V.G.3 Low Cost Durable Seals for PEMFCs

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Subcontractors:
- Freudenberg-NOK General Partnership, Plymouth, MI
- Henkel Corporation, Rocky Hill, CT
- Virginia Polytechnic Institute and State University (Virginia Tech), Blacksburg, VA

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Objectives

- Develop a working material specification to guide the development of proton exchange membrane fuel cell (PEMFC) seal materials.
- Synthesize and compound materials that meet the requirements of the materials specification.
- Evaluate candidate materials through accelerated ex situ testing to predict whether the material will meet durability objectives given in Table 1.
- Validate the performance of the best performing material candidate through in-cell testing.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

(A) Durability

Technical Targets

The aim of this project is to develop and evaluate new non-silicone liquid injection moldable (LIM) and dispensable materials to improve durability for both transportation and stationary applications while maintaining or improving on the cost benefits of LIM silicone materials (Table 1).

TABLE 1. DOE Targets Receiving Focus as Part of this Project

Characteristic	Units	2009 Status	2010 / 2011[a]
Durability	hours	>10,000 hours achieved[b]	5,000/40,000
Sealability at Low Temperature	°C	Testing to-date indicates success	-40/-35
Cost[c]	$\$/kW_{net}$	To be evaluated	(2.00 – 3.88)[d]

[a] DOE Transportation/Stationary targets
[b] Real-time out-of-cell testing at 90°C
[c] Based on high volume production (500,000 transportation systems per year/2,000 stationary units per year). Note: A cost target for seals is not currently carried in the Hydrogen, Fuel Cells & Infrastructure Technologies Program Multi-Year Research, Development, and Demonstration Plan. See footnote (d) below for an explanation of how this target was derived.
[d] Suggested cost target range for transportation applications derived from Reference 1 ($3.88/kW), Reference 2 ($2.00/kW) and conversations with the Fuel Cell Tech Team ($2.00/kW). Based on Reference 1, a reasonable suggested target for stationary applications may be $6.03/kW.

Accomplishments

- Completed more than 10,000 hours of out-of-cell testing at 90°C.
- Completed more than 4,500 hours of out-of-cell testing at 120°C.
- Developed a new material candidate that is expected to have greater high temperature stability.
- Began comparisons against other materials previously used or being considered for use.

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Introduction

Seal durability is critical to achieving the 2010 DOE operational life goals for both stationary and transportation PEMFC stacks. The seal material must be chemically and mechanically stable in an environment consisting of aggressive operating temperatures, humidified gases, and acidic membranes. The seal must also be producible at low cost. Currently-used seal materials do not meet all these requirements.

High consistency hydrocarbon rubber compounds that show promise for compatibility with the PEMFC environment are difficult to process in a way that leads to low cost PEMFC production. Silicone-based LIM rubber compounds which are easy to process in ways leading to low-cost production are highly gas permeable and have been shown to be unstable in PEMFC applications. To produce PEMFC stacks which are both highly durable and low in cost, a seal material with the stability of high consistency hydrocarbon rubber and the processing ease of a LIM silicone is sought.

Approach

To accomplish the objectives of this project, the approach is to develop and evaluate non-silicone LIM seal materials that can meet the specialized mechanical, compatibility, and cost requirements inherent to the design and operation of PEMFCs. To guide material development, a working material specification was developed. Materials developed to this specification by Henkel are evaluated through out-of-cell testing at Virginia Tech, Henkel and UTC Power in simulated environments. Using an appropriate set of accelerated testing techniques, an initial lifetime estimate will be made for the candidate materials. The best candidate or candidates will be selected for in-cell testing to validate the performance of the material in a PEMFC environment. Specimens for out-of-cell testing and full size prototypes for in-cell testing are produced by Freudenberg-NOK General Partnership.

The outcome of the project will benefit the PEMFC industry by providing a seal material specification, a material that satisfies it, and verification that the specification and the material enable a low-cost and durable seal.

Results

With the specification completed early in the project, the work over the last year has been focused on two primary tasks:

1. Synthesizing and formulating new materials for evaluation.
2. Out-of-cell material testing.

Additional material development experiments were conducted over the last year. The work has yielded another material candidate, FCS2, for evaluation. It is a one-part material with the cure characteristics of the two-part material evaluated earlier in the project. This new candidate is also designed to have improved high temperature stability.

