vehicle fleet is expected to grow from 1 to 2.5billion by 2050 [1,2] as global population rises to 10billion [5].

After two decades of intensive research, costing billions of dollars, the commercialization of fuel cell vehicles has commenced with several automakers launching FCEVs in the USA, Asia and Europe (Table1). While BEV sales exceed 1million annually [6,7], FCEVs are trickling, rather than flooding into the hands of consumers, in part because the hydrogen infrastructure is a decade behind BEV recharging posts [8].

FCEVs offer many advantages over BEVs (Table1): very fast refueling time (ca. 3-5minutes), freedom from “range anxiety” with up to 600km between refueling, greater longevity (>200,000km), better driver experience and safety [9]. However, FCEVs still have higher capital and operating costs when compared to BEVs, with current models around 250% more expensive [9]. High FCEV cost is primarily due to the use of platinum (Pt) catalysts and current low production volumes. Although precious metal loadings have fallen dramatically in the last decade [10], it still remains a significant issue (Fig.1). For example, Daimler has cut Pt content in its FCEVs (Mercedes GLC F-Cell vs. B-Class F-Cell) by 90% since 2009 and Toyota is targeting a 50% reduction from current levels. However, it is anticipated that (ultra)-low loading Pt or

Non-Precious Metal Catalysts (NPMC), together with increased mass-production of FCEVs, could achieve cost parity with BEVs by 2030 [9].

Misconceptions about Platinum

The automotive industry is a major user of Pt, with catalytic converters requiring around 40% of annual global production. Since the 1990's, R&D has sought to replace Pt in FCEVs with cheaper, durable, highly performing and easily accessible catalysts. The reasons are threefold: (i) high mining and refining costs mean Pt accounts for around a 1/3rd of the total cost of an automotive fuel cell stack*, (ii) the mineral is "scarce" and (iii) Pt mining is concentrated in geopolitically and economically unstable regions [11]. At the time of writing, raw Pt trades at around US$30 per gram [12], which is around 50,000times more expensive than stainless steel. *Note there are no economies of scale, so the cost of raw PGM stays constant independent of the quantity used.

Pt availability is thought of as a problem; however, global reserves are estimated at ~69,000tonnes [13]. According to Cawthorn [14], the Bushveld Complex in South Africa alone could supply global Pt demand for up to a century, with a current annual production of 140tonnes and the possibility of extracting Pt up to 10,000tonnes (350million oz) per km of vertical depth. 2.5billion FCEVs each containing 30g of Pt (i.e. no technical progress from today) would require 75,000tons of Pt. This excludes the potential of recycled Pt from spent automotive catalysts, with up to 95% recovery using present-day technologies [15]. It is therefore very unlikely that Pt availability will prove a major bottleneck for the automotive sector, especially given the likelihood that the present target of $<0.1 g_{Pt}\ kW^{-1}$ will be achieved by 2050. This would place FCEV stacks *on par* with current Pt loadings for catalytic converters (ca. 1g for petrol, 8-10g for diesel).

Status of FCEVs

Unlike the extensive global rollout of BEVs, FCEVs are leased and sold in small quantities and in limited areas. A decade since Honda publicly launched the FCX Clarity, the fuel cell variant is still only available in Japan and California. The Toyota Mirai and Hyundai ix35 are available more widely, although only 15 countries have public stations to refuel them at present [9]. 5,600 FCEVs now operate in the USA, with a comparable number in the rest of the world combined [16].

Worldwide, governments are preparing for a major push towards FCEVs. By 2030, the USA targets 1million FCEVs in California alone [17], while China, Japan and South Korea aim for 1, 0.8 and 0.6million respectively [9]. Japan aims to deploy 200,000 FCEVs by 2025, costing ~US$6,000 more than a standard hybrid, versus US$27,000 premium today, with a cost target for the fuel cell stack falling from U$200 to US$50 per kW.

Increasingly it is argued that hydrogen's main role may lie beyond passenger vehicles, decarbonizing heavy transportation sectors that batteries cannot easily serve [1,2]. The suitability of PEMFC for buses, trucks, trains [9], shipping [19] and even aviation [20] is of significant interest worldwide.

There is broad agreement that hydrogen-powered transport is an essential component of climate change mitigation, especially if global warming is to be limited to 1.5°C (Fig.2).

