Title
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Journal
Nature Energy, 6(5)

ISSN
2058-7546

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Publication Date
2021-05-01

DOI
10.1038/s41560-021-00775-z

Peer reviewed

New roads and challenges for fuel cells in heavy-duty transportation

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Abstract:

The recent release of hydrogen economy roadmaps for several major countries emphasizes the need for accelerated world-wide investment in research and development activities for hydrogen production, storage, infrastructure, and utilization in transportation, industry, and the electrical grid. Due to the high gravimetric energy density of hydrogen, the focus of technologies that utilize this fuel has recently shifted from light-duty automotive to heavy-duty vehicle (HDV) applications. Decades of development of cost-effective and durable polymer electrolyte membrane fuel cells must now be leveraged to meet the increased efficiency and durability requirements of the HDV market. This review summarizes the latest market outlooks and targets for truck, bus, locomotive, and marine applications. Required changes to the fuel cell system and operating conditions for meeting Class 8 long-haul truck targets are presented. The necessary improvements in fuel cell materials and integration are also discussed against the benchmark of current passenger fuel cell electric vehicles.

Interest in hydrogen as an energy carrier has been reinvigorated in recent years, driven by the need for more flexible utilization of renewable energy sources and increased security of the global energy market.[1] By 2050, hydrogen could meet 14% of the energy demand in US[2] and 24% of world's energy needs,[3] of which 30% is projected for use in transportation.[4] The hydrogen roadmap for Europe anticipates a potential to generate 25% of European Union energy demand from hydrogen by 2050 to mobilize a European fleet of over 50M fuel-cell vehicles.[4] Significant action has come from local and national governments, which are increasingly enacting legislation banning internal combustion engines (ICEs) in favor of electric vehicles (EVs). China, in particular, has drastically expanded its support of fuel cells, with a reported 2018 funding/subsidy level of ¥85 billion ($12.4 billion USD),[5] and Germany recently announced €9 billion ($10.2 billion USD) in funding for hydrogen and fuel cells.[6] The United States Department of Energy (US DOE) has supported fuel cell transportation R&D since the late 1970's, and their research and development targets to enable commercialization, including those of the Hydrogen at Scale (H2@Scale) initiative, have been widely cited.[7,8]

Polymer electrolyte membrane fuel cells (PEMFCs) have many benefits over conventional ICEs used in passenger light-duty vehicles (LDVs), including higher efficiencies (>60%), while providing similar driving range (> 300 miles) and fueling times (< 5 min). Fuel-cell electric vehicles (FCEVs) are in the early stages of commercialization, with approximately 7,000 sold or leased in the United States to date.[9] Nearly every major automotive manufacturer is developing FCEVs. However, the initial cost of these vehicles is not yet competitive, and current approaches to stack cost reduction can dramatically decrease the stack lifetime. Lack of hydrogen refueling stations and cost parity with gasoline are also significant constraints for widespread FCEV commercialization.

While fuel cells for LDVs have been in development for over two decades, heavy-duty applications have only recently attracted significant attention.[10] This shift has been catalyzed by the unique scalability of fuel cells in terms of both power and energy, which can be achieved by increasing the size of the fuel cell stack or hydrogen tank at a much smaller additional weight penalty than Li-ion batteries. Commercial deployment of HDVs also requires less infrastructure investment as fewer refueling stations are required due to dedicated and more predictable routes. However, the different drive cycles and operating conditions of heavy-duty vehicles (HDVs), as well as their significantly longer lifetimes, mean a substantial improvement in durability and a greater focus on fuel efficiency is required as compared to LDVs. For instance, over 75% of LDV miles are used for non-interstate travel, while 55% of truck travel is spent on interstate roads (bts.gov). Further, the average Class 8 trucks travels 6 times further than the average LDV.[11] The US in particular has a large industry relying on HDVs that exceeded 300 billion annual vehicle miles traveled (AMT) in 2018, of which over 180 billion was attributed to long-haul trucking.[2,12] The heavy-duty market is also a critical market for reducing energy consumption and emissions, as medium- and heavy-duty trucks consume 25% of the total annual vehicle fuel use and produce 23% of the total CO_2 emissions in the US today.[13,14] Furthermore, annual freight truck miles traveled is projected to increase by 54% by 2050.[15]

Herein, we review the recent roadmaps, market outlooks, and targets for FCEVs, with a focus on applications in the HDV sector. Taking on the specific case of Class 8 long-haul trucks, simulated drive cycles are presented which inform system level strategies for achieving the high efficiency and durability operation required by this application. An increase in operating cell voltage is presented as the most viable path forward to improved efficiency, with key changes in

system design proposed to address high power operation where the majority of fuel is consumed. The materials and electrode design breakthroughs required for this new operating paradigm are discussed in detail. A shift in focus to end-of-life performance is emphasized in the development of advanced catalysts and supports, high-temperature membranes and ionomers, novel electrode designs, and durable gas diffusion layers and bipolar plates. Finally, the materials utilized in the Toyota Mirai passenger FCEV are presented as a benchmark for materials development and innovation.

Emerging transportation markets for FCEVs

The substantial decrease in Li-ion battery cost and their introduction into various vehicle markets, as well as increased electrification of the transportation sector, have resulted in a rapid expansion of electrochemical transportation technologies.[16] While current fuel-cell and battery LDV costs are estimated to be close to parity for similar driving,[17] the lack of a hydrogen fueling infrastructure has severely limited the adoption of fuel cells for LDVs. An exception to the implementation of PEMFCs is the material handling sector, where the central refueling nature of these applications coupled with cost and operation advantages have allowed this technology to be implemented without subsidies.[18] Working from that model, PEMFCs are increasingly being examined for use in HDV applications where there is a need for high power and reduced emissions, such as in ports for drayage or delivery vehicles with well-defined routes. Equally as important are HDV applications that can benefit from the separation of energy storage and power output, the inherent strength of fuel cell systems. Factoring in refueling time advantages and the intrinsic power density, PEMFCs have significant performance advantages for both medium-duty (MDV) and HDV applications.[19] Until recently, HDV applications utilizing PEMFCs have primarily been limited to fuel-cell buses which have been employed by various US transit authorities since the early 2000s. The fuel and battery system lifetimes in these applications have exceeded 30,000 hours, with an average of 13,236 hours.[20] Much like their LDV counterparts, buses have seen limited adoption due to both the high initial capital cost (>$1M) and the scarcity of hydrogen fueling stations. However, these barriers are being lowered thanks to a mix of government investment towards production of less expensive hydrogen, expanded policies focused on increased electrification, and continuous technological advancements. This progress is evident in the ever-expanding number of bus and truck fleet demonstration projects, especially in Europe and China.[21]

In a roadmap to a US hydrogen economy developed by the Fuel Cell & Hydrogen Energy Association (FCHEA), an ambitious plan from early scale-up in 2025 to broad roll out in 2030 could result in a 17M-ton demand for hydrogen with over 5600 fueling stations. In such a scenario, the number of FCEVs is expected to dramatically increase to 200,000 in 2025 and 5.3M in 2030 for passenger cars and commercial vehicles.[2] The hydrogen roadmap for Europe includes a 2030 market penetration of 45,000 HDVs and projects, by 2050, 1.7M HDVs (trucks and buses) and 5000 trains.[4] Similarly, Japan projects the use of 1200 buses by 2030 (www.iphe.net/japan) and the Republic of Korea projects 70,000 trucks and buses by 2040 (www.iphe.net/republic-of-korea).

Similar opportunities are emerging for fuel cells for freight, regional, and switcher locomotives due primarily to the tightening standards for NO_x, SO_x, particulates, and noise. Freight locomotives consume a significant amount of fuel at high notch levels where the diesel engine is most efficient, while yard switchers operate at idle and low notch levels where fuel cells are most efficient. Depending on the service route, suburban passenger trains have frequent start-stops and

acceleration events. Preliminary indication is that fuel cells can have a 30-35% efficiency advantage over diesel engines in freights and regionals, but >75% in yard switchers.[21] Durability is a major challenge since the required engine lifetime is 10-15 years, 35,000 operating hours, and ~3.6M miles.[22]

Fuel cells are also being considered for maritime applications primarily due to the International Maritime Organization's ambition to cut sulfur emissions by 86% and CO_2 emissions by 50% by 2050.[5] Preliminary indication is that the efficiency advantage of fuel cells over marine diesel engines may be small for medium and large container ships, although the emission advantages are much greater. Again, durability is a major challenge since the required engine lifetime for container ships is 25 years and 75,000-100,000 operating hours.[23]

These emerging fuel cell applications are summarized in **Figure 1**, which depicts the significance and implication of the shift from LDV to HDV applications both in terms of size and mileage driven. The dramatic increase in the average daily and lifetime mileage of these applications shifts the focus from capital to operating costs (*CapEx* to *OpEx*). At the cell level, this signals a switch from the current LDV focus on low Pt-loaded electrodes and very high power densities to new material and integration solutions that will mitigate degradation and increase efficiency. The following sections explore the case for long-haul HDV trucks, but the same approach, challenges, and solutions are relevant to the other HDV applications.

System strategies for increased efficiency and durability

The recently-released DOE targets for hydrogen Class 8 long-haul trucks (**Figure 2**) emphasize the longer lifetimes and increased efficiency demands of HDV applications, with nearly a four-fold increase in system lifetime to a total of 12 years and 1M miles of operation.[24] For LDVs, PEMFC peak efficiencies have reached approximately 65%, a significant gain over the tank-to-wheels efficiency of gasoline-powered passenger vehicles around 19%. Such efficiency advantages at 25% rated power will be harder to come by for MDV/HDVs, where diesel trucks routinely obtain 40% efficiency at much higher rated powers of 50-100%.

These enhanced targets offer some distinct differences and opportunities for PEMFC design. The specific materials and system requirements are determined by the drive cycle, which can differ significantly even within a given application.[25,26] For example, the drive cycle for a Class 8 truck illustrated in **Figure 3a** is just one of three under consideration by the EPA's Greenhouse Gas Emissions Model (GEM2).[27,28] The PEMFC voltages corresponding to this drive cycle were simulated by the system modeling team at Argonne National Laboratory assuming a minimum idle power of 20kW to limit the peak cathode potential, a strategy implemented to limit cathode catalyst degradation.[29]

Figure 3b also illustrates how the system efficiency at rated power is dramatically affected by the cell voltage and coolant temperature. For example, a stack operated at 0.675V (comparable to current LDV operation at rated power) would need to run above 90°C to reject waste heat and enables efficiencies of 43-46%. Efficiencies ≥ 50% can only be reached by operating above 0.7V. Higher voltages also allow lower operating temperatures that extend the durability of electrodes and membranes. This modeling illustrates one of the difficulties encountered in HDV operation: heat rejection at low speeds on steep gradients where the battery needs to be used to

supplement the PEMFC power. This hybrid strategy has also been used to limit the cell voltage of the stack to 0.9V, as illustrated in the drive cycle in **Figure 3a**.

As noted in Fig. 3b, heat rejection in HDVs is facilitated by higher cell voltage at rated power, which translates to lower power densities and bulkier stacks, and elevated operating temperatures for smaller radiator fans (less parasitic power). Initial total cost of ownership analysis of Class-8 trucks also favors selection of higher cell voltage at rated power for higher efficiency (better fuel economy) and higher permissible Pt loadings (initial capital cost). Since HDVs require extended lifetime (25,000 h), the issue of durability is equally, if not more, critical. Thus, some of the high-payoff material development related areas of research are a) high activity catalysts without the restriction of very low PGM loadings, b) catalysts and ionomers stable at temperatures above 90-100^{o}C, c) highly stable ionomers and membranes under dry conditions, and d) high durability catalysts, catalyst supports, and membranes, which are discussed in detail in the materials strategies section. The strategies for engineered solutions include limiting peak potentials in drive cycles transients, and hybridization with a battery to idle stacks during periods of low power demand. Implementing these strategies requires complete understanding of the relationship between stack and membrane degradation rates and operating conditions (potential, temperature, relative humidity) and the methods of controlling them. Voltage optimization in LDVs focused on decreasing the overall ohmic and mass-transport resistances in a push towards higher current densities. For the HDV space, the increased voltage and temperature requirements for higher efficiencies require a more dedicated focus on reducing kinetic losses, mixed potentials due to crossover hydrogen and oxygen, and localized issues due to catalyst/ionomer interactions in the MEA, as discussed later. The specific mix of materials- and system-mitigation strategies will need to be tailored to the stressors identified in the drive-cycle analysis and specific hybridization strategy.

In terms of durability concerns, cell reversal can occur during unprotected startup-shutdown transients and during fuel starvation resulting in the oxidation of the catalyst-carbon support.[30-32] There has been extensive research to develop more stable supports, but solely materials solutions have thus far been inadequate and engineering approaches of preventing formation of an H_2/air front by depleting oxygen from the cathode during shutdown have proven more successful as discussed in Box 1. Furthermore, voltage clipping is an effective mitigation strategy for limiting carbon corrosion and catalyst particle coalescence during operation, and systems-level mitigation strategies such as limiting upper and/or lower cell voltage (*i.e.*, cathode potential) have also been investigated to prevent catalyst dissolution.[33,34] This strategy appears to have been implemented in the current generation of PEMFC LDVs, but has the drawback of restricting the minimum power that the system is allowed to generate, thus limiting overall system efficiency. Operating at air flow rates only slightly higher than those needed for the required stack power (*i.e.*, high oxygen utilization) and lower oxygen concentrations are some of the available options to expand the operating power window by reducing the allowable idling power.