This brings the total of material candidates evaluated to three. These materials have been given the designations FCS0, FCS1 and FCS2. Some key properties of the three are compared to each other and to the materials specification in Table 2. All three meet the minimum requirements in the material specification. This means that seal components suitable for use in PEMFC applications could be produced from

TABLE 2. Comparison of Key Properties for Three Candidates Evaluated To-Date

Henkel LIM Hydrocarbon Elastomer Property Table for DOE					
Properties	Project Requirements		FCS0	FCS1	FCS2
	Minimum	Ultimate			
Process Properties					
LIM processable	Yes	Yes	Yes	Yes	Yes
Viscosity @ room temperature (cPs)	<= 700,000	<= 600,000	~ 500,000	~ 543,000	~ 543,000
Mold temperature (°C)	< 135	<= 110	120 to 130	120 to 130	120 to 130
Mold time (second)	<= 400	<= 60	60 to 120*	60 to 120*	60 to 120*
Mechanical Propreties					
Hardness (Shore A)	15 to 68	30 to 55	31	30	30
100% Modulus (Mpa)	0.25 to 3.5	1 to 2.5	0.75	0.68	0.69
Tensile strength (Mpa)	>= 0.5	>= 0.8	1.3	1.3	1.1
Elongatin (%)	> 125	> 150	163	171	160
Tear strength Die C (kN/m)	>= 2.7	>= 5.0	3.7	3.7	3.9
Environmental Requirements					
Temperature resistance (°C)	-40 to 85	-40 to 90	-40 to 90	-40 to 90	-40 to 120
Notes					
*cure schedule: 120 second in the mold @ 120°C and then 1 hour post cure @ 130°C					

Green: meets minimum & ultimate requirements
Yellow: meets minumum requirements
Red: below minimum requirements

any of the three. Additional development continues. The next candidate, FCS3, is expected to meet the ultimate requirements of the specification. Meeting the ultimate requirements broadens the range of design options available. The material will also provide a step improvement in strength and elongation.

A variety of out-of-cell tests are being used in the evaluation of the materials being developed for this project. The aim is to evaluate the beginning of life properties and to evaluate how those properties change with time when subjected to combinations of temperatures and chemical environments typical of PEMFC operation. One of the most important measures for seals is compressive stress relaxation (CSR). This is a measure of the decay in sealing force with time. During the past year, over 10,000 hours of CSR testing was completed on the first material candidate evaluated for the project. The results are summarized in Figure 1. The test was run at 90°C in de-ionized water, a 50/50 water/ethylene glycol (EG) mix, and in air. The 50/50 water/EG mix had the greatest effect. However, the sealing force decay in all cases was less than 20%. This is a significant result. While significantly greater values can typically be accounted for in the design of sealing systems, lower values translate directly into increased durability and greater design flexibility. For reference, the sample was prepared in accordance with ASTM D 6147 Section 7.1.1.

Recognizing that there is still interest throughout the PEMFC industry, particularly in the automotive sector, to operate at temperatures as high as 120°C, CSR testing at 120°C was conducted on the second material evaluated for the project. The result is summarized in Figure 2. The test ran for 4,500 hours. Force decay at the end of the test was approximately 10%. A comparison of this data to the 90°C air aging case in Figure 1 reveals that the force decay results are similar at 4,500 hours. This suggests that physical rather than chemical relaxation still dominates after 4,500 hours at 120°C.

As hydrocarbon-based elastomers, it is expected that one of the most significant threats to the stability of the candidate materials is oxidation. Based on this expected importance, a comparison of the oxidative stability to some other materials which have been considered for PEMFC applications was begun and is still underway. A key interim result of this work is provided in Figure 3. In this figure, relative change in mass with time is tracked for testing at 150°C. In this case, FCS1 is being compared to three ethylene propylene diene monomer (EPDM)-based materials. Significant changes in the

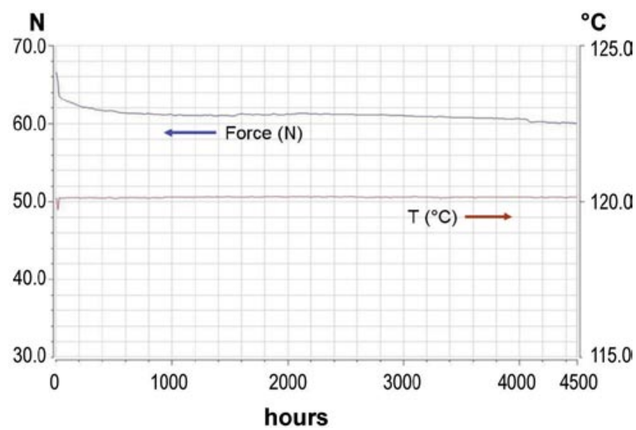

FIGURE 2. Results for CSR testing at 120°C in air. Approximately 10% load loss was observed at the end of the 4,500 hour test. This result taken in conjunction with the result from Figure 1 strongly suggest that material candidates being developed for this project will meet the 2010 DOE durability goals for automotive PEMFC applications.

