Fuel cell vehicles (FCVs) using polymer electrolyte fuel cells (PEFCs) have been attracting much attention due to their low levels of exhaust emissions and the general transition from gasoline-fueled vehicles to electromotive vehicles. For further proliferation of FCVs, reductions of cost and increases in durability are required. Perfluorosulfonic acid (PFSA) membranes such as Nafion have been widely used as the electrolyte membrane for PEFCs due to their high proton conductivity and mechanical stability. However, their fully fluorinated chemical structure leads to high cost, low glass transition temperature (Tg), and a high gas permeability through the membrane, which are manufactured by DuPont and Gore, respectively.

In our previous study,32 we investigated a sulfonated benzophenone poly(arylene ether ketone) semiblock copolymer (SPK-bl-1, SPK)33 and a phenylene poly(arylene ether ketone) semiblock copolymer (SPP-bl-1, SPP)34 membrane under accelerated open circuit-voltage (OCV) conditions appropriate for FCVs. The results indicate that a simple hydrophilic structure that does not include ketone group leads to increased chemical stability versus radical attack decomposition. On the other hand, an SPP membrane quickly decomposed when subjected to a Fenton's reagent test, which may be attributed to the presence of heteroatoms in the hydrophobic structure. Based on these results, we developed a new hydrocarbon membrane that consists solely of phenylene groups, which is designated a sulfonated poly(phenylene) quinquephenylene (SPP-QP, Figure 1).35

In our previous study,32 we investigated a sulfonated benzophenone poly(arylene ether ketone) semiblock copolymer (SPK-bl-1, SPK)33 and a phenylene poly(arylene ether ketone) semiblock copolymer (SPP-bl-1, SPP)34 membrane under accelerated open circuit-voltage (OCV) conditions appropriate for FCVs. The results indicate that a simple hydrophilic structure that does not include ketone group leads to increased chemical stability versus radical attack decomposition. On the other hand, an SPP membrane quickly decomposed when subjected to a Fenton’s reagent test, which may be attributed to the presence of heteroatoms in the hydrophobic structure. Based on these results, we developed a new hydrocarbon membrane that consists solely of phenylene groups, which is designated a sulfonated poly(phenylene) quinquephenylene (SPP-QP, Figure 1).35

Durability of Newly Developed Polyphenylene-Based Ionomer Membranes in Polymer Electrolyte Fuel Cells: Accelerated Stress Evaluation

Ryo Shimizu,1 Kanji Otsuji,1 Akihiro Masuda,2 Nobuyuki Sato,2 Masato Kusakabe,3 Akihiro Iiyama,4,∗ Kenji Miyatake,4,∗ and Makoto Uchida 4,∗

1 Interdisciplinary Graduate School of Medicine and Engineering, University of Yamanashi, Kofu 400-8511, Japan
2 Toyosem Research Center, Inc., Otsu 520-8567, Japan
3 Kaneka Corporation, Settsu 566-0072, Japan
4 Fuel Cell Nanomaterials Center, University of Yamanashi, Kofu 400-0021, Japan
5 Clean Energy Research Center, University of Yamanashi, Kofu 400-8510, Japan

The chemical durability of the hydrocarbon (HC) polymer electrolyte membrane, sulfonated poly(phenylene) quinquephenylene (SPP-QP), is evaluated at 90°C and 160 kPaG pressure of hydrogen and air supplying anode and cathode, respectively, under open circuit voltage (OCV) conditions as an accelerated stress test in a polymer electrolyte fuel cell (PEFC). To evaluate the degree of deterioration of the membranes, exhaust water is collected from both electrodes periodically during the tests, and the chemical species are analyzed by ion chromatography (IC). The SPP-QP membrane-based cell with appropriate gaskets and gas diffusion layers (GDLs) shows the highest durability, in comparison with several other cells, and exhibits a high OCV for more than 1000 h and the lowest emission rate of sulfate (30 μg cm⁻² 2.6% loss) accumulated over 1000 h. We conclude that the simple hydrophilic structure and hydrophobic structure of the SPP-QP membrane, consisting solely of phenylene groups, leads to remarkably high intrinsic chemical stability and stability during the OCV stress evaluation; however, it also makes the membranes brittle. We also suggest that this negative aspect of the SPP-QP membrane might be mitigated by use of appropriate cell components such as gaskets and GDLs.