Another durability issue is ice formation when starting from subfreezing temperatures, which can lead to electrode delamination and other performance losses arising from water-management-related transport resistances. Many patents are available on approaches to prevent ice formation. These include operating at low voltages (*i.e.*, low cathode potentials) during cold start to maximize in-stack heat production to warm the stack above 0°C rapidly, which comes at the expense of overall system efficiency, or purging the stack during shutdown at subfreezing

temperatures to remove liquid water and dehumidify the MEA.[35,36] An additional concern arising from the extended HDV lifetime is the cumulative effect of contaminants introduced into the MEA through the fuel stream, air stream, or from other components in the PEMFC stack or balance of plant. There are numerous studies and reviews on this topic describing the poisoning mechanisms and mitigation strategies.[37] The fuel and air purity requirements for HDV applications are not expected to be any different from LDV applications, with increased Pt loading offsetting the increasing durability requirements of HDVs. However, contaminants such as iron, that could potentially lead to enhanced membrane degradation, are of great concern in HDV applications and should be avoided until more robust membranes are synthesized or mitigation strategies that either prevent the migration of contaminant ions to the membrane or eliminate their degrading effects are developed.

Materials strategies for increased efficiency and durability

Many of the fundamental LDV durability limitations and failure mechanisms will be exacerbated by the longer lifetime and potentially higher operating temperatures of HDV systems. New operating, materials, and materials integration strategies are needed to address these challenges. Possible means to address these challenges while also meeting lifetime and efficiency requirements are hierarchical assemblies, optimized catalyst/support interactions, alternative support or composite materials, chemically-modified membranes, tuned membrane/electrode interfaces, and self-healing or hybrid ionomers. These concepts are discussed by MEA component (**Figure 4**) in the following sections.

Catalyst and supports

The development of durable cathode oxygen reduction reaction (ORR) catalysts that minimize or eliminate the use of Pt is a particularly active research area that has been summarized in recent reviews.[38-41] While immense progress has been made in improving the activity and durability of low and non-Pt ORR electrocatalysts, further improvements and significant R&D effort, especially in the area of durability, are needed for these materials to be considered for the interim target of 25,000 h for HDV applications.[38,41]

Fortunately, the shift in balance between capital and operating costs means the requirement for ultralow loadings of precious metal catalysts (<0.125 mg cm^{-2}) for LDVs are not as critical during the initial transition to HDVs, allowing for loadings around 0.3 mg cm^{-2}. Considering fleet size and the current use of noble metals in Class 8 trucks for catalytic converters and particulate filters; replacing diesel HDVs with FC-HDVs should not pose a significant increase in Pt demand.

However, due to the extended lifetime and potentially higher operating temperatures of HDV stacks, many of the same concerns remain or are exacerbated regarding loss of catalyst electrochemically-active surface area (ECSA),[42-46] and the resulting loss of catalyst performance and stack power density and system efficiency (*i.e.*, increase in voltage loss at a given current density). The loss of ECSA is driven by Pt dissolution with subsequent deposition of the Pt in the membrane[47] and redeposition of Pt onto existing particles (termed Ostwald ripening) and/or particle coalescence.[39,48-52] It is noteworthy that platinum dissolution is especially prevalent during potential cycling in the 0.6 to 0.95 V range, and thus is a concern for the cathode catalyst and not a major concern for the anode catalyst, which operates at close to the reversible

hydrogen potential. Recent work suggests catalyst particle-size distribution plays a more significant role than overall particle size in mitigating ECSA loss via Ostwald ripening. However, monodispersed catalyst particles are difficult to produce and the development of scalable, cost-effective synthesis routes is an area particularly worthy of further research.[53]

In addition to tailoring particle size and dispersion, the ORR activity of Pt can be enhanced through alloying with transition metals (TMs), such as Co and Ni. Despite pre-treatment of Pt-TM alloys to remove leachable TM, additional leaching occurs during MEA assembly and operation causing loss of the enhancing effects of the TM on ORR activity. [42,43,54] In current LDV systems, like that of the Toyota Mirai, the TM content of the cathode catalyst is only ~ 10 at.% and system controls are employed that limit the highest potential experienced by the cathode to prevent TM loss during operation, while still allowing use of a catalyst with a higher ORR activity than that of Pt.[55,56] In addition to performance penalties due to the impact of loss of TM on ORR activity, dissolved TMs can also affect the proton and oxygen transport properties of the ionomer in the cathode catalyst layer, thus further decreasing high-current-density performance.[57] Thus, the longer lifetime requirements for HDV MEAs compared to those of LDVs will present a significant challenge for alloy catalysts. **Figure 5** illustrates this challenge by comparing the *beginning-of-life* (BOL) and *end-of-life* (EOL) performance of MEAs composed of a commercial PtCo alloy or Pt cathode catalyst with similar average particle size and distribution. These two cells were subjected to the accelerated stress test (AST) recommended by the US Department of Energy, which typically comprises 30,000 cathode potential cycles between 0.6 and 0.95 V with 3 s holds at each potential, which has been shown to equate to a projected system lifetime of 5,000 h.[7] Since the system lifetimes for HDV applications are expected to be >25,000 h, the number of AST cycles was extended from 30,000 to 75,000. While the PtCo alloy-containing MEA does show improved BOL performance at low current densities, this advantage is lost over the period of the test with the EOL performance of the two catalysts being almost identical at the lower current densities (**Figure 5**). This durability issue of current commercial alloy catalysts, combined with the inferior performance of alloy catalysts at higher current densities due to mass-transport losses, is a significant issue that must be addressed prior to implementation in HDVs.[43] There are several synthetic routes and structures for Pt-based catalysts that have shown improved retention of TM and thus better ability to maintain ORR activity and performance over time. One of the most promising strategies is the use of Pt-TM intermetallics for which stability is imparted by the ordered arrangement of Pt and TM in the catalyst particles.[38,58,59] Recently, there have been single-cell lab-scale demonstrations showing improved TM and activity retention with ordered Pt-Co, Pt-Ni, and Pt-Fe intermetallics with TM contents as high as 27 at%.

Increased attention is also being given to the carbon support structure, which plays a critical role in catalyst particle size, dispersion, and interaction with the ionomer, as well as catalyst durability. The geometry of the support's pores as well as the location of the catalyst particles with respect to the pores impacts oxygen transport, ORR kinetics, and ECSA loss due to reduced particle coalescence.[60] Carbon supports vary from highly-graphitized, corrosion-resistant carbons with low porosity to the generally-favored higher surface area (HSA), but less stable, mesoporous carbons, which show improved mass activity and accessibility to particles to oxygen.[61] In particular, electrocatalysts with high mass activities (>600 mA/mg_{Pt}) have been

almost exclusively supported on porous carbons, where the location of Pt and Pt alloys within the carbon pores prevents interaction with ionomer that could otherwise impede ORR kinetics.

While system-level strategies, such as depleting oxygen from the cathode during shut-down can significantly mitigate the corrosion of the HSA mesoporous carbon supports, the longer HDV lifetimes will require the development of catalyst and support combinations with low catalyst particle mobility, lower extent of poisoning by ionomer, and inherent resistance of both catalyst and support to corrosion. There has been extensive research to develop supports that are more stable and that interact more strongly with catalyst particles to inhibit mobility. These include oxides of refractory metals, carbides, nitrides, nitrogen-doped carbons, and graphitized carbons.[62,63] To date, alternative materials to carbon have suffered from either insufficient BOL electronic conductivity, loss of conductivity during operation, and/or low surface area and poor Pt particle dispersion. While graphitized carbons have been shown to decrease carbon corrosion, they also have lower surface areas than non-graphitized supports and thus suffer from issues with adequately dispersing the catalyst particles and maintaining the dispersion. Promising approaches for overcoming this conundrum include doping the graphitized supports with elements, such as nitrogen, that strongly interact and anchor the catalyst particles.[64] Other novel approaches include deposition of metal oxide, such as zirconia or silica, or carbon overlayers to protect and stabilize the Pt particles against dissolution and migration and coalescence.[65-67]

A final key consideration is the benchmarking of catalyst activity and durability. The aqueous electrolyte thin-film rotating disk electrode technique (TF-RDE) is an efficient way to screen catalysts and develop structure-property relationships,[68] but the exceptional mass activities observed in RDE rarely translate to high performance in MEAs, and, in many cases, even fall short of the performance of baseline materials.[38] This not only emphasizes the need to consider support effects in catalyst development, but also the stability of materials in the MEA vs. aqueous environment[69] and the effects of electrode structure on catalyst performance, discussed in following sections. Consequently, while novel catalysts with high mass activities are still of interest,[70] HDV requirements necessitate a shift in focus towards durable catalyst-support combinations, with evaluation and reporting including EOL performance in MEA, rather than just BOL ORR activity in an aqueous environment.

Ionomers and Membranes

Ionomers are used as the ion-conducting solid-electrolyte in PEFCs, where they play a dual role of both the proton-exchange membrane (PEM) separator that facilitates ion transport between the electrodes, as well as the electrolyte binder in the CLs, where it provides percolated ion-conduction pathway and binds the supported catalyst particles.[75,90] Within the CL, catalyst ionomers exist as nanometer-thick electrolyte “thin films” that allow for reactant gas and water transport in addition to proton conductivity. Ionomer properties within the CL can deviate significantly from bulk membrane ones and are highly dependent on the interactions with the rest of the CL constituents and are discussed in a later section below. Thus, performance and durability requirements in fuel cells necessitate optimized ionomer multi-functionalities tailored for PEM and CLs.

As a PEM, membrane chemical-mechanical stability becomes even more critical for HDVs due to the longer required lifetime, higher operating temperatures, and the importance of minimizing hydrogen and oxygen crossover for higher efficiencies. In addition, new degradation mechanisms could emerge in HDVs over timescales that were previously not assessed in LDVs,

including a much longer period of exposure of other system components that can induce new or accelerated degradation pathways (such as iron ions acting as Fenton's reagents). Membranes for LDVs are based on perfluorinated sulfonic acid (PFSA) ionomer chemistry. PFSA-based membranes are the current state-of-the-art due to their ability to combine high ionic conductivity and inherent chemical-mechanical stability owing to their fluorocarbon chemistry. Ionomers with hydrocarbon-based random and block-co-polymers have also been investigated for PEMFC applications, resulting in ionomers that can exhibit conductivity comparable to that of PFSAs, however their long-term durability in a cell has not yet been demonstrated. While some hydrocarbon ionomers allow for reduced cost as well as tailored structures with a wider range of equivalent weight (inverse of the ion-exchange capacity) that can be tuned for transport, the challenge remains of meeting both mechanical toughness (to withstand the humidity cycling during operation) and chemical stability (to survive cell operation) requirements in a single material.[71,72] State-of-the-art PFSA membranes are mechanically supported, usually with an intra-membrane reinforcement, and chemically, with dopants, such as oxides of cerium and manganese, to scavenge the radicals that are known to attack the ionomers.[34,73] These radical scavengers, however, are mobile and readily move into the cathode at operating potentials and can result in performance losses.[34] In addition to additives, side-chain modifications as well as pretreatment and processing effects could provide additional parameters and tunability in membrane design.[71,73-77] Membrane thickness could become another design parameter for simultaneous optimization of performance and durability. This is in light of the increased emphasis on durability and fuel cost, making issues such as local non-uniformities, hydrogen and oxygen crossover, Ce depletion and mobility, and reinforcement strategies more important and requiring a different optimization function compared to LDVs discussed elsewhere.[34]

In addition, higher temperature operation, near or above 100°C, requires different operating strategies as higher humidification becomes untenable due to the need for pressurization, thereby necessitating novel, high-temperature membranes that maintain conductivity at lower relative humidity. Another important area is the exploration of the effect of other components on membrane durability, such as bipolar plates (through corrosion and leaching of TMs), cathode-catalyst layers (via interface misalignment and interactions), and gas-diffusion-layer (GDL) structure (through penetration by carbon fibers), even though some of these could be ameliorated using system-design strategies. Improved understanding of the effects of air contaminants, component leaching, radical scavenger migration, or any new additives that may be present in the hybrid membranes will be critical for improving membrane durability.

Electrode structure

An increase in cathode electrode performance will be necessary to achieve the MEA-level ORR mass activities required (>1 A/mg_{Pt}) and enable the high efficiencies that would yield cost savings vs. conventional diesel engines. Approaches to improving performance at high potentials include modifying the carbon support structure, manipulating support/catalyst interactions as mentioned above, and tailoring ionomer/ORR catalyst interactions either through material or integration strategies (6a). However, with total lifetime fuel cost as a primary driver, specific protocols will need to be developed to understand how MEA efficiency changes over the projected 1M mile lifetime of operation as performance assessments at BOL and after ASTs will not be enough to correctly project fuel cost for an MEA or system.

