PEM Fuel Cells: Status and Challenges for Commercial Stationary Power Applications

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The past decade has seen tremendous advances in proton exchange membrane fuel cell (PEMFC) technology. However, there remain many challenges to bring commercially viable stationary PEMFC products to the market. This review, from a manufacturer's perspective, focuses on system reliability and materials compatibility and their strong impact on stack life and overall system durability. Statistical analysis is based on field data from more than 600 stationary PEMFC systems for both continuous and back-up power applications. Sealing materials and coolants are used to illustrate the approaches taken to evaluate materials compatibility studies.

INTRODUCTION

Proton exchange membrane fuel cell (PEMFC) technology is a promising alternative for secure and clean energy but it has to compete against established energy sources. Comprehensive reviews have been published on general PEMFC technology,[1–6] fuel cell components,[7–10] electrode catalysts,[11–19] membrane electrolytes,[20–24] bipolar plates,[25–26] fuel reforming,[27–30] and hydrogen storage.[31–33] However, little information is available on system component reliability, probably due to a lack of sufficient field data. This review provides a manufacturer's perspective of PEMFC technology and the challenges for stationary applications based on field data from over 600 stationary PEMFC systems. The large sample size allows for meaningful statistical analysis of system component reliability and its relationship to the overall system durability.

REMOTE CONTINUOUS POWER PRODUCTS

A stationary PEMFC system must demonstrate greater than five years of durability to compete with other distributed power generation systems.[34] System durability is often defined as the maximum lifetime of a system with no more than 10% loss in efficiency at the end of life.[2] The state-of-the-art PEMFC stack life is between 12,000 hours and 13,000 hours on simulated reformate gas.[35,36] Plug Power, a fuel-cell development company based in Latham, New York, has demonstrated a stack life of over 13,000 hours in its latest field systems operating with an integrated natural gas fuel processor. For a PEMFC stack operating with neat H_2 and air at 60–75°C and ambient pressure, the maximum voltage degradation rate needs to be 1.7 μV/h to achieve 40,000 hours of stack life.[2] In most cases, however, stationary PEMFC systems operate on an H_2-containing fuel (reformate) from a fuel processor, with natural gas and liquid petroleum gas as the most common feeds. An even lower voltage degradation rate (~1 μV/h) is required for a five-year stack life when operating a stack on a reformate containing 40–50% H_2 and 10 ppm or less CO.

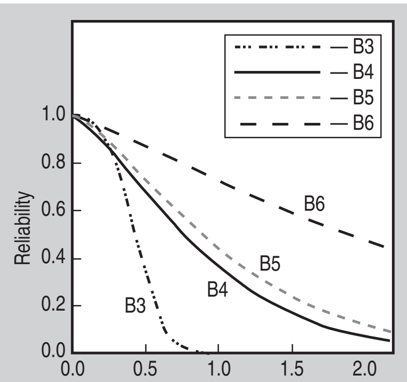

Figure 1. Weibull distributions of stack reliability for GenSys products (B = Block). B3 to B6 represent major product revisions from the original B1 design (adapted from Reference 37).

A recent study of fuel cell system reliability gave a statistical metric of durability that covered field test results obtained between 2001 and 2005 from more than 300 GenSys™ stationary PEMFC systems.[37] Over this time period, a fourfold increase in stack life was achieved almost exclusively through hardware and software design optimizations using essentially the same membrane electrode assembly (MEA).[37] Also, the average system availability improved from ~88% to over 95%.[38–40] The improvement in system reliability (see Figure 1) contributes directly to an improved stack life.

Premature stack failure has been attributed to various system component failures. Many of these are repairable and, if done in time, have minimal impact on the stack life. Such is often the case for a system tested under simulated field conditions. However, a system component failure in the field could inflict irreversible damage to a stack if the repair is not performed in time. Examples of system component failures include failed pumps, solenoid valves, flow controllers, and shift reactors. A broken coolant pump or a shift reactor with a bad catalyst bed could accelerate MEA degradation, and hence shorten stack life.