© The Author(s) 2019. Published by ECS. This is an open access article distributed under the terms of the Creative Commons Attribution 4.0 License (CC BY, http://creativecommons.org/licenses/by/4.0/), which permits unrestricted reuse of the work in any medium, provided the original work is properly cited. [DOI: 10.1149/2.0131907jes]
In the present research, we focus on the durability of the SPP-QP membrane, in comparison with previous results for HC membranes (SPP and SPK) under identical accelerated OCV conditions. During the durability evaluations, the exhaust water from both sides of the cells was collected and then analyzed by ion chromatography (IC). After equilibrating the membrane samples for at least 3 h under 80°C and 60% RH conditions, the stress-strain curves were obtained at a stretching rate of 10 mm min⁻¹. For the gasket samples, the curves were obtained at room temperature and humidity conditions (25°C, ca. 30% RH).

Fuel cell operation.—In this study, we selected the same accelerated OCV stress-evaluation protocol that was provided by Honda R&D Co., Ltd., as described in a previous report. Hydrogen (76% RH, anode) and air (86% RH, cathode) were supplied at 90°C and 160 kPaG pressure. The flow rates were 200 mL min⁻¹ for both electrodes. During the tests, exhaust water from both electrodes was collected.

Measurement of cell performance parameters during operation.—At the initial point and every 200 h during durability testing, cyclic voltammetry (CV) was carried out with a potentiostat (PGST30 Autolab System, Eco-Chemie) to evaluate the

| Table I. Initial membrane properties. |
| Membrane | IEC (meq g⁻¹) | Thickness (µm) | Sulfonic acid conc. (µg cm⁻²) |
|----------|--------------|---------------|-------------------------------|
| SPP-QP   | 2.37         | 25            | 1.15 × 10⁴                   |
| SPP      | 2.39         | 25            | 1.15 × 10⁴                   |
| SPK      | 2.40         | 25            | 1.16 × 10⁴                   |

*Initial sulfonic acid concentration as a reference for the sulfate concentrations detected in the exhaust water after durability testing.

Membrane and gasket stress-strain curves.—Tensile tests of the membranes and gaskets were conducted with a Shimadzu AGS-J 500N universal test machine equipped with a temperature and humidity-controllable chamber. The tests followed the DIN-53504-S3 standard (dumbbell shape of 35 × 6 mm (total) and 12 × 2 mm (test area)). After equilibrating the membrane samples for at least 3 h under 80°C and 60% RH conditions, the stress-strain curves were obtained at a stretching rate of 10 mm min⁻¹. For the gasket samples, the curves were obtained at room temperature and humidity conditions (25°C, ca. 30% RH).

Fuel cell operation.—In this study, we selected the same accelerated OCV stress-evaluation protocol that was provided by Honda R&D Co., Ltd., as described in a previous report. Hydrogen (76% RH, anode) and air (86% RH, cathode) were supplied at 90°C and 160 kPaG pressure. The flow rates were 200 mL min⁻¹ for both electrodes. During the tests, exhaust water from both electrodes was collected.