Drawing from a decade of LDV work focused on reducing overall Pt loading and improving local oxygen transport,[7,78] **Figure 6a** illustrates how ionomer loading, Pt particle location and accessibility,[61] ionomer adsorption,[79] ionomer conformation,[80] chemistry and confinement,[81] as well as Pt/C/ionomer aggregation[82] offer opportunities to tailor electrocatalyst/ionomer interactions and integration, resulting in improving overall electrode design for high-current-density operation. The well-documented oxygen transport losses (R_{nF}), which occur near the reaction site when operating low Pt roughness (*i.e.*, f_{Pt} [cm^2_{Pt} cm^{-2}_{MEA}] < 40) electrodes at high current densities, have been shown to be the largest contributor to transport-related voltage loss for LDV applications.[78,83] While 6b shows that the increased Pt content in HDV PEMFCs significantly reduces these losses at BOL, local transport losses may continue to be a concern as electrocatalyst surface area decreases at the end of stack life.

In PEMFC catalyst layers, ionomer is present across a distribution of nm-thick films on the catalyst/support and as free aggregates (Figure 4). In the 5-50 nm film thickness regime, ionomer behavior is influenced by confinement and substrate interactions, altering the film's structure and limiting hydration and species mobility (*e.g.* reduced proton and O_2 transport).[74,79,84-92] The variations in R_{nF} due to modifications of ionomer to carbon (I:C) ratio are related both to the increasing proportion of thicker films[88,91,92] as well as increased ionomer aggregation resulting in increased pore blocking in the bulk of the electrode and within the electrocatalyst aggregates. While changes to ionomer equivalent weight (EW) have not been found to significantly affect R_{nF} (**Figure 6a**), it has been found to affect the level of electrocatalyst agglomeration. Consequently, modifications to ionomer chemistry/structure could further alter the ionomer-catalyst interactions and local transport functionalities.[74,84,85,87-89,91,93]

Since the exact approaches and materials used to improve LDV performance may not be directly translatable to HDVs, additional studies must be performed to begin to unlock the full potential of high-performance materials within the electrode structure. In a recent example, a sulfonated polymerized ionic liquid block copolymer was combined with Nafion and a Pt/Vu based electrocatalyst to produce an MEA where Pt-oxide formation was suppressed at high potential, enabling ORR specific activities on par with those obtained from ionomer-free (i.e., unpoisoned) RDE experiments. As a result, the mixed ionomer electrode achieved a doubling of catalyst mass activity.[94] While perhaps only pertinent to catalysts using non-porous supports where the vast majority of electrocatalysts particles interact directly with the ionomer, there is an opportunity to tailor the electrocatalyst/ionomer interface to enable higher efficiency HDV electrodes. Such electrodes could further benefit from development of novel ionomers with high-gas permeation and tunable substrate-interactions. Furthermore, despite an improved understanding across the community, there is still a need to develop an ink-to-electrode structure understanding with which one can dictate catalyst-layer structures by controlling catalyst/ionomer interactions in the ink, utilizing optimized ink processing, and through tailored electrode break-in (conditioning) procedures.

Gas Diffusion Layers

GDLs typically consist of a carbon black microporous layer (MPL) on top of carbonized fibers. The whole layer is teflonated to form a mixed wettable material. These GDLs tend to show decreasing hydrophobicity with operating time, which changes its ability to remove water, thereby affecting performance. While these hydrophobicity changes have not been a primary

life-limiting factor for LDVs, the effects will be exacerbated with the longer HDV lifetime, possibly requiring new materials or additional treatments. For example, the addition of carboxymethyl-cellulose has been demonstrated to improve GDL durability when subjected to both chemical and mechanical ASTs, dramatically reducing the change in power density, Ohmic resistance, and mass-transport resistance.[95] Similarly, the use of microporous layers (MPLs) with hydrophilic pores[96] and renewed investigations on the GDL/channel interface where droplet shedding occurs demonstrate areas of increased interest for HDV PEMFC optimization.[97]

Bipolar Plates

Cost estimates indicate that bipolar plates represent approximately $8.2/kW or 30% of the PEMFC stack cost at production volumes of 500,000 systems per year.[98] Preferred materials, like stainless steel or titanium, show inherent corrosion resistance such that imperfect corrosion-resistant coatings do not result in significant release of corrosion products. However, as HDVs require significantly longer lifetimes, small amounts of corrosion products such as Fe^{2+} may prove problematic for membrane degradation as they act as Fenton's reagents to catalyze radical formation. In addition, the cost of stainless steel (SS 316L) is estimated to be $\$3.5/kW_{net}$, which is currently higher than the DOE target of $3/kW for a complete plate,[98] thus less expensive substrate materials are needed to meet commercialization targets. Titanium is an even more expensive base material than stainless steel. A strong candidate is aluminum due to its low cost and weight. Emphasis should be placed on developing cost-effective coatings for aluminum that are defect-free, conductive, and corrosion resistant. Carbon-composite bipolar plates provide another path forward to replace stainless steel; efforts should focus on improving their volumetric power density.[99]

Current status of commercial MEAs

The current state-of-the-art (SOA) materials used in commercial LDVs can effectively serve as the starting point and benchmark for the next-generation HDV components. The United States Council for Automotive Research (USCAR) provided the Fuel Cell Performance and Durability (FC-PAD) consortium with components from the Toyota Mirai FCEV, following 300hr of real-world driving.[14,56] A small portion of this analysis is presented in **Figure 7.** Cross-sectional views of the cell in **Figure 7a,b** show a 10µm-thick ionomer membrane reinforced with an expanded hydrophobic polytetrafluoroethylene (ePTFE) layer for enhanced mechanical stability. The electrodes are comprised of Pt (anode) and Pt-Co alloy (cathode) nanoparticle catalysts supported on high surface area carbon, with a total Pt loading of approximately 0.4 mg_{Pt}/cm^2 (**Figure 7c**), similar to loadings expected for HDV applications. The mean particle size of the $Pt_{0.92}Co_{0.08}$ cathode catalyst was 4.7 nm (**Figure 7f-g**), with the low Co loading reinforcing the emphasis on durability over BOL performance. Despite the low loading, Co was found in the membrane, supporting the earlier discussion regarding the need for dissolution-resistant catalysts.

A non-homogeneous ionomer distribution within the cathode, with larger ionomer aggregates near the membrane (shown in green) and regions of dense agglomerates (shown in red) that exhibit limited ionomer infiltration (**Figure 7d**) illustrate the need for further electrode optimization. Large ceria particles were added to the MPLs of the GDL for membrane degradation mitigation. The bipolar plates (not pictured) were carbon-coated titanium with a 3-D fine mesh flow field on the cathode. These observations from commercial LDVs demonstrate

that while much progress has been made, significant research and development is required to meet not only LDV targets, but the enhanced durability and efficiency requirements for HDVs. There remains a critical need to develop robust, efficient, and cost-effective MEAs tailored to the HDV application with more emphasis on efficiency and durability rather than the cost of components.

Outlook

It is an exciting time for hydrogen and fuel cells. The rapid developments in fuel-cell materials, integration, and manufacturing over the last decade will continue to accelerate as world-wide interest in hydrogen grows as both a value proposition and means for decarbonizing our energy system. In the transportation sector, we see an imminent shift in focus from light- to heavy-duty applications. This shift is due in part to the decrease in Li-ion battery costs that are making battery electric vehicles the incumbent technology for LDVs, as well as infrastructure challenges with distributed hydrogen. While fuel cells remain advantageous compared to batteries in many areas (*e.g.*, cost for increased driving range, refueling time, etc.) and will remain so with further improvements, such differences greatly increase as one moves towards the heavy-duty and fleet applications. Such a movement also ameliorates the hydrogen infrastructure challenges by concentrating refueling stations.

However, even with this focus, early electrified HDVs will likely be battery driven due to existing recharging infrastructure and earlier deployments. As fuel cell technology evolves, the inherent advantages will provide for its rapid adoption, especially for high power applications such as long-haul trucks, trains, buses, and maritime. The change in focus from light to heavy duty vehicles exacerbates durability and efficiency challenges for fuel cells, necessitating material and system innovations that enable new classes of hydrogen vehicles that take advantage of the high efficiency, power density, and scalability of this technology. As HDVs make their way to the road, installation of the required hydrogen infrastructure, in addition to the increased levels of hydrogen production, distribution, use, and storage through the DOE H2@Scale initiative, will pave the way for widespread adoption of light-duty FCEVs.

Acknowledgements

This work was supported by the U.S. Department of Energy's Hydrogen and Fuel Cell Technologies Office (DOE-HFTO) through the Fuel Cell Performance and Durability (FC-PAD) consortium, technology managers Greg Kleen and Dimitrios Papageorgopoulos. This research used resources of the Advanced Photon Source (APS), a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357, with electron microscopy conducted at the Center for Nanophase Materials Sciences, which is a DOE Office of Science User Facility. It is also supported by Lawrence Berkley National Laboratory under Contract No. DE-AC02-05CH11231, Oak Ridge National Laboratory under Contract No. DE-AC05-00OR22725, Los Alamos National Laboratory under Contract No. 89233218CNA000001, and the National Renewable Energy Laboratory, operated by Alliance for Sustainable Energy, LLC, for the U.S. Department of Energy (DOE) under Contract No. DE-AC36-08GO28308. The views expressed in the article do not necessarily represent the views of the DOE or the U.S. Government.

Competing Interests

The authors declare no competing interests

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101 US Department of Energy. *Fuel Cell Truck Powertrain R&D Activities and Target Review Workshop* (2018); https://www.energy.gov/eere/fuelcells/fuel-cell-truck-powertrain-rd-activities-and-target-review-workshop-h2-scale-end-use
102 Borup, R.L. & Weber, A.Z. *FC-PAD: Fuel Cell Performance and Durability Consortium* (Hydrogen & Fuel Cells Program Annual Merit Review Proceedings, 2020); https://www.hydrogen.energy.gov/pdfs/review20/fc135_borup_weber_2020_o.pdf
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105 Haug, A. *Novel ionomers and electrode structures for improved PEMFC electrode performance at low PGM loadings* (Hydrogen & Fuel Cells Program Annual Merit Review Proceedings, 2019); https://www.hydrogen.energy.gov/pdfs/review19/fc155_haug_2019_o.pdf

106 Borup, R.L. & Weber, A.Z. *FC-PAD: Fuel Cell Performance and Durability Consortium* (Hydrogen & Fuel Cells Program Annual Merit Review Proceedings, 2019); https://www.hydrogen.energy.gov/pdfs/review19/fc135_borup_2019_o.pdf

107 Borup, R.L., More, K.L. & Weber, A.Z. *FC-PAD: Fuel Cell Performance and Durability Consortium* (Hydrogen & Fuel Cells Program Annual Merit Review Proceedings, 2018); https://www.hydrogen.energy.gov/pdfs/review18/fc135_borup_2018_o.pdf

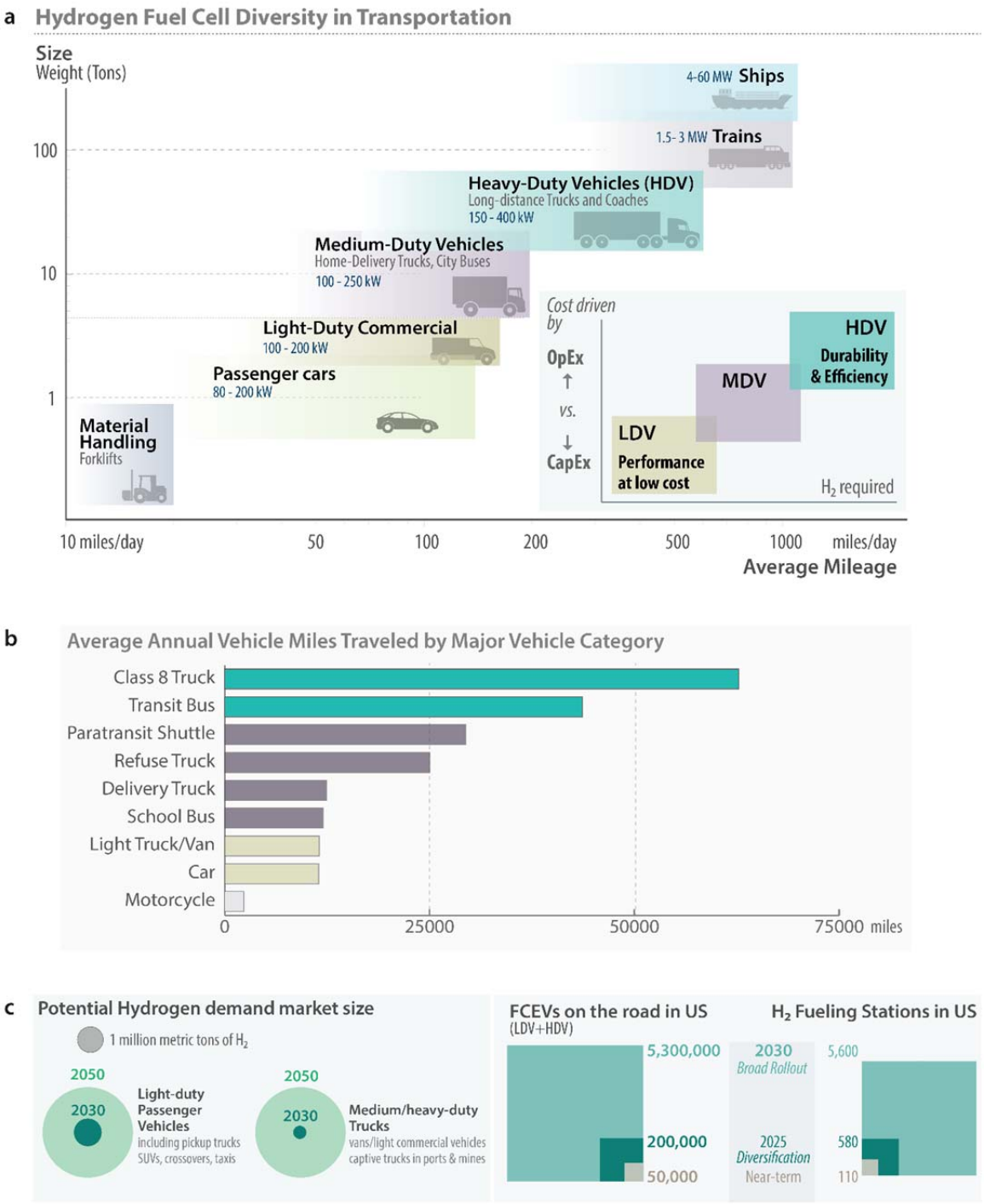


Figure 1. Roadmap to Hydrogen Fuel-Cells for Transportation. (a) Illustration of roadmap for transition from light-duty and automotive fuel cells to medium- and heavy-duty applications highlighting the paradigm shift in daily mileage and power output needs. The diagram is created by compiling information from various reports.[4,19,100,101] The inset demonstrates the trade-off between the OpEx and CapEx-driven costs for three classes of vehicles. **(b)** Annual miles traveled per major vehicle category in the US.[11] **(c)** Projected hydrogen economy and market for LDV and HDVs.[2]