Malfunctioning solenoid valves and flow controllers could lead to permanent damage or "sudden death" of a stack through fuel starvation,[41–42] for example. Repeated start/stop cycles may also lead to premature MEA failure due to "fuel/air front" induced cathode thinning.[43] A reduction in component failure rate, site problems, and extraneous factors lowers the chance of premature stack failure and increases the system durability. This is especially true during the early development stage of a commercial PEMFC product as illustrated in Figure 2. All but

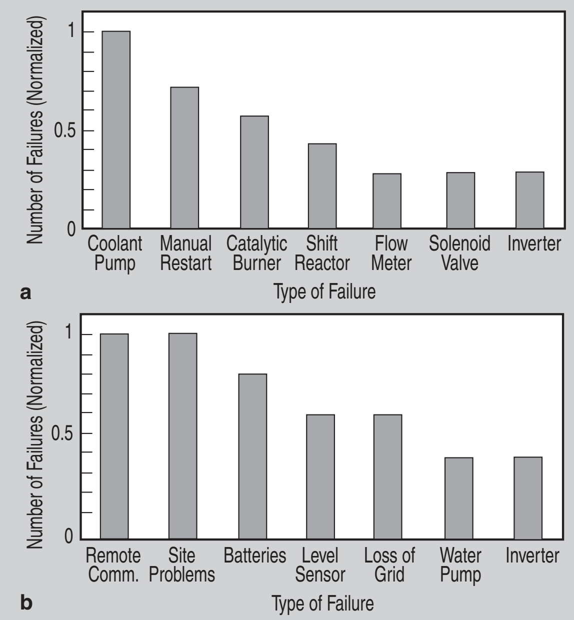

Figure 2. (a) Top seven failure modes from a sample of 75 GenSys field units commissioned in October 2002 and (b) a sample of 45 GenSys field units commissioned in June 2004 (adapted from Reference 37). Stack failures were left out of these charts since they were often linked to other system failures. Hence, the number of stack failures at the early product development stage does not accurately represent the stack reliability (comm. = communications).

one of the top seven failures are related to system components. With the hardware and software design improvements, six of the top seven failure modes were removed from the chart by June 2004. The only remaining problem, still ranked at 7 (but at a much lower frequency), is related to the inverter in the power conditioning module. Six new problems moved to the top of the chart. The limiting factors in system reliability that had indirect effects on stack life were level sensor (No. 4) and water pump (No. 5). This leaves room for further improvement in system reliability even without any technology advances in the fuel cell stack.

BACK-UP POWER PRODUCTS

To date, over 300 GenCore™ systems have been delivered to locations ranging from Iceland to Saudi Arabia and South Africa. The wide range of applications and ambient environments has provided valuable insight into the unique reliability challenges facing a PEMFC back-up power system.

A back-up power product differs from a continuously run product in several key areas: duty cycle, response time, and operating environment. A back-up power supply usually has limited run hours, often less than 50 hours a year over a typical life span of 8–10 years. When a power outage occurs, it is also critical for the back-up power system to immediately take over and support the customer load for the entire outage. Furthermore, most back-up power installations are outdoors and they must be able to operate over a wide range of environmental conditions. Despite all these challenges, PEMFC back-up power provides a durable and robust back-up solution that can be customer sized with predictable and scalable run time, as compared to the limited run time (~8 hours) of a typical battery system. Moreover, it enables remote monitoring and diagnostics and reduces personnel callouts during power outages. This leads to a lower maintenance cost than for a battery system, which requires frequent site visits to keep the batteries at full charge during idling periods. It is also more adaptable than a battery system toward harsh environmental conditions, such as hot/cold (–40 to 46°C) and dry/wet weather. Lastly, a PEMFC back-up power supply is environmentally friendly with a small carbon footprint.