Measurement of cell performance parameters during operation.—At the initial point and every 200 h during durability testing, cyclic voltammetry (CV) was carried out with a potentiostat (PGST30 Autolab System, Eco-Chemie) to evaluate the

| Table II. MEA configurations for each type of cell. |
| Cell type | Type 1 | Type 2 | Type 3 |
|-----------|--------|--------|--------|
| GDL       | SGL 25BCH | Soft GDL | SGL 25BCH |
| Gasket    | Si/PEN/Si | Si/PEN/Si | PTFE   |

universal test machine equipped with a temperature and humidity-controllable chamber. The tests followed the DIN-53504-S3 standard (dumbbell shape of 35 × 6 mm (total) and 12 × 2 mm (test area)). After equilibrating the membrane samples for at least 3 h under 80°C and 60% RH conditions, the stress-strain curves were obtained at a stretching rate of 10 mm min⁻¹. For the gasket samples, the curves were obtained at room temperature and humidity conditions (25°C, ca. 30% RH).

Fuel cell operation.—In this study, we selected the same accelerated OCV stress-evaluation protocol that was provided by Honda R&D Co., Ltd., as described in a previous report. Hydrogen (76% RH, anode) and air (86% RH, cathode) were supplied at 90°C and 160 kPaG pressure. The flow rates were 200 mL min⁻¹ for both electrodes. During the tests, exhaust water from both electrodes was collected.

Measurement of cell performance parameters during operation.—At the initial point and every 200 h during durability testing, cyclic voltammetry (CV) was carried out with a potentiostat (PGST30 Autolab System, Eco-Chemie) to evaluate the

| Table II. MEA configurations for each type of cell. |
| Cell type | Type 1 | Type 2 | Type 3 |
|-----------|--------|--------|--------|
| GDL       | SGL 25BCH | Soft GDL | SGL 25BCH |
| Gasket    | Si/PEN/Si | Si/PEN/Si | PTFE   |
electrochemical surface area (ECA) of Pt and peak shifts in the high potential region induced by adsorbates from the decomposition of the membranes on Pt in the cathode catalyst layer at 90°C and ambient pressure. Prior to the measurements, hydrogen (100 mL min⁻¹) and nitrogen (150 mL min⁻¹) humidified at 76% RH and 86% RH were supplied to the anode and cathode, respectively. The cathode potential was scanned between 0.06 and 1.0 V at 20 mV s⁻¹, with a H₂ flow rate of 50 mL min⁻¹ at the anode and the cathode N₂ flow halted just prior to the measurement. The ECA values were determined from the hydrogen adsorption charge in the negative-going potential scan, referenced to ΔQHgc = 210 μC cm⁻², which is a standard value for polycrystalline platinum. The crossover hydrogen from the anode to the cathode was measured by use of linear sweep voltammetry (LSV) at 90°C and ambient pressure in a cell purged with hydrogen (100 mL min⁻¹) at the anode and nitrogen (150 mL min⁻¹) humidified at 76% RH and 86% RH at the cathode. The cell potential was swept from 0.15 to 0.6 V at a sweep rate of 0.5 mV s⁻¹. The current-potential (I-E) polarization curves of the MEAs were measured by supplying hydrogen and oxygen/air at 90°C and ambient pressure. The flow rates of all gases were controlled by mass flow controllers. The utilisations of the reactant gases were 70% for hydrogen and 40% for oxygen/air. Hydrogen gas and oxygen/air were pre-humidified at 76% RH and 86% RH respectively (Figure 2a). The emitted concentration values of sulfate decreased in the order SPP > SPP-QP type 1 > SPP-QP type 2 ≈ SPP-QP type 3. This was thought to be attributable to the intrinsic chemical stabilities of the membranes themselves. To check these, the intrinsic chemical stabilities of the SPP-QP, SPP, and SPK membranes were evaluated with Fenton’s reagent. For this test, membranes samples with dimensions of 5 cm × 5 cm × 25 μm were used, with a mass of ca. 100 mg; these were immersed in an aqueous solution containing 3% H₂O₂ and 2 parts per million (ppm) Fe²⁺ at 80°C for 1 hour. The residual Mₘₜ and Mₚ values were 99% and 99% for SPP-QP, 43% and 35% for SPP, and only 11% and 10% for SPK, respectively, and ion exchange capacity (IEC) of SPP-QP hardly changed after test (Table III). These results indicated that the SPP-QP membranes had much higher intrinsic chemical stability compared to those of SPP and SPK membranes, which was reflected in the results of the accelerated OCV stress evaluation.