Decarbonization scenarios see hydrogen supplying a tenth of global transportation energy demand as early as the mid-2040s [21]. This requires a sustained period of scale-up: the median pathway to achieving 1.5°C sees hydrogen usage growing by >20% year-on-year for the next three decades. Reduced ambition to mitigate climate change still presents a major role for hydrogen, albeit delayed by several decades [21].

Status of fuel cell systems

At this time, durability and cost are the primary challenges still to be met. The 2020 US DoE targets for fuel cell systems are a cost of US$40/kW with an efficiency of 65% at peak power and with 12.5g of Pt, based on 500,000 automotive fuel cell systems produced per year (Figs.1&3). The PEMFC stack cost breakdown identifies that the catalyst contributes significantly to the total cost (41%) when compared to the bipolar plate, membrane, gas diffusion layer (GDL), electrodes and gaskets, and balance of plant (BoP) costs [22] (Fig.3). Manufacturing scale-up has reduced the costs of PEMFCs at a comparable pace to lithium ion batteries [23]. According to Moreno *et al.* [24] reducing the membrane electrode assembly (MEA) cost up to 30% makes the US$40/kW cost target by 2020 reachable, corresponding to a reduction in catalyst cost to <US$4/kW and the membrane to <US$1/kW. However, the catalyst cost is predominantly a material cost and does not fall with the number of systems produced per year. Thus, lowering the amount of Pt-based catalyst used while maintaining the durability is essential. The catalyst loading can be lowered by finding an improved catalyst with a higher oxygen reduction reaction (ORR) activity, increasing the catalyst surface area and lowering the mass transport losses at high current densities.

Cathode ORR Catalysts

Tremendous progress has been made over the last two decades in improving the anode and cathode catalyst layers that form the heart of the fuel cell, the two important layers that are sandwiched with the polymeric proton exchange membrane in-between to form MEAs. However, continued research on alternative catalysts for the slow ORR on the cathode has been a prime focus, with the goal of lowering the total Pt content of the stack from 30g to less than 10g.

Current attention on the MEA is mainly focused on improving the cathode catalyst layer. The ORR reaction is sluggish, and Pt/C loadings of ~$0.35mg_{Pt}/cm^2$ are typically needed. Improving ORR catalytic activity (e.g. alloy catalysts, novel structures) is being intensively studied as it would lower the total loading of Pt in the fuel cell stack and hence lower the cost of the stack [24]. Electrocatalyst activity may be enhanced through both improved surface area accessible to reactants (better mass activity mA/mg_{Pt}) and higher intrinsic activity (mA/cm_{Pt}^2). Additionally, the cathode catalyst layer is subject to cyclic load cycling between 0.60V and OCV, variable RH (Relative Humidity), water generation as a function of current density and start-up/shut-down losses. Each of these conditions degrades the performance of the catalyst layer over time.

Numerous binary and ternary Pt-based electrocatalysts were discovered while conducting research on PAFCs (Phosphoric Acid Fuel Cells) in the late 20th century, such as Pt-Co, Pt-Ni, Pt-Cr and Pt-Co-Cr, Pt-Rh-Fe. These have shown durability of over 40,000 hours in commercial stationary fuel cells at 190°C [25], and this learning has been transferred to PEMFCs, with Pt-Co/C and Pt-Ni/C being the most commonly utilized binary alloys. Additional work had to be conducted to modify these catalysts for use in PEMFCs, in part due to the different electrode structure in PEMFCs as compared to acid filled GDEs (Gas Diffusion Electrodes) of PAFCs. PTFE has been eliminated and the catalyst layer consists of solely catalyst/C and ionomer. Fundamentally, base metals tend to move to an alloy's surface and cannot be stopped due to surface segregation. The limited amount of ionomer (30%) in PEMFC cathodes means base metal leaching into the ionomer is a much more serious issue than in liquid acid-based fuel cells. For PEMFCs, Pt-alloys are typically pre-leached to remove excess Co from the surface and minimize the increase in catalyst layer resistance (protonic resistance) over time. In addition, smaller Pt particles can be used in PEMFC cathodes since the operating temperature does not exceed 90^{o}C.

MEAs have been widely reported with exotic catalyst structures that exhibit >10x the activity of nanoparticle Pt/C [26,27,28]. These novel classes of electrocatalysts tackle the problem that most atoms in a Pt nanoparticle remain unused since only the surface participates in reactions. Particles below ca. 2nm are unstable and will double in size after 1,000hours of operation due to dissolution and re-deposition.