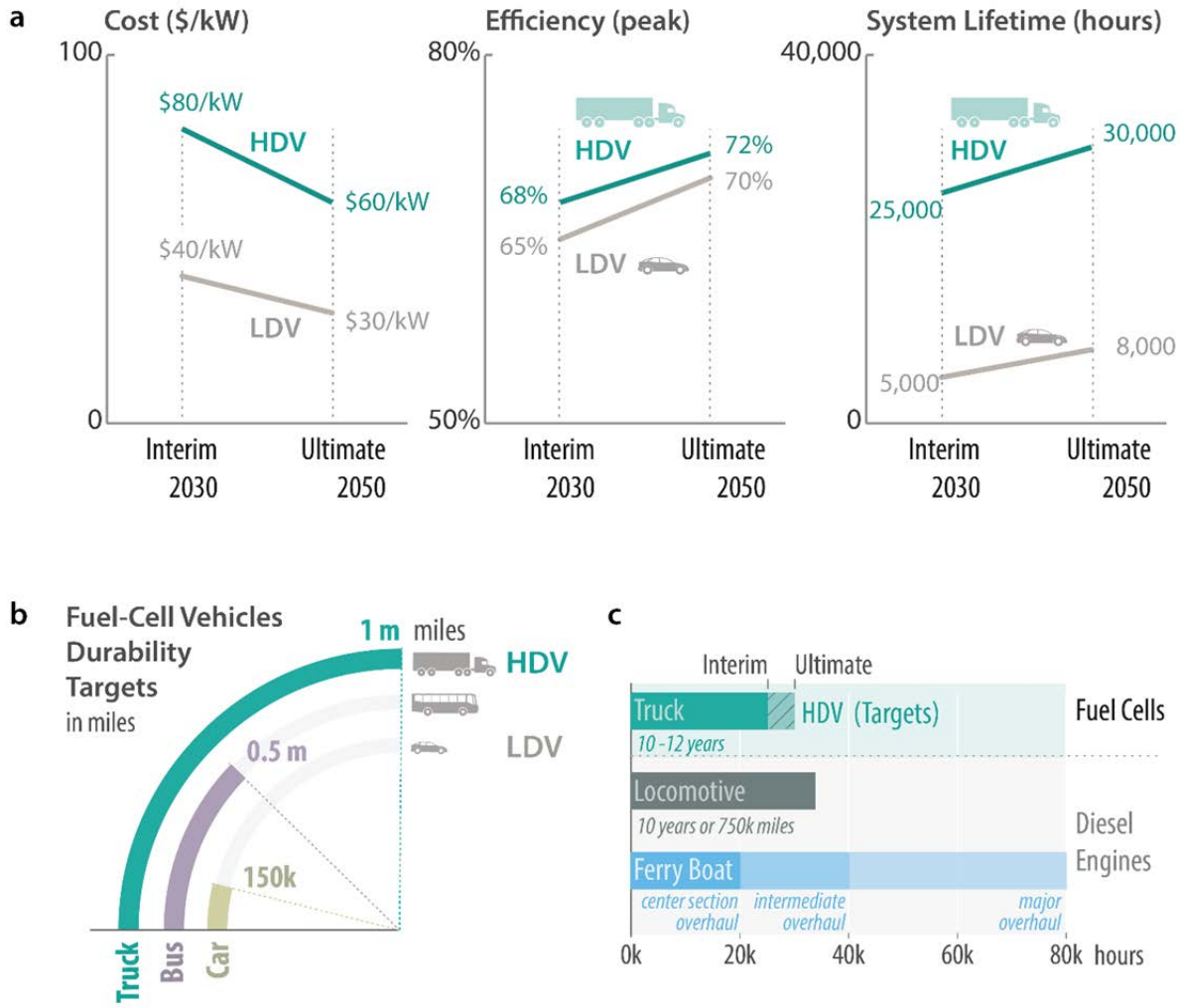


Figure 2. Summary of fuel cell targets and lifetimes. (a) Key interim and ultimate DOE fuel cell targets for automotive and trucks.[24] Current target of $50/kW for LDV is based on 100,000 units/year; HDV Targets are for Class 8 Tractor-Trailers. Ultimate targets are based on simple cost of ownership assumptions and reflects anticipated timeframe for market penetration. (**b)** Fuel cell durability targets for light- and heavy-duty vehicles expressed in terms of miles.[24] **(c)** Comparison of HDV fuel cell lifetime targets with the useful service lifetime of current diesel engines for rail (www.efrc.gov) and marine (www.wsdot.wa.gov/ferries/) applications.[22,23]

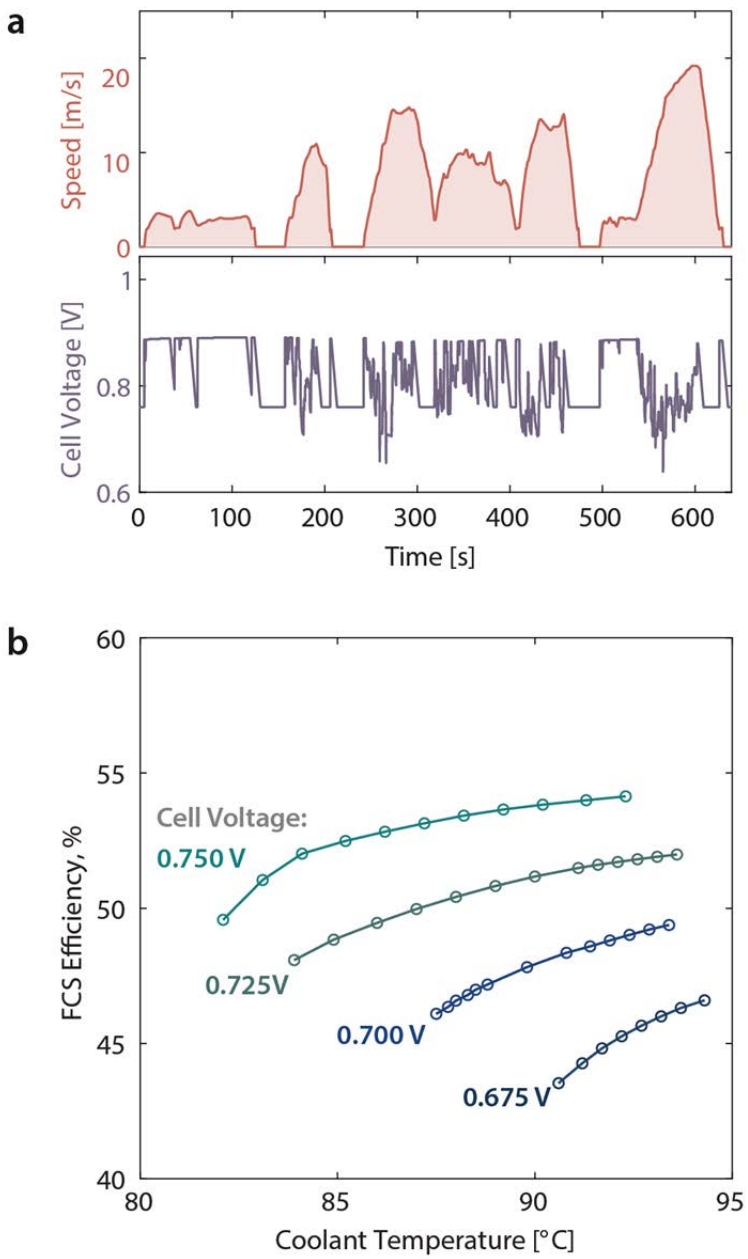


Figure 3. System demand and strategies for a HDV fuel cell system (FCS). (a) A representative HDV drive cycle for a Class 8 truck defined by the California Air Resource Board (ARB).[27] The cell voltages on this drive cycle were simulated for a hybrid 275kW FCS, 35kWh battery traction power system by assuming a minimum idle power of 20kW to limit the upper potential. (b) Relationship between the cell voltage at rated power and coolant temperature needed to reject waste heat at 6% grade.[29] Higher voltages allow operation at lower temperature and lead to higher efficiency but also lower power density (larger stack size). Panels adapted from pages 20 and 21 of ref.[29], 2020 DOE Annual Merit Review.

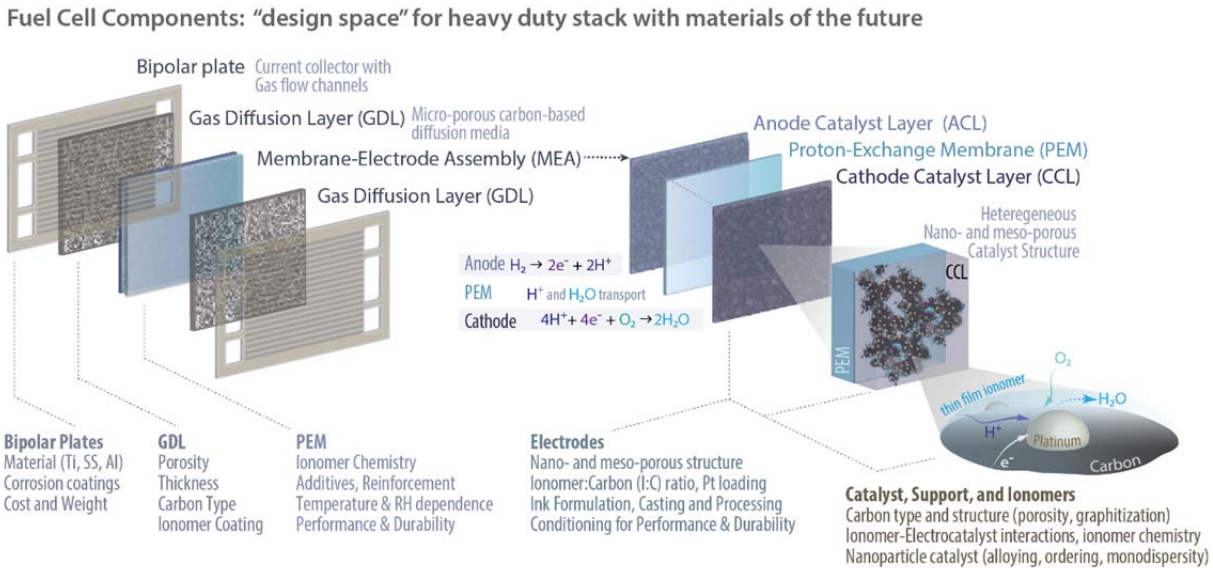


Figure 4. Design space for fuel cells. Components of a PEMFC stack with materials and design parameters and close-up view of the MEA and cathode catalyst structure, shown with anode/cathode reactions and illustration of heterogeneous porous structure of the catalyst layer and the interactions between ionomer thin-film, carbon support and Pt catalyst particles.

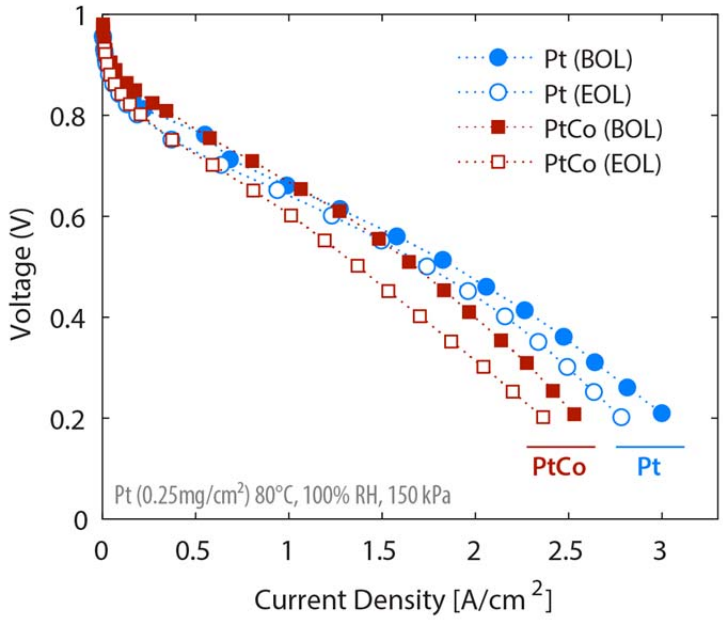


Figure 5. Impact of catalyst alloying on fuel-cell performance change over lifetime. Polarization curves at BOL and after 75,000 cycles of the square wave AST, for a Pt/C (Blue circles) and a PtCo/C (Red square) cathode catalyst-based MEA with a Pt loading of $0.25 mg_{Pt}/cm^2$. Reproduced from page 21 of ref.[102], 2020 DOE Annual Merit Review.