CVs were measured initially and every 200 h for all cells (Figures 3a–3e). The Pt surface oxidation/reduction peaks changed remarkably with time in the case of the SPK cell: the onset potentials of the oxidation peaks moved from 0.6 V to 0.8 V, and large oxidation current over 0.9 V increased slightly (Figure 3b). In contrast, the CV peaks for the SPP-QP cells hardly changed over 1000 h (Figure 2a). The hydrogen leak current density and the composition of the water that was drained from each cell were measured every 200 h by use of LSV and IC, respectively, as shown in Figures 2a–2c. The cell voltage for the SPP-QP type 1 cell decreased significantly after 700 h. In the case of SPP-QP type 2 and type 3 cells, the cell voltages maintained high values for over 1000 h, as those for the SPP cell and SPK cell (Figure 2a). The hydrogen leak current density also behaved nearly the same for each of the cells, except for the SPP-QP type 1 cell, which reached ca. 100 times higher H₂ leakage at 800 h than those of the others (Figure 2b). The amount of emitted sulfate ions for the SPP-QP type 1 cell reached 59 μg cm⁻² (geometric area 29.2 cm², 5.1% loss) at 800 h, which was lower than those of the SPP and SPK cells. Additionally, those for the SPP-QP type 2 and type 3 cells were 28 and 32 μg cm⁻² (2.4% and 2.8% loss) respectively (Figure 2c). The emitted concentration values of sulfate decreased in the order SPP > SPP-QP type 1 > SPP-QP type 2 ≈ SPP-QP type 3. We consider this to be attributable to the intrinsic chemical stabilities of the membranes themselves. To check these, the intrinsic chemical stabilities of the SPP-QP, SPP and SPK membranes were evaluated with Fenton’s reagent. For this test, membranes samples with dimensions of 5 cm × 5 cm × 25 μm were used, with a mass of 100 mg; these were immersed in an aqueous solution containing 3% H₂O₂ and 2 parts per million (ppm) Fe²⁺ at 80°C for 1 hour. The residual Mₘₜ and Mₚ values were 99% and 99% for SPP-QP, 43% and 35% for SPP, and only 11% and 10% for SPK, respectively, and ion exchange capacity (IEC) of SPP-QP hardly changed after test (Table III). These results indicated that the SPP-QP membranes had much higher intrinsic chemical stability compared to those of SPP and SPK membranes, which was reflected in the results of the accelerated OCV stress evaluation.

CVs were measured initially and every 200 h for all cells (Figures 3a–3e). The Pt surface oxidation/reduction peaks changed remarkably with time in the case of the SPK cell: the onset potentials of the oxidation peaks moved from 0.6 V to 0.8 V, and large oxidation current over 0.9 V increased slightly (Figure 3b). In contrast, the CV peaks for the SPP-QP cells hardly changed over 1000 h for the full potential range (Figures 3d, 3e). Even the SPP-QP type 1 cell maintained a similar shape for the CV at 600 h (Figure 3c). The CVs after 800 h for the SPP-QP type 1 cell were not obtainable due to large H₂ cross-over. Additionally, after each performance evaluation, we found that the CVs for the SPK and SPP membranes recovered to nearly the same shapes as those of the initial CVs. We consider the reason to be as follows. Our previous research reported that the changes that occurred in the CVs could mainly be attributed to the adsorption of membrane degradation products on the catalyst; these products accumulated during the OCV durability test and were then washed out from the MEA with the water that was produced during the interim performance evaluation.²

Therefore, the results from the CV measurements also indicate that the decomposition of the SPP-QP membrane was much milder than that of the SPP membrane and by far milder than that of the SPK membrane, because the amounts of adsorbates in the SPP-QP cell were smaller than those in the SPP and SPK cells. Hence, we conclude that the durability of the membranes increased in the order SPK < SPP < SPP-QP, according to the single cell evaluation.