Cathode Electrode Structure

As the automotive industry employs ever-higher activity Pt-alloy catalysts, Pt loadings are reduced correspondingly, in turn yielding thinner and mechanically weak catalyst layers and MEAs. For example, Pt-Co/C with 4x the activity of Pt/C requires a catalyst layer with one quarter the thickness, impacting on stability and durability. This problem is partly mitigated by reducing the wt% of Pt. The H_2-air performance curves do not shift uniformly over the range of current densities, and at high current densities the limiting current has an early onset.

Numerous studies have been conducted to understand this so-called *anomalous effect* [27]. Looking at specific current density, catalyst sites may become more severely stressed when loadings are reduced. The losses from this phenomenon have been attributed to the ionomer/catalyst interface and poisoning of the catalyst by the sulfuric acid groups on the ionomer.

Research into PGM-free ORR catalysts

Since the 2000s, some 65,000 papers have been published in the area of NMPC [29]. It is questionable whether PGM-free catalysts have a role to play as catalyst loadings approach the target of 10g Pt per 100kW stack. However, there has been renewed interest in PGM-free cathode catalysts, and their activity has been improved significantly albeit only under oxygen [30]. Since the intrinsic activity of PGM-free catalysts for ORR is low, more catalyst has to be applied to provide similar performance, especially under air. Higher loadings of PGM-free catalysts result in a much thicker catalyst layer (up to 100times than those containing PGM) concomitant with higher catalyst layer resistance and mass transport losses. Finally, most PGM-free catalysts still suffer from poor durability [30].

Closing remarks

The overall goal for automotive PEMFCs is to match the performance, cost and durability of conventional ICEs. The performance and durability of all major stack components including bipolar plates, membranes, catalysts, gas diffusion layers as well as balance of stack have been significantly improved over the last decade. It is now very likely that cost and durability targets will be met in the next decade, providing a commercially viable alternative to the internal combustion engine which has dominated for the last century.

References and recommended reading

Papers of particular interest, published within the period of review, have been highlighted as: •Paper of special interest ••Paper of outstanding interest.

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List of Figures and Tables

Figure 1: Evolution of total platinum group metal (PGM) cost and loading for a 100kW FCEV, showing historic development, current status and targets. Bars show the total PGM cost based on current raw Pt prices (≈US$30/g). Values inside bars give the total fuel cell stack cost for a manufacturing volume of 500,000 per year. Values outside bars give the total PGM loading per unit cell area (mg/cm²) and for a 100kW FCEV (g). Inset figure shows a broader set of technical targets for FCEVs in 2020.