The system architecture of a PEMFC back-up power product is largely driven by its duty cycle requirement. Since the stack life requirement is only about 800 hours for a back-up power system, the operating latitude is greater than that of a continuous power supply. GenCore products use neat H_2 instead of reforming gas feed. Limited run hours also allow a stack to be run at a higher current, thereby reducing the number of cells required to produce the desired power. All of these allow for a greatly simplified system design. As a back-up power system often requires frequent start/stop cycles, proper start-up and shutdown procedures must be implemented to avoid cathode thinning.[43] The idle periods of a back-up power system also pose unique design challenges for components like blowers and pumps that are best suited for continuous operation. Periodic conditioning cycles are programmed to run automatically to ensure system readiness.

The reliability of GenCore systems is measured in terms of unscheduled service calls per year (USCR/yr)—system failures that requires a service technician to repair. Figure 3 shows a normalized plot of the field experience for two design iterations.

The USCR/yr for the Block 1 systems decreased significantly after the initial release. Similar to GenSys products, the early development challenges were mostly related to system control and non-stack-component problems. System reliability improvement was achieved

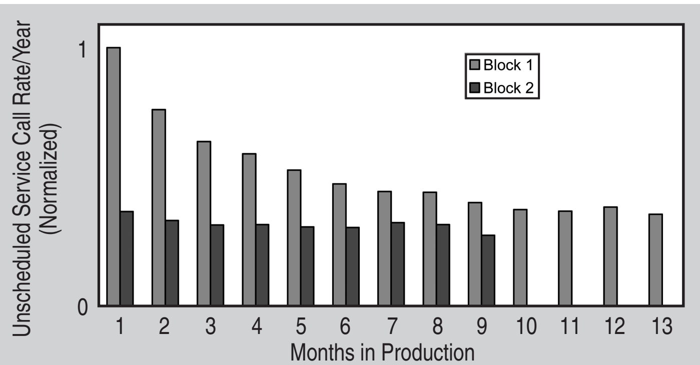

Figure 3. GenCore system reliability improvement between December 2004 and 2005. The Block 1 (B1) design was the initial release of the GenCore™ system. The Block 2 (B2) design was released approximately nine months after B1. Because of the difference in time-in-service for each fleet, all of the data were normalized for statistical analysis. This chart is compiled from the B1 products from a 13-month production period and the B2 products from a 9-month production period, with the x-axis representing the months from the design release to when a particular fleet of products was manufactured.

through control optimization and hardware design changes with the same stack materials. Initially, one of the primary failures was control issues associated with an incomplete understanding of customer applications. These failures were quickly addressed through software changes, which accounted for the rapid decrease in failure rate. The remaining failures were associated with the unique challenges related to a back-up power system, such as frequent start/stop and long idle time. These issues were addressed through the implementation of proper operating procedures.

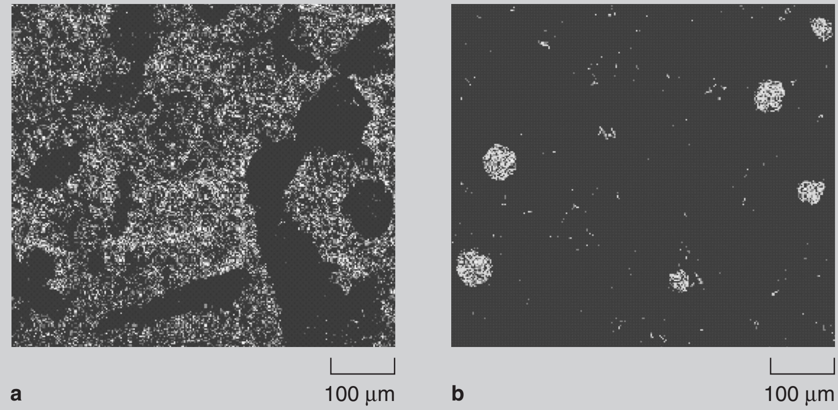

Figure 5. Scanning-electron microscopy–energy-dispersive x-ray spectroscopy images of an embrittled membrane sample. The holes and tears resulted from reduced mechanical integrity caused by the crystallization of (a) silicon- and (b) calcium-containing particles from degradation of incompatible sealing materials inside and on the surface of a membrane.