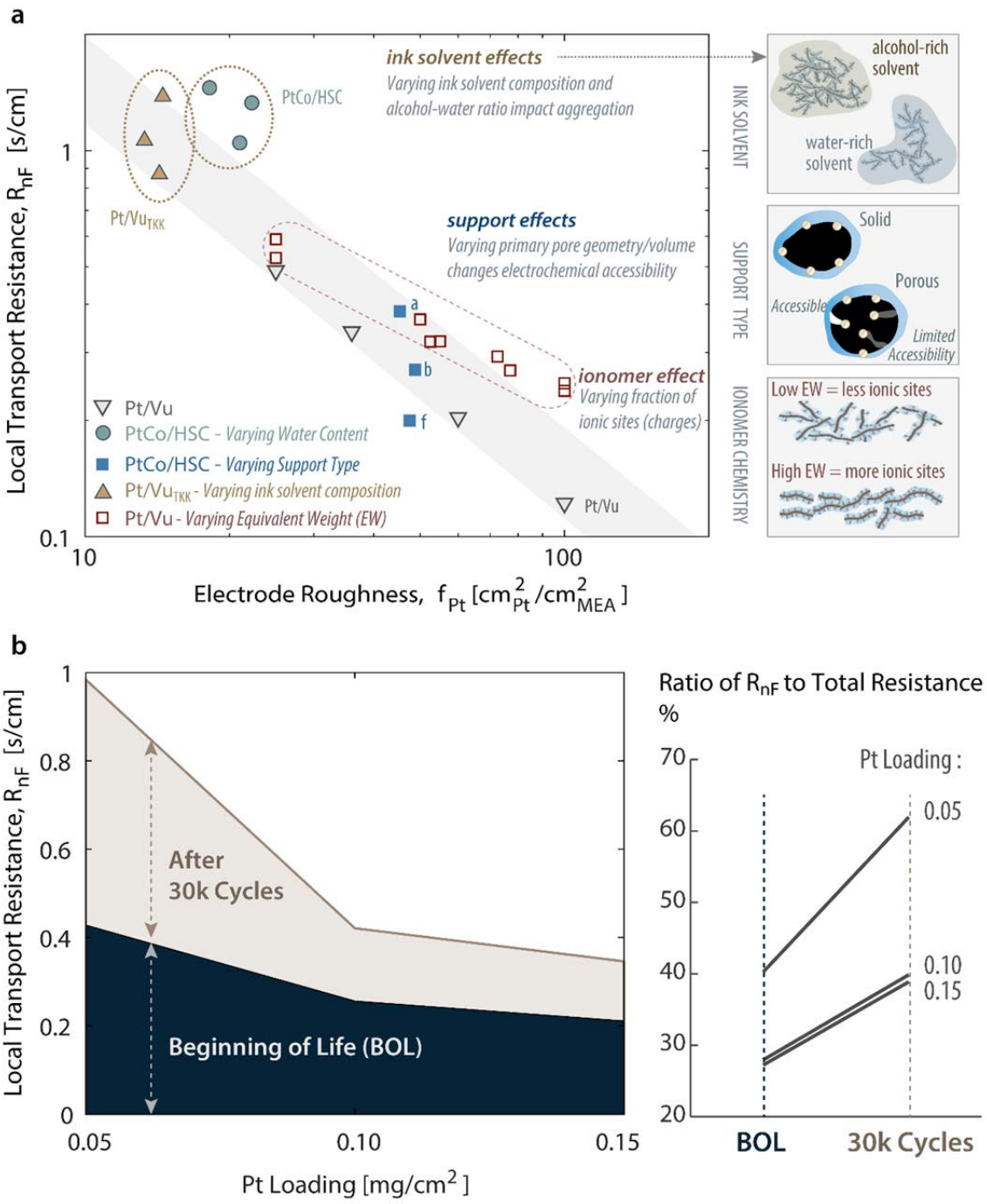


Figure 6. Performance-Durability impact on transport resistance in fuel cell electrodes. (a) Cathode CL local transport resistance (non-Fickian oxygen transport resistance - R_{nF}) as a function of Pt electrode roughness.Pictorial description of the underlying phenomena (on right). Data points adapted from ref.[80] (Pt/Vu$_{xx}$), ref.[103] (PtCo HSC - varying water content), ref.[104] (Pt/Vu), ref.[61] (PtCo/HSC – varying support type), and page 11 of ref.[105] (Pt/Vu – varying EW). **(b)** Effect of (Pt) loading on local resistance, R_{nF}, BOL and after 30,000 cycles, calculated from the breakdown of the measured polarization curves, with breakdown of contribution of R_{nF} to total resistance. Reproduced from page 19 of ref.[106], 2019 DOE Annual Merit Review.

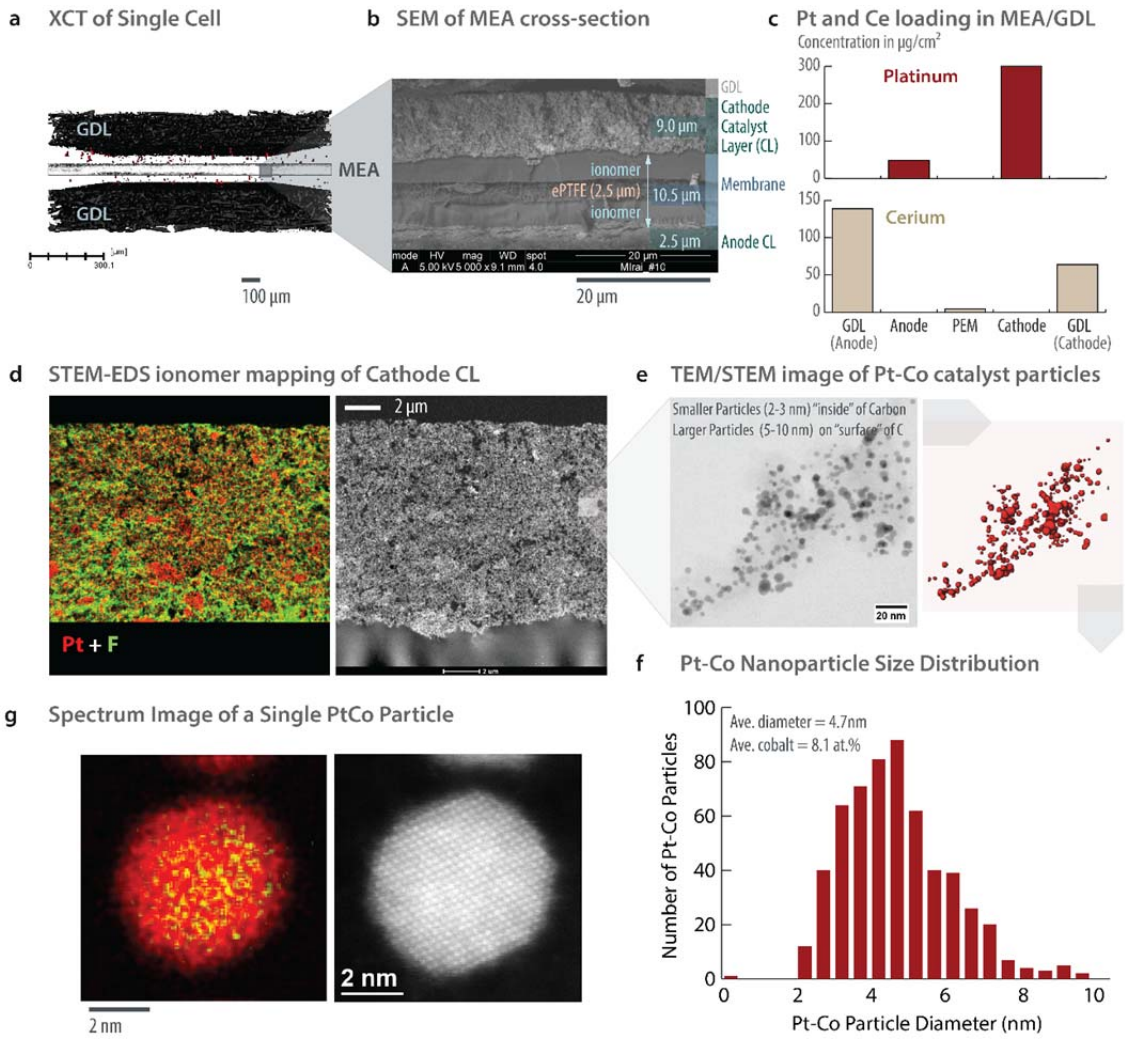
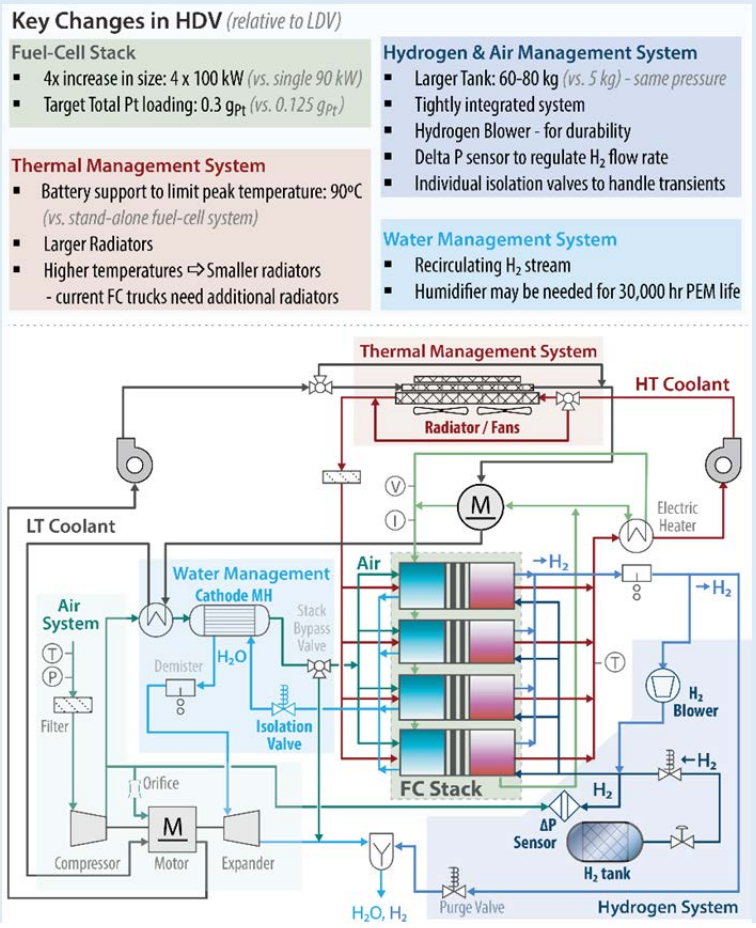