### Results and Discussion

**Durability of the cell under accelerated OCV stress evaluation.**

The cell voltages were continuously measured during the accelerated OCV stress evaluation, and the hydrogen leak current density and the composition of the water that was drained from each cell were measured every 200 h by use of LSV and IC, respectively, as shown in Figures 2a–2c. The cell voltage for the SPP-QP type 1 cell decreased significantly after 700 h. In the case of SPP-QP type 2 and type 3 cells, the cell voltages maintained high values for over 1000 h, as those for the SPP cell and SPK cell (Figure 2a). The hydrogen leak current density also behaved nearly the same for each of the cells, except for the SPP-QP type 1 cell, which reached ca. 100 times higher H₂ leakage at 800 h than those of the others (Figure 2b). The amount of emitted sulfate ions for the SPP-QP type 1 cell reached 59 μg cm⁻² (geometric area 29.2 cm², 5.1% loss) at 800 h, which was lower than those of the SPP and SPK cells. Additionally, those for the SPP-QP type 2 and type 3 cells were 28 and 32 μg cm⁻² (2.4% and 2.8% loss) respectively (Figure 2c). The emitted concentration values of sulfate decreased in the order SPP > SPP-QP type 1 > SPP-QP type 2 ≈ SPP-QP type 3. This was thought to be attributable to the intrinsic chemical stabilities of the membranes themselves. To check these, the intrinsic chemical stabilities of the SPP-QP, SPP, and SPK membranes were evaluated with Fenton’s reagent. For this test, membranes samples with dimensions of 5 cm × 5 cm × 25 μm were used, with a mass of ca. 100 mg; these were immersed in an aqueous solution containing 3% H₂O₂ and 2 parts per million (ppm) Fe²⁺ at 80°C for 1 hour. The residual Mₘₜ and Mₚ values were 99% and 99% for SPP-QP, 43% and 35% for SPP, and only 11% and 10% for SPK, respectively, and ion exchange capacity (IEC) of SPP-QP hardly changed after test (Table III). These results indicated that the SPP-QP membranes had much higher intrinsic chemical stability compared to those of SPP and SPK membranes, which was reflected in the results of the accelerated OCV stress evaluation.

CVs were measured initially and every 200 h for all cells (Figures 3a–3e). The Pt surface oxidation/reduction peaks changed remarkably with time in the case of the SPK cell: the onset potentials of the oxidation peaks moved from 0.6 V to 0.8 V, and large oxidation current over 0.9 V increased slightly (Figure 3b). In contrast, the CV peaks for the SPP-QP cells hardly changed over 1000 h for the full potential range (Figures 3d, 3e). Even the SPP-QP type 1 cell maintained a similar shape for the CV at 600 h (Figure 3c). The CVs after 800 h for the SPP-QP type 1 cell were not obtainable due to large H₂ cross-over. Additionally, after each performance evaluation, we found that the CVs for the SPK and SPP membranes recovered to nearly the same shapes as those of the initial CVs. We consider the reason to be as follows. Our previous research reported that the changes that occurred in the CVs could mainly be attributed to the adsorption of membrane degradation products on the catalyst; these products accumulated during the OCV durability test and were then washed out from the MEA with the water that was produced during the interim performance evaluation.²

Therefore, the results from the CV measurements also indicate that the decomposition of the SPP-QP membrane was much milder than that of the SPP membrane and by far milder than that of the SPK membrane, because the amounts of adsorbates in the SPP-QP cell were smaller than those in the SPP and SPK cells. Hence, we conclude that the durability of the membranes increased in the order SPK < SPP < SPP-QP, according to the single cell evaluation.
Figure 3