COMPONENT COMPATIBILITY AND MANUFACTURING VARIABLES

Many studies have been conducted on MEA component degradation research,[1–24] but only recently stack components other than the MEA have been studied for their effects on stack life.[44–47] To the authors' knowledge, there is no published study on component specification variation and its impact on PEMFC system performance and reliability. The importance of system component compatibility and manufacturing variables is demonstrated in this paper using examples from product development experience (see Figure 4).

The stack is only one of several subsystems in a PEMFC system with hundreds of parts and components. Component compatibility, which includes both chemical and mechanical properties, plays an important role in system reliability and overall performance (Figure 4). For example, membrane edge failure and pinhole formation, two primary causes of premature stack failure, have been linked to contaminants leached out from gaskets, bipolar plates, hoses, and other components upstream of the stack.[48] Another example is the compatibility of the sealing materials and the stack coolant required to maintain stable sealing properties.[44] Similarly, the graphite/polymer binder of molded plates must be stable and maintain proper mechanical strength during its lifetime.[25-26] Components must also be chemically stable under the stack operating conditions so that they will not leach out species that are poisonous to the electrode catalysts,[45] harmful to membrane stability and its proton conductivity,[46,47] or having adverse effects on the electrode/gas diffusion layer (GDL) properties such as hydrophilic/hydrophobic character.[49]

To select the best materials/design for a system component, one must first study its operating environment such as temperature, pressure, and its chemical/electrochemical environment. For example, the reactant side of a PEMFC bipolar plate (all sealing materials and plate components) must be able to tolerate high humidity, temperature changes, reactive chemicals, highly redox environment, and certain trace hydrocarbons and inorganic species in the gas streams. On the other side of a bipolar plate, the coolant must have stable chemical properties and must not generate any harmful species that could attack the seals, plates, or delivery hoses. At the same time, components in contact with coolant should not exacerbate its degradation. Coolant leakage should be minimized because it could lead to temporary or permanent performance loss as a result of MEA contamination and/or a change in GDL/electrode properties. Two examples are used here to illustrate the impact of component compatibility on fuel cell system durability.

Sealing Materials and Coolant Compatibility

One widely used commercial fuel cell grade sealing material did not meet materials compatibility requirements because its filler leached out and the silicon component degraded when in contact with coolant and other fluids. This led to the loss of the force retention of the sealing material and, subsequently, coolant leakage, plate shorting, and gas crossover. When leaked into the MEA, the degradation of certain coolants was accelerated through electrochemical oxidation catalyzed by platinum. Furthermore, the decomposition products of the silicon-based sealing material appeared at the membrane edges and inside the membrane through a diffusion mechanism.[44] This led to a decrease in the membrane conductivity and a reduction in membrane mechanical integrity

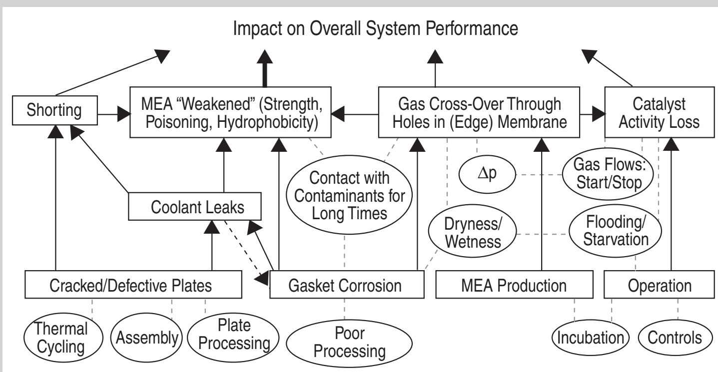

Figure 4. The impact of system components on performance.