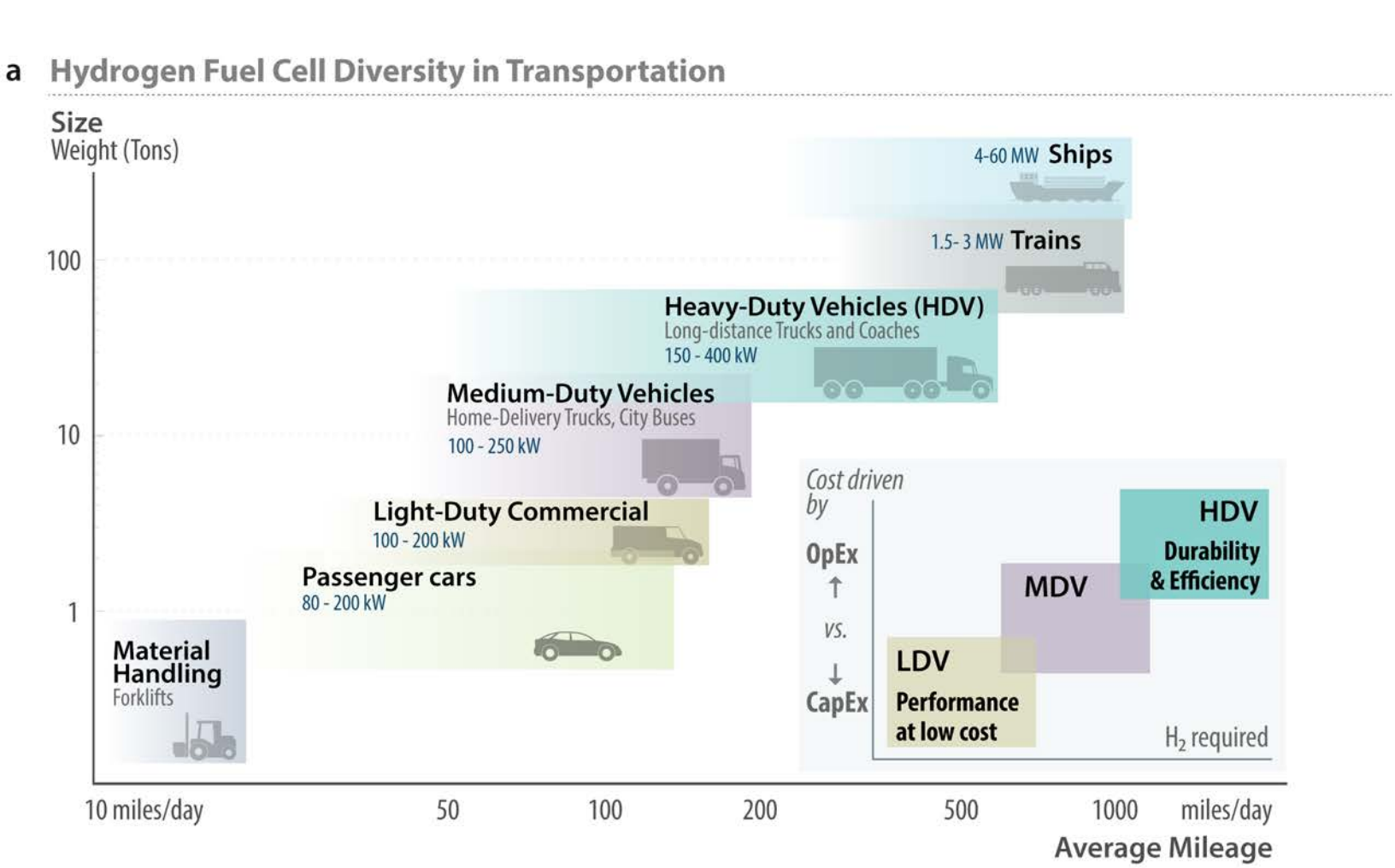
Figure 7. Current state-of-the-art materials used in passenger fuel cell vehicles and their characterization results. Summary of materials used in a single cell from the 370 cell, 114kW PEMFC stack of a Toyota Mirai FCEV; characterization performed by the FC-PAD consortium. (a) X-ray computed tomography (XCT) of the MEA and GDL showing thickness of GDL and MEA, (b) Scanning electron microscopy (SEM) of the MEA cross-section showing thickness of membrane and catalyst layers, (c) Pt and Ce loadings in cell components determined by X-ray fluorescence, (d) Scanning transmission electron microscopy and energy dispersive X-ray spectroscopy (STEM-EDS) of the cathode-catalyst layer showing fluorine (green) elemental map, representing ionomer distribution, overlaid on platinum (red), (e) STEM tomography showing distribution of Pt-Co particles over and within the carbon support, (f) size distribution of cathode catalyst particles, and (g) STEM-EDS map of individual Pt-Co nanoparticle showing alloy core covered with a Pt-rich shell. Panels adapted from pages 23-30 of ref.[107], 2018 DOE Annual Merit Review

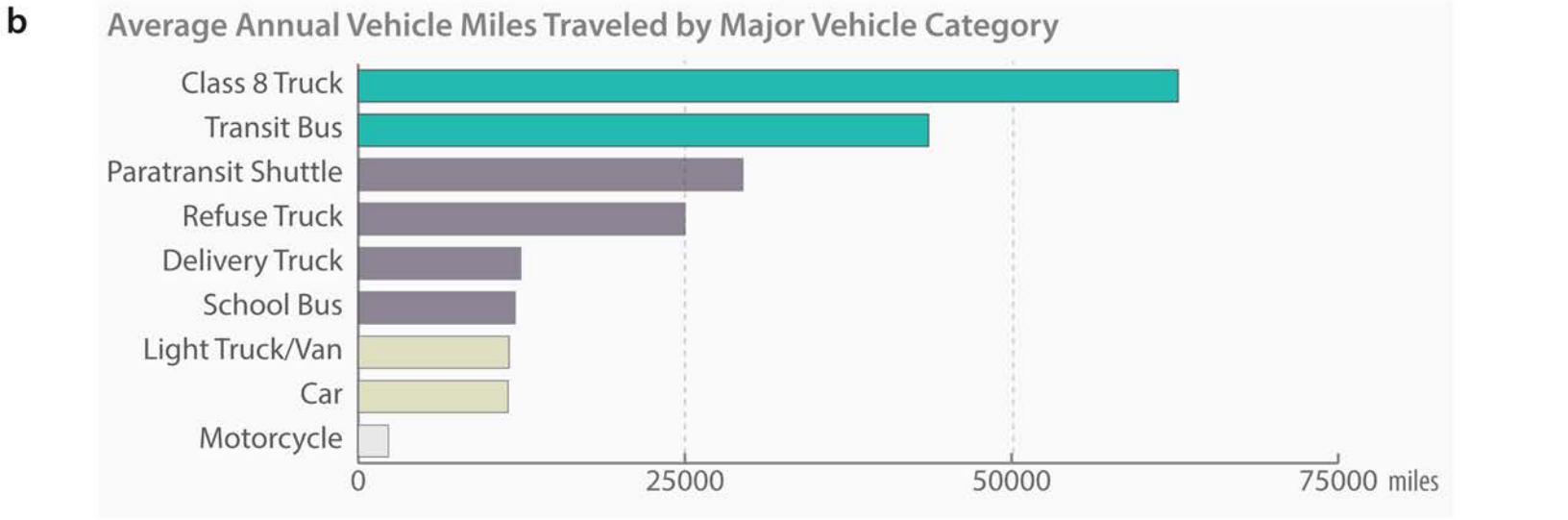
Box:

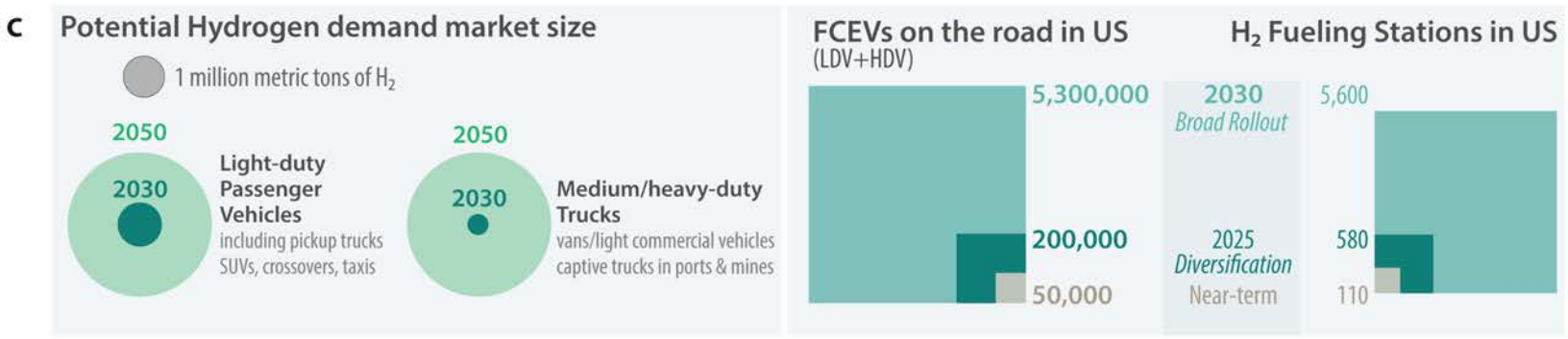
Title: Conceptual design for next-generation PEMFC system for HDVs

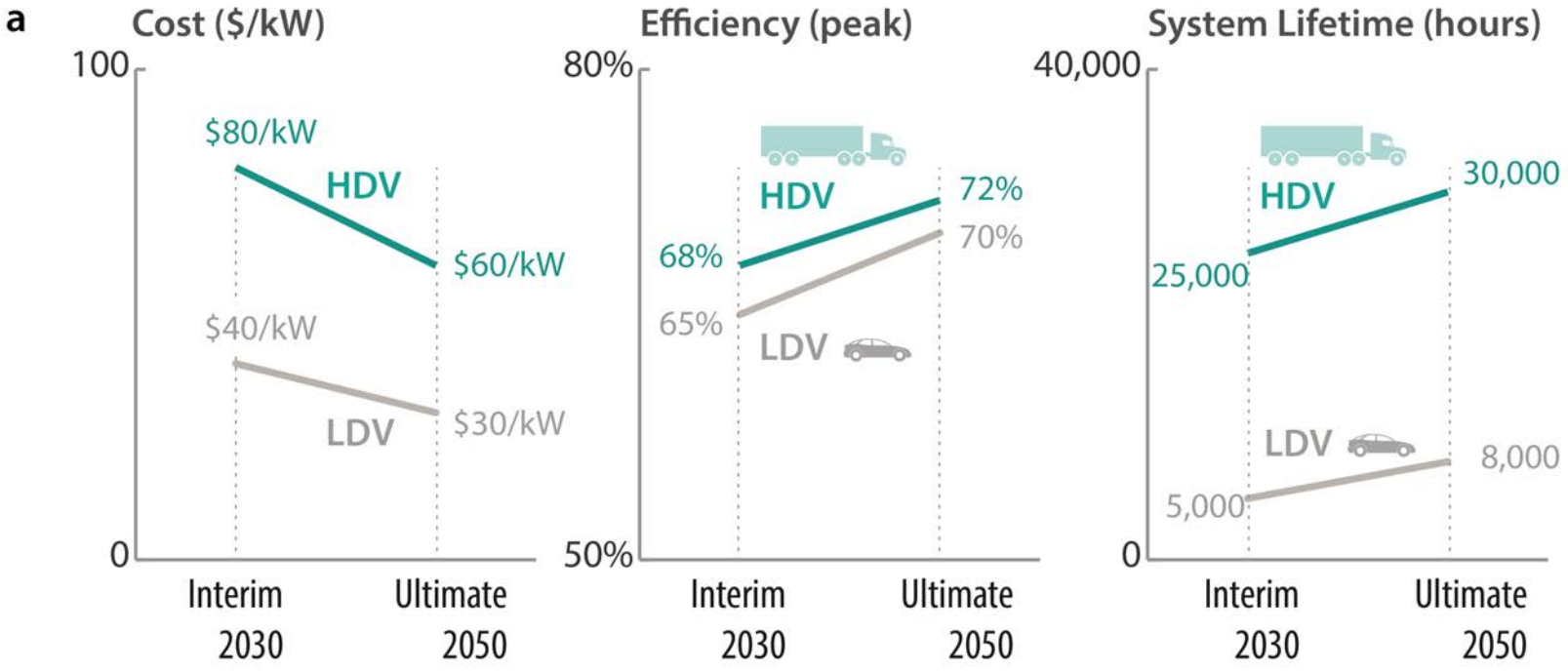
While the blueprint for HDV systems can be derived from the layouts for LDVs, the balance of plant components in HDVs have some significant differences to accommodate the larger number of stacks and to aid with the increased efficiency and durability requirements. The conceptual system is built around four 100-kWe gross stacks, standardized to promote manufacturing at volumes necessary for economy of scale. The air management system incorporates a single turbo compressor patterned after superchargers for long-haul trucks, but is driven by a motor with a motor-controller and an optional expander to reduce the parasitic power. The fuel-management system consists of a single recirculation blower, demister, and valves for regulating the inlet pressure of fresh hydrogen. The water-management system includes a cathode membrane humidifier to ensure longer membrane lifetimes by avoiding hot dry operations. The thermal-management system consists of a low-temperature (LT) circuit for cooling the compressor discharge air and a high-temperature (HT) circuit for the stack coolant. The overall system incorporates minimum controls for isolating the anode and cathode during startup and shutdown and for depleting H_2 and O_2 to avoid the formation of H_2/O_2 fronts at the anode that lead to damaging high internal cathode potentials (i.e., cell reversal). Adapted from page 16 of ref.[29], 2020 DOE Annual Merit Review