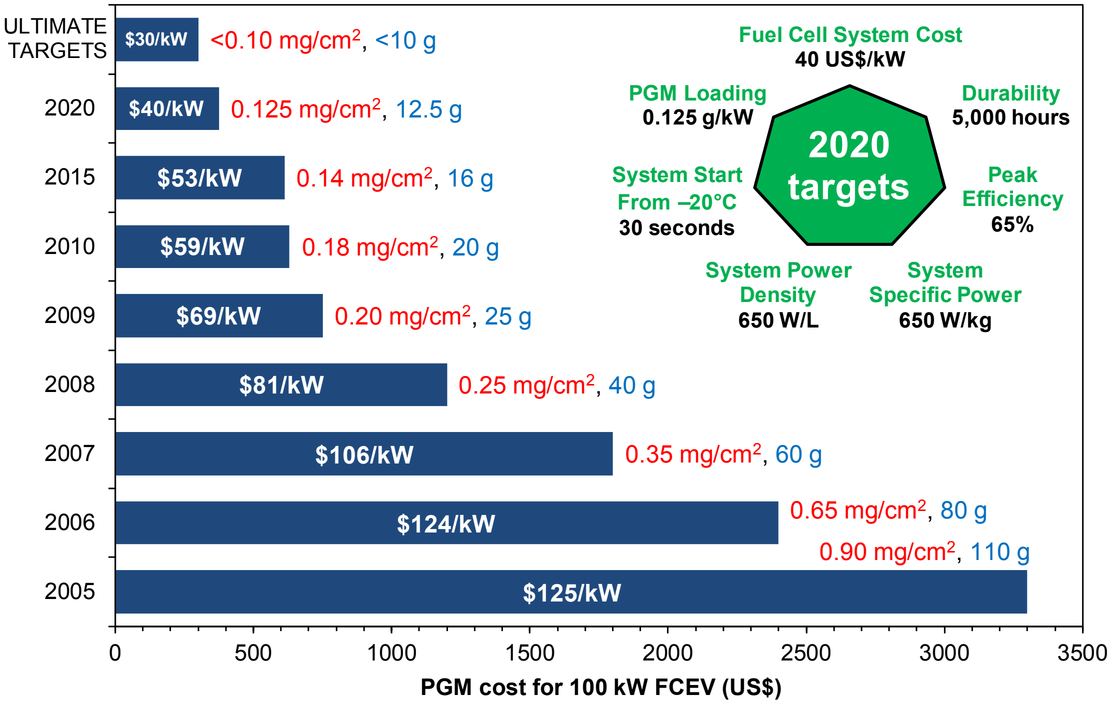

Figure 2: The share of global transportation provided by hydrogen during the 21st century, as modelled in scenarios for the IPCC Special Report on Global Warming of 1.5°C [21]. The hydrogen share is measured in terms of primary energy input across all modes of transport. Lighter shaded areas show the central two-thirds of scenarios (17th to 83rd percentile), darker shaded areas show the central one-third (33rd to 67th percentile). Colors classify scenarios by their warming impact in 2100.

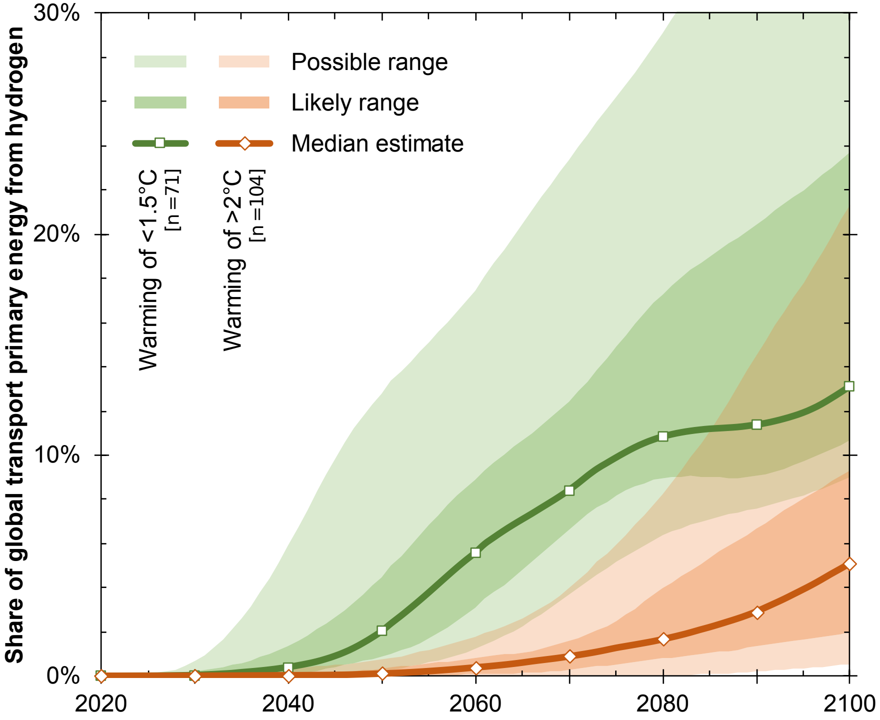

Figure 3: Automotive fuel cell cost evolution and projection. Bars show the PEMFC stack cost (US$), while the cost of the total FCEV is printed for selected years. Inset figure: Automotive fuel cell component cost distribution based on 500,000 fuel cell systems produced per year.