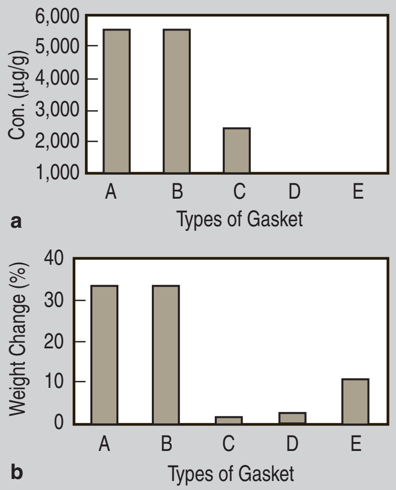

Figure 6. Examples of sealing material ranking studies. (a) A ranking of the level of harmful degradation products. (b) A comparison of the gain of mass (indicative of mechanical strength).

(Figure 5). Finally, certain fragments of this sealing material found their way into the GDL and the cathode, which resulted in the loss of cathode electrode activity and an adverse change in GDL hydrophilic/hydrophobic character. All of these factors led to premature stack failures and shortened stack life. A new sealing material was selected through an extensive ex-situ/in-situ screening process on scores of potential candidates. Each of them was subjected to a series of accelerated ex-situ degradation tests defined by a test matrix similar to the one shown in Table I. The results were ranked against each other (see, for example, Figure 6). A handful of promising candidates were further examined in situ under real fuel cell operating conditions. The final selection was based on the overall chemical and mechanical properties of the materials.

Coolant and Bipolar Plate Compatibility

Even with the best sealing material, coolant leakage is still a potential problem, especially for a system that is expected to be in continuous operation for at least 40,000 hours. Furthermore, coolant leakage can occur through the bipolar plates if the fillers or binders are susceptible to leaching. This is particularly true in the case where a coolant is incompatible with plates made from the graphite-polymer composite materials. The coolant decomposition products in turn accelerate corrosion of other components such as the radiator and gasket. The mixture of these degradation species, in the form of metal ions and organic/inorganic species, subsequently lead to MEA contamination, shunt currents in the coolant loop, and electrical shorting (*via* local precipitation) of bipolar plates. One well-documented situation is the shunt current effect on coolant stability and bipolar plate compatibility.[50–56] There is a substantial electric potential across a fuel cell stack. This requires the use of a dielectric coolant system or a coolant loop electrically insulated from the rest of the system to avoid component corrosion induced by shunt currents.[53–56] In the presence of an electric field, ionic species in a coolant loop are forced to move in certain directions depending on their electric charges, generating a shunt current. The result is the formation of a concentration gradient of charged species. This gradient facilitates leaching of trace metals and other ionic species from the bipolar plates, sealing materials, and other components in contact with the coolant. The shunt current slowly but gradually increases as this "autocatalyzed" ion enrichment process accelerates over time. In the absence of a coolant polishing system, this could lead to the blockage of coolant channels at one end of a stack and, potentially, shorting of bipolar plates under extreme circumstances. Even with a coolant polishing system, the continuous leaching of trace ionic species from bipolar plates could eventually lead to coolant leakage through the plates and, consequently, loss of stack performance. Finally, certain leaching species serve as catalysts for coolant degradation which may in turn accelerate the leaching process. A thorough materials compatibility study is therefore critical in the selection of a coolant and all components that may potentially come in contact with it. These results emphasize that, in addition to having appropriate physical properties (such as heat capacity, thermal conductivity, thermal expansion, dielectric constant, and viscosity), a coolant must also be compatible with manifolds, pumps, hoses, radiators, gaskets, and seals. Furthermore, a coolant must meet the requirements of safety standards on toxicity and flammability, regulations on shipping and waste recycling, and on environmental impact assessment regarding aquatic species and biodegradability.

Component Manufacturing Variables and System Reliability

To build a reliable PEMFC system, it is critical to minimize the component manufacturing variations. Currently, with the small scale of PEMFC production, commercial-grade fuel cell components often display substantial deviations from their product specifications. An example is the electrode thickness of commercial MEAs (Figure 7). It is evident that, even within the same sample, the thicknesses of the electrode layers can vary greatly from one region to another. Such component manufacturing issues hamper the overall effort toward improving commercial PEMFC system reliability. In the case of MEA production, part of the problem is the lack of a non-destructive in-line quality control method to ensure batch-to-batch consistency. Without adequate MEA quality control, it is difficult to interpret autopsy results and to link apparent membrane/electrode problems of used MEAs to a particular failure mechanism. Overall, the component reliability is a challenge to fuel cell manufacturers as well as their component suppliers.