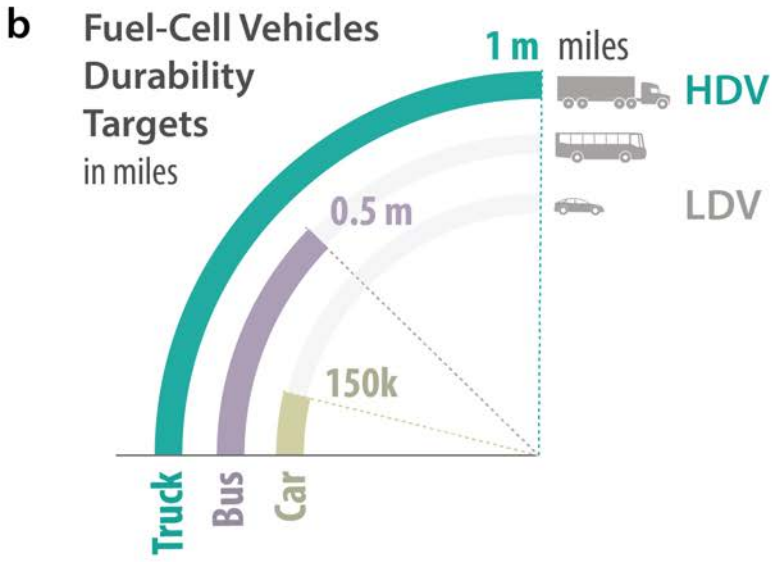


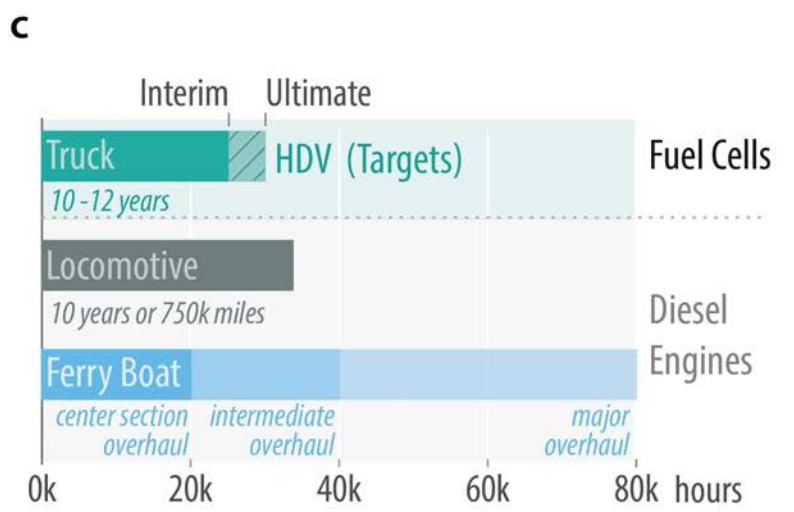

a
Hydrogen Fuel Cell Diversity in Transportation
Size
Weight (Tons)
100
10
1
4-60 MW Ships
1.5- 3 MW Trains
Heavy-Duty Vehicles (HDV)
Long-distance Trucks and Coaches
150 - 400 kW
Medium-Duty Vehicles
Home-Delivery Trucks, City Buses
100 - 250 kW
Light-Duty Commercial
100 - 200 kW
Passenger cars
80 - 200 kW
Material Handling
Forklifts
Cost driven by
OpEx
vs.
CapEx
HDV
Durability & Efficiency
MDV
LDV
Performance at low cost
H2 required
10 miles/day
50
100
200
500
1000
miles/day
Average Mileage

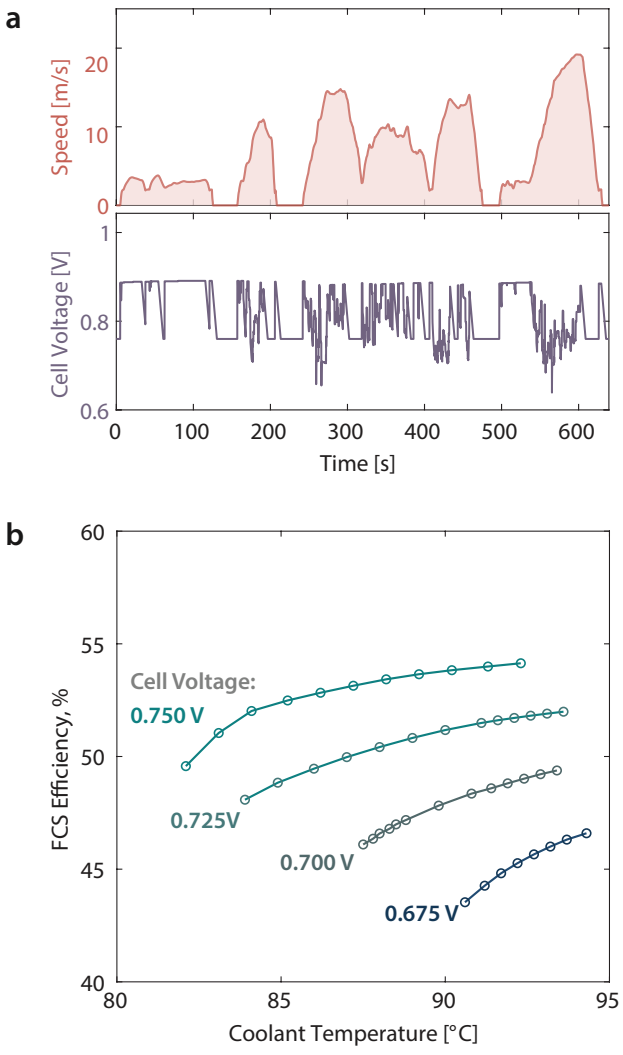

b
Average Annual Vehicle Miles Traveled by Major Vehicle Category
Class 8 Truck
Transit Bus
Paratransit Shuttle
Refuse Truck
Delivery Truck
School Bus
Light Truck/Van
Car
Motorcycle
0
25000
50000
75000 miles


c
Potential Hydrogen demand market size
1 million metric tons of H2
2050
2030
Light-duty Passenger Vehicles
including pickup trucks SUVs, crossovers, taxis
2050
2030
Medium/heavy-duty Trucks
vans/light commercial vehicles captive trucks in ports & mines
FCEVs on the road in US
(LDV+HDV)
5,300,000
200,000
50,000
2030
Broad Rollout
2025
Diversification
Near-term
H2 Fueling Stations in US
5,600
580
110

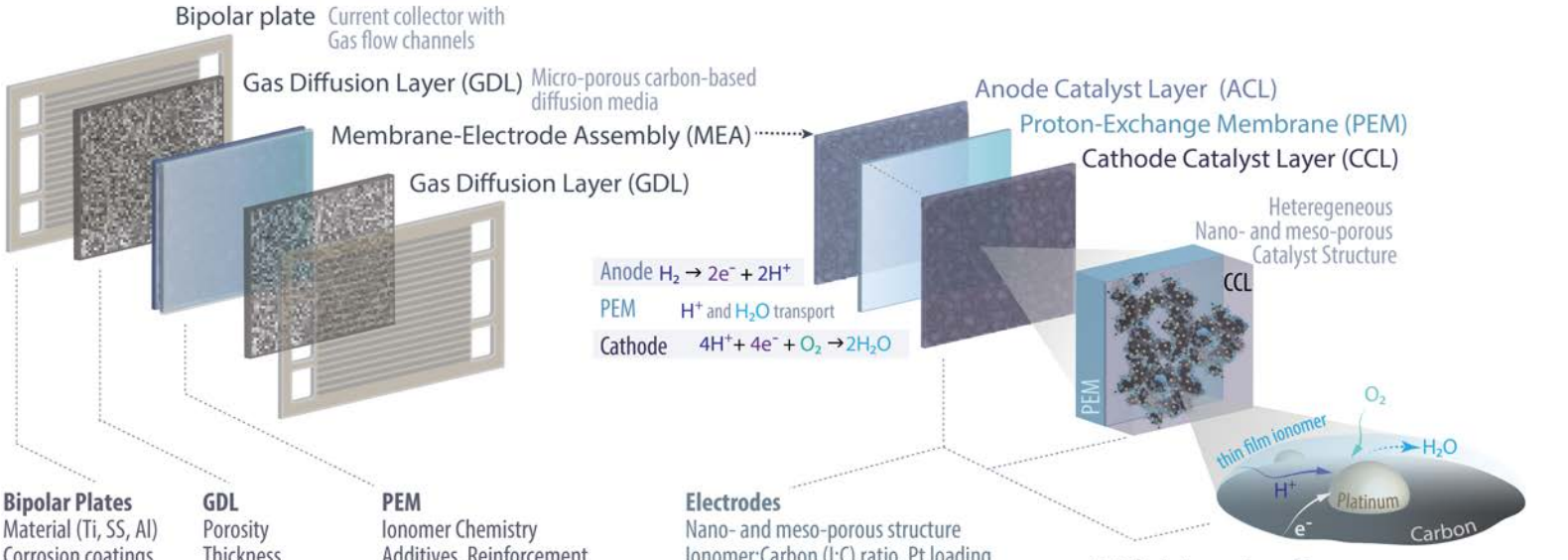

a
Cost ($/kW)
100
0
$80/kW
HDV
$60/kW
$40/kW
LDV
$30/kW
Interim 2030
Ultimate 2050
Efficiency (peak)
80%
50%
HDV
72%
68%
70%
65%
LDV
Interim 2030
Ultimate 2050
System Lifetime (hours)
40,000
0
HDV
30,000
25,000
LDV
8,000
5,000
Interim 2030
Ultimate 2050


b
Fuel-Cell Vehicles Durability Targets
in miles
1 m
miles
HDV
0.5 m
LDV
150k
Truck
Bus
Car


c
Interim
Ultimate
Truck
HDV (Targets)
10 -12 years
Fuel Cells
Locomotive
10 years or 750k miles
Diesel Engines
Ferry Boat
center section overhaul
intermediate overhaul
major overhaul
0k
20k
40k
60k
80k
hours


a
Speed [m/s]
20
10
0
Cell Voltage [V]
1
0.8
0.6
0
100
200
300
400
500
600
Time [s]
b
FCS Efficiency, %
60
55
50
45
40
80
85
90
95
Coolant Temperature [°C]
Cell Voltage:
0.750 V
0.725V
0.700 V
0.675 V

Fuel Cell Components: "design space" for heavy duty stack with materials of the future



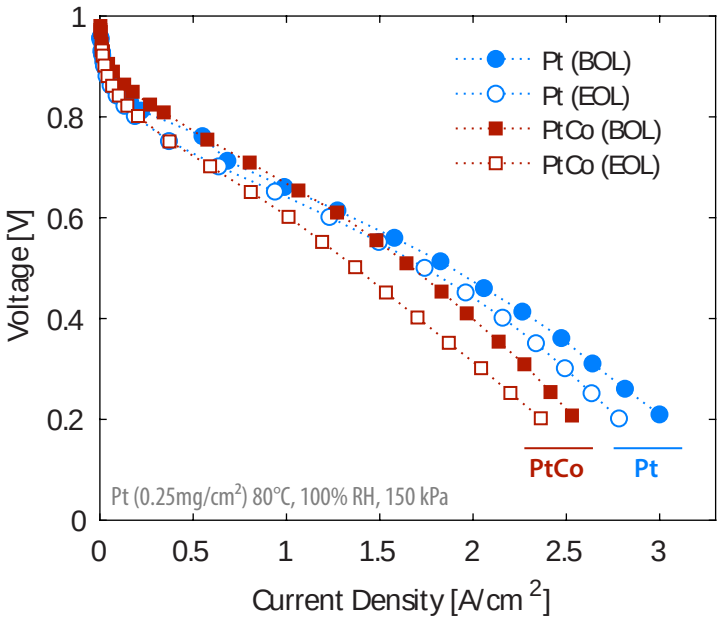
Bipolar Plates
Material (Ti, SS, Al)
Corrosion coatings
Cost and Weight

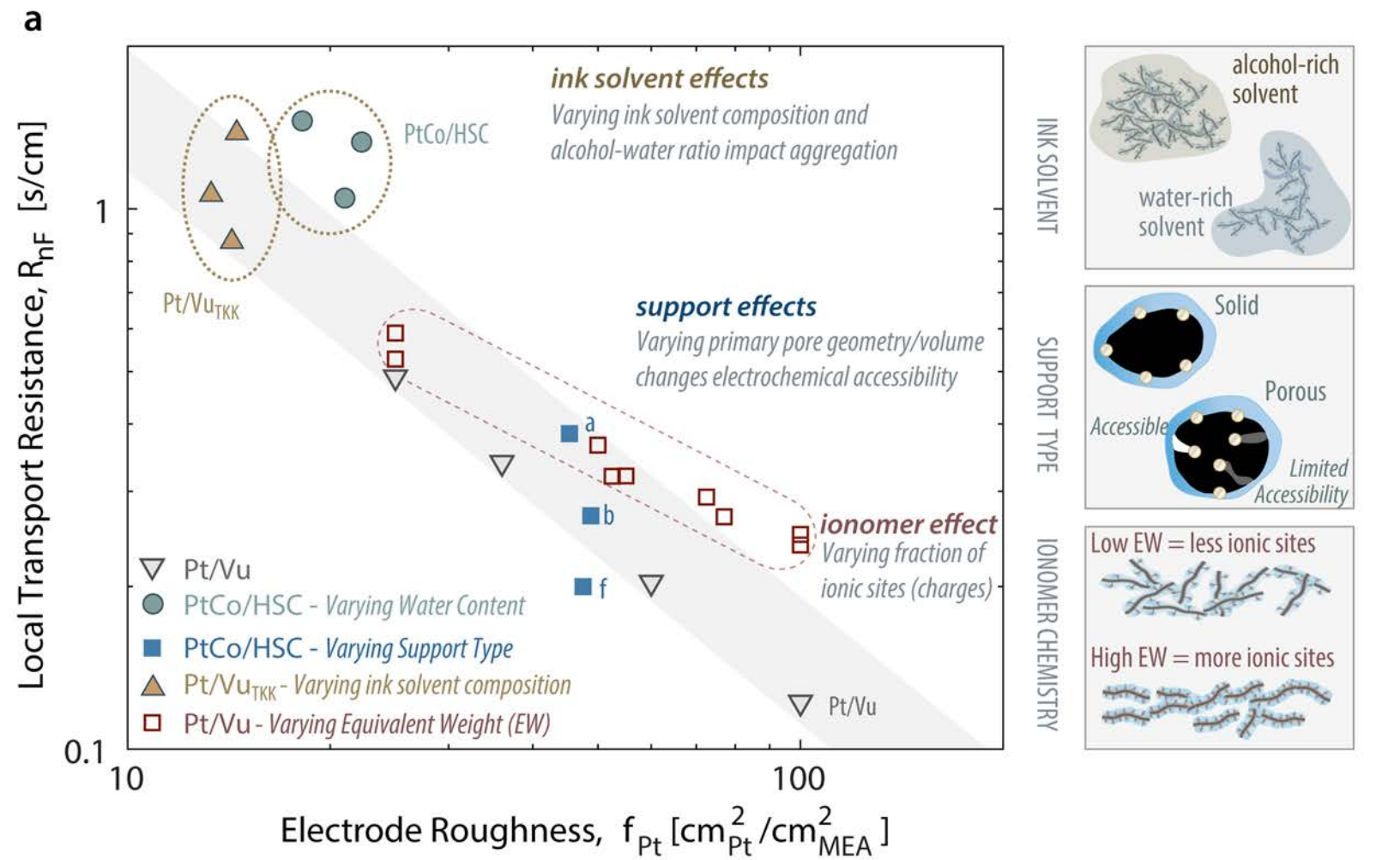
GDL
Porosity
Thickness
Carbon Type
Ionomer Coating