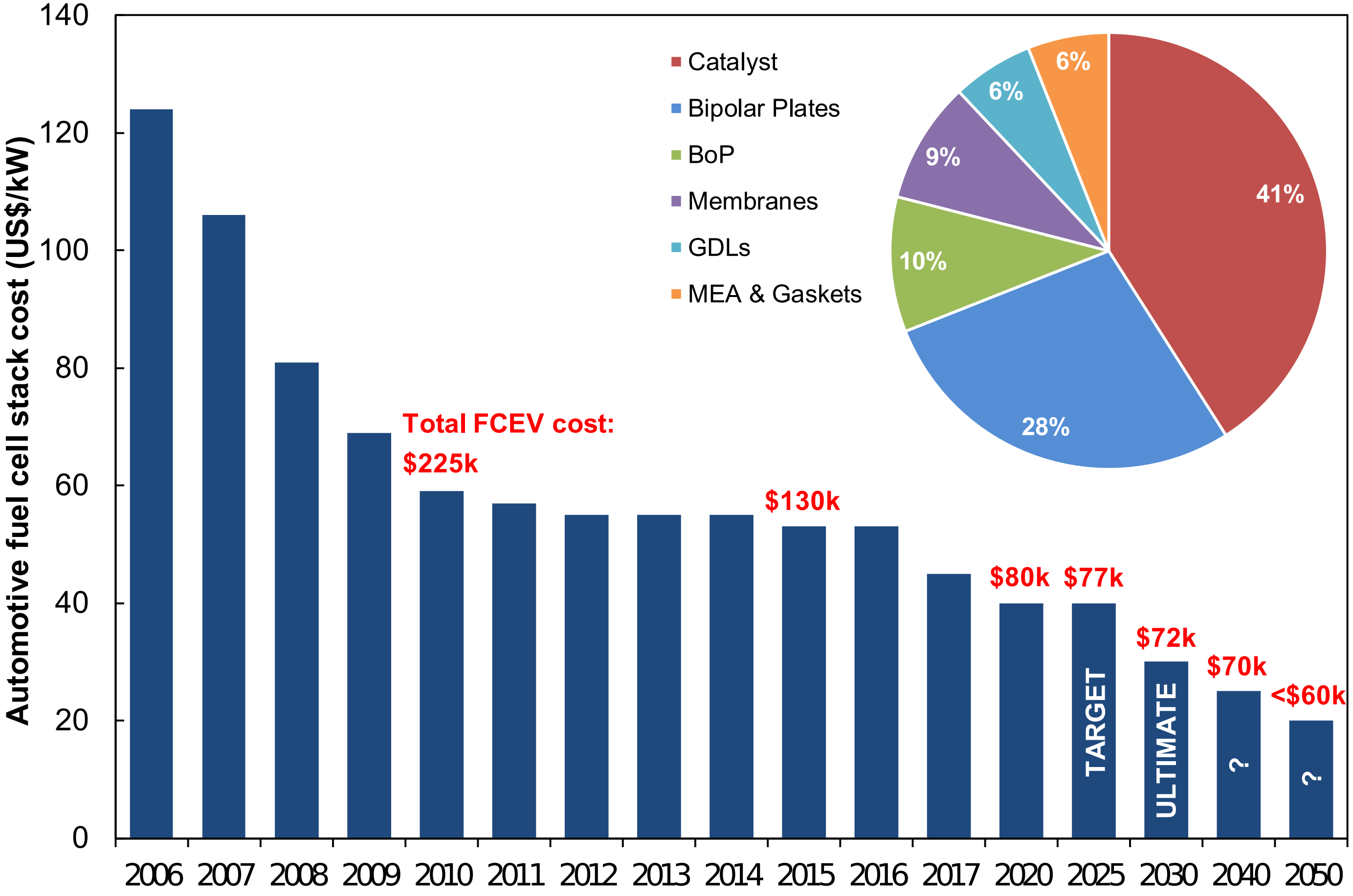

Table 1: Specifications for the latest FCEV models currently in production. For other FCEVs, see [1,2].

FCEV	Launch date	Mass (kg)	Fuel cell/ motor power (kW)	Power density (kW/L)	Acceleration time (s) 0-60 mph [100 km/h]	Fuel tank capacity (kg) [wt%]	Fuel pressure (MPa)	Estimated range (miles - km)	Fuel economy Kg hydrogen / 100 km	Fuel consumption (mpg gasoline equivalent)*
Hyundai Nexo	2018	1873	95/120	3.10	9.5 [10]	6.33 [7.18wt%]	70	370 - 595	0.84	57-61
Honda Clarity	2016	1875	103/130	3.12	9.2 [9.7]	5.46 [6.23wt%]	70	366 - 589	0.97	67
Hyundai ix35 FCEV or Tucson FCEV	2014	1980	100/100	1.65	12.5 [13.2]	5.64 [6.43wt%]	70	369 - 594	0.95-1.0	66
Toyota Mirai	2014	1850	90/114	3.10	9 [9.5]	5.0 [5.70wt%]	70	312 - 502	0.76	67

*compared to sales-weighted average fuel economy in the USA of 25-43 mpg, depending on vehicle class.

Conflict of interest

We wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.