CONCLUSION

Statistical analysis, based on field data from several large commercial PEMFC product deployments, has demonstrated the importance of system reliability on

Table I. Sample Matrix for Coolant-Sealing Material Compatibility Study

Tests	Coolant A (wt.%)	Acid 1 ppm	O1 ppm	O2 ppm	Acid 2 M	H_2O_2 (wt.%)
A	0	10	0	0	0.35	3
A1	0	10	0	0	0	1.5
B	0	1,000	0	0	0.5	3
C	5	0	200	300	0	0
D	55	0	0	0	0	0
E	55	0	200	300	0	0

Note: A through E are different test environments for a certain sealing material in contact with Coolant A. O1 and O2 are potential coolant degradation products. Acid 1 and Acid 2 are acidic species commonly presented in a PEMFC environment.

the stack life and overall system durability, especially during the early stages of commercial product development. The reduction in system software and component failures has been responsible for dramatic improvements on the system reliability for both GenSys and GenCore product lines.

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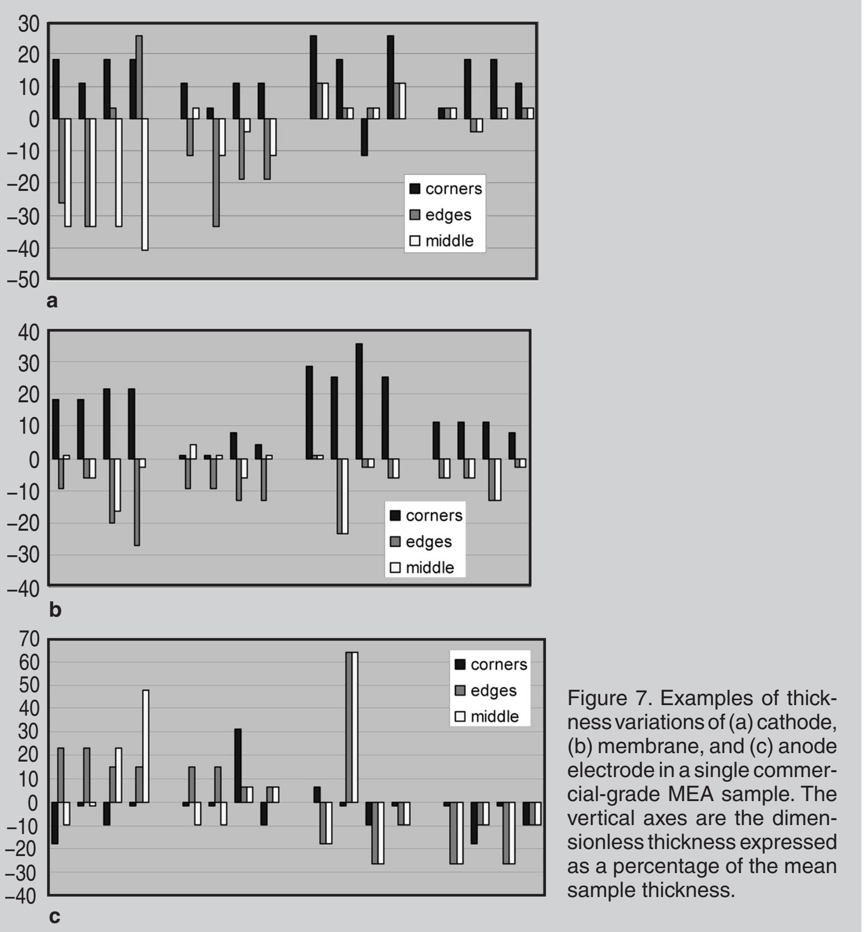

Figure 7. Examples of thickness variations of (a) cathode, (b) membrane, and (c) anode electrode in a single commercial-grade MEA sample. The vertical axes are the dimensionless thickness expressed as a percentage of the mean sample thickness.

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