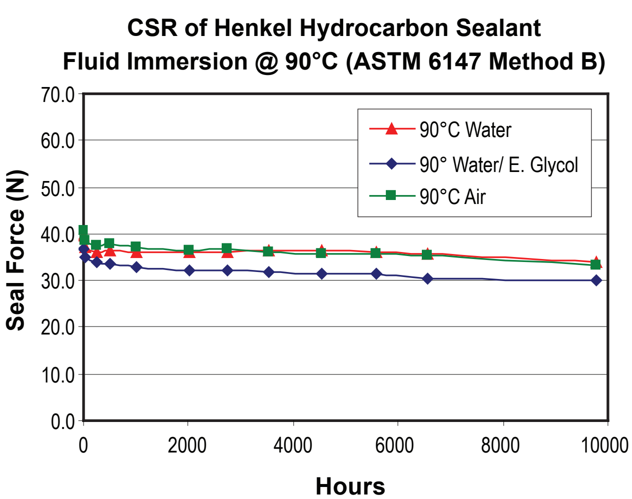

FIGURE 1. Results for CSR testing at 90°C in three aging media. Less than 20% loss in sealing force was observed in 10,000 hours for all cases.

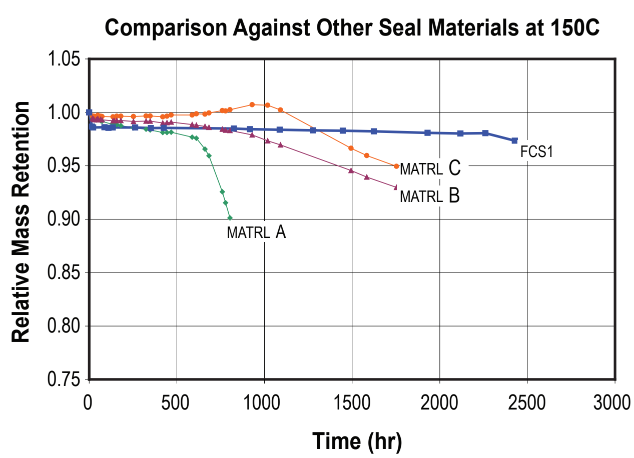

FIGURE 3. Comparison of EPDM and candidate material oxidative stability. The relative change in mass was tracked with time for three EPDM-based materials and FCS1, one of the developed candidate materials. The result indicates that FCS1 has higher oxidative stability.

rate of either weight gain or loss can be correlated to significant degradation in mechanical properties. The result indicates that the candidate material has greater oxidative stability than the EPDM-based materials tested. Several tests are also currently underway to compare the candidate materials to other materials such as silicone.

In addition to primary tasks discussed above, work on other tasks has also continued. Hundreds of sub-scale molded O-ring seals have been produced. The parts are seals over-molded around the perimeter of a disc of gas diffusion material. They are being used to determine appropriate molding parameters and to explore the role of geometry on load relaxation by comparison with results obtained from ASTM 6147 style samples. Some limited sub-scale molding was initiated and full-size mold flow analyses were conducted.

Conclusions and Future Directions

Based on the work performed to date for the project, the following can be concluded:

- The general material development strategy (synthesis and compounding) appears to be sound.
- Based on CSR, oxidation or other on-going testing, the materials developed have a high probability of success for meeting the 2010 DOE goals for use in automotive PEMFC applications.

Activities for the remainder of the project will include the following:

- Further development and evaluation of the next material candidate, FCS3.
- Molding and evaluation of seals over-molded around the perimeter of subscale membrane electrode assemblies (MEAs).
- Development of a high volume capable injection molding technique for over-molding seals around the perimeter of full-scale MEAs.
- Additional out-of-cell testing designed to validate test results obtained to date.
- Full-size in-cell testing to validate out-of-cell test results.

References

1. Brian James, Jeffrey Kalinoski, "Mass Production Cost Estimation of Automotive Fuel Cell Systems," DOE Hydrogen Program 2009 Annual Merit Review Proceedings, Washington, DC, 2009.

2. Jayanti Sinha, Stephen Lasher, Yong Yang, "Direct Hydrogen PEMFC Manufacturing Cost Estimation for Automotive Applications," DOE Hydrogen Program Annual Merit Review Proceedings, Washington, D.C., 2009.