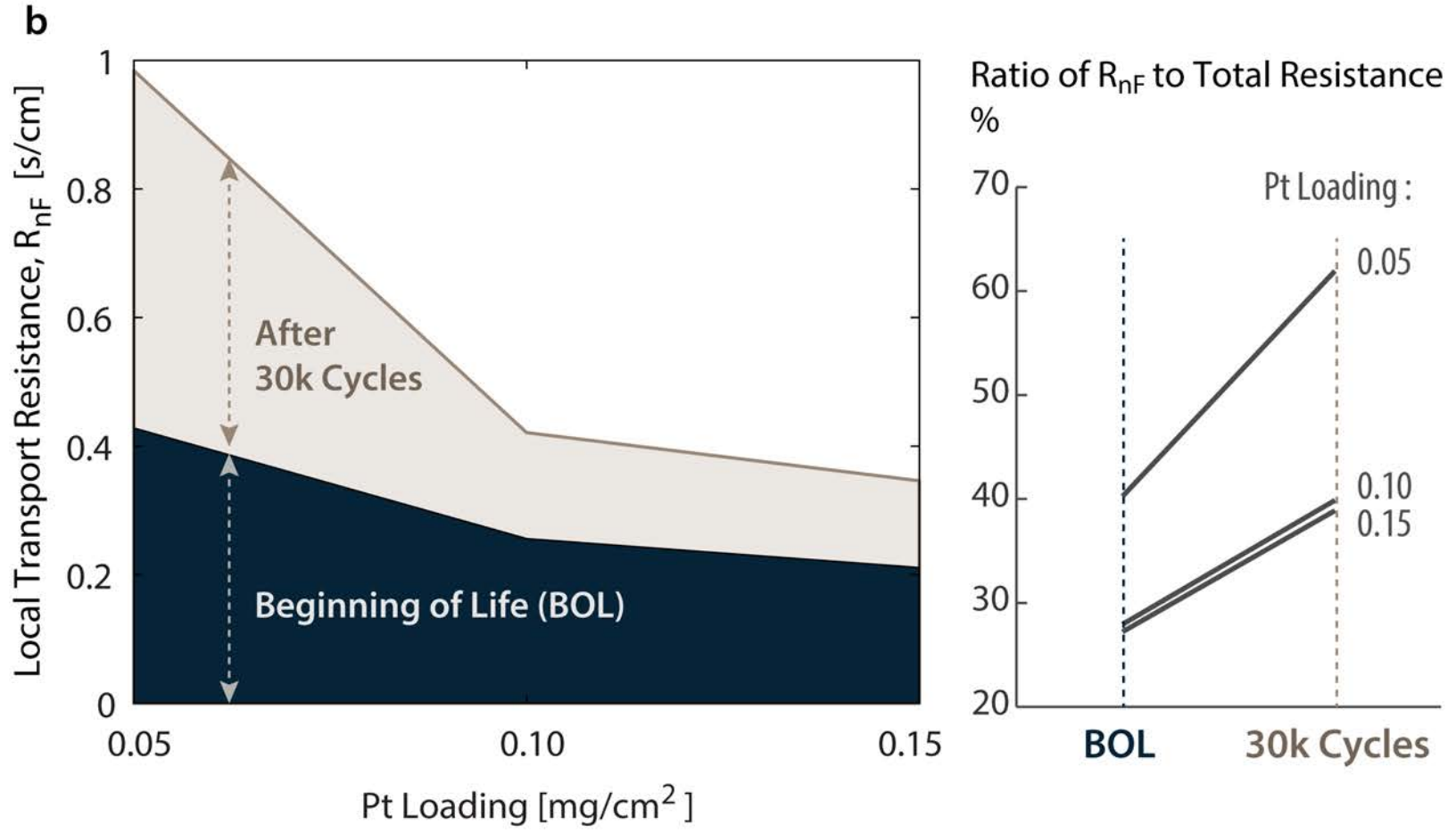
PEM
Ionomer Chemistry
Additives, Reinforcement
Temperature & RH dependence
Performance & Durability

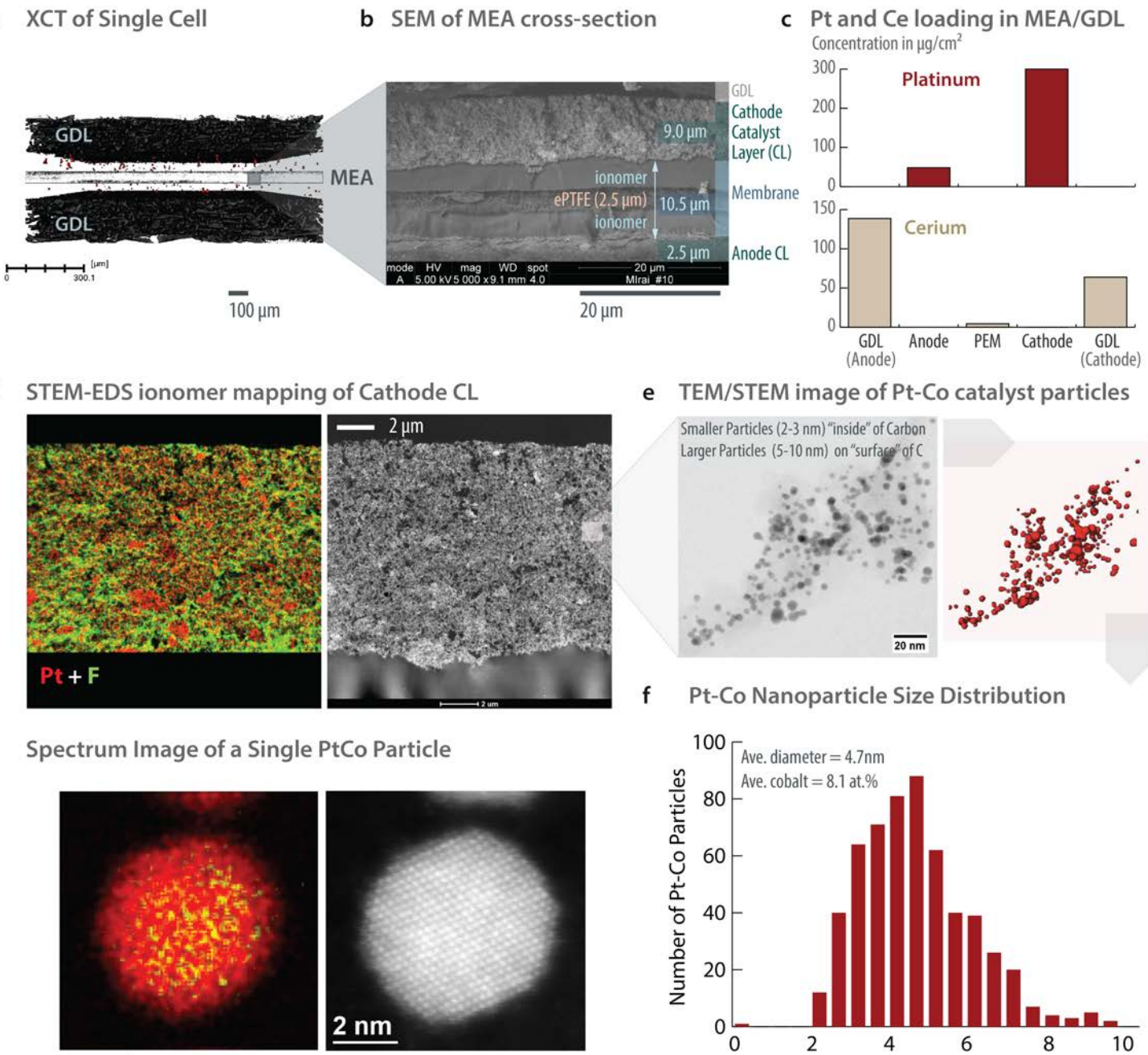
Electrodes
Nano- and meso-porous structure
Ionomer:Carbon (I:C) ratio, Pt loading
Ink Formulation, Casting and Processing
Conditioning for Performance & Durability

Catalyst, Support, and Ionomers
Carbon type and structure (porosity, graphitization)
Ionomer-Electrocatalyst interactions, ionomer chemistry
Nanoparticle catalyst (alloying, ordering, monodispersity)


Pt (BOL)
Pt (EOL)
PtCo (BOL)
PtCo (EOL)
PtCo
Pt
Pt (0.25mg/cm2) 80°C, 100% RH, 150 kPa
Voltage [V]
Current Density [A/cm 2]
0
0.2
0.4
0.6
0.8
1
0
0.5
1
1.5
2
2.5
3


a
ink solvent effects
Varying ink solvent composition and alcohol-water ratio impact aggregation
PtCo/HSC
Pt/Vu$_{TKK}$
support effects
Varying primary pore geometry/volume changes electrochemical accessibility
ionomer effect
Varying fraction of ionic sites (charges)
a
b
f
Pt/Vu
PtCo/HSC - Varying Water Content
PtCo/HSC - Varying Support Type
Pt/Vu$_{TKK}$ - Varying ink solvent composition
Pt/Vu - Varying Equivalent Weight (EW)
Pt/Vu
Local Transport Resistance, R_{nF} [s/cm]
1
0.1
10
100
Electrode Roughness, f_{Pt} [cm^2_{Pt}/cm^2_{MEA}]
INK SOLVENT
alcohol-rich solvent
water-rich solvent
SUPPORT TYPE
Solid
Porous
Accessible
Limited Accessibility
IONOMER CHEMISTRY
Low EW = less ionic sites
High EW = more ionic sites


b
Local Transport Resistance, R_{nF} [s/cm]
1
0.8
0.6
0.4
0.2
0
After 30k Cycles
Beginning of Life (BOL)
0.05
0.10
0.15
Pt Loading [mg/cm^2]
Ratio of R_{nF} to Total Resistance
%
70
60
50
40
30
20
Pt Loading :
0.05
0.10
0.15
BOL
30k Cycles


a XCT of Single Cell
GDL
GDL
MEA
100 µm
b SEM of MEA cross-section
GDL
Cathode Catalyst Layer (CL)
Membrane
Anode CL
9.0 µm
ionomer
ePTFE (2.5 µm)
ionomer
10.5 µm
2.5 µm
20 µm
c Pt and Ce loading in MEA/GDL
Concentration in µg/cm²
Platinum
Cerium
GDL (Anode)
Anode
PEM
Cathode
GDL (Cathode)
d STEM-EDS ionomer mapping of Cathode CL
Pt + F
2 µm
e TEM/STEM image of Pt-Co catalyst particles
Smaller Particles (2-3 nm) "inside" of Carbon
Larger Particles (5-10 nm) on "surface" of C
20 nm
f Pt-Co Nanoparticle Size Distribution
Ave. diameter = 4.7nm
Ave. cobalt = 8.1 at.%
Number of Pt-Co Particles
Pt-Co Particle Diameter (nm)
g Spectrum Image of a Single PtCo Particle
2 nm
2 nm

Key Changes in HDV *(relative to LDV)*

Fuel-Cell Stack

- 4x increase in size: 4 x 100 kW *(vs. single 90 kW)*
- Target Total Pt loading: 0.3 g_{Pt} *(vs. 0.125 g_{Pt})*

Thermal Management System

- Battery support to limit peak temperature: 90°C *(vs. stand-alone fuel-cell system)*
- Larger Radiators
- Higher temperatures ⇨ Smaller radiators - current FC trucks need additional radiators

Hydrogen & Air Management System

- Larger Tank: 60-80 kg *(vs. 5 kg) - same pressure*
- Tightly integrated system
- Hydrogen Blower - for durability
- Delta P sensor to regulate H_2 flow rate
- Individual isolation valves to handle transients

Water Management System

- Recirculating H_2 stream
- Humidifier may be needed for 30,000 hr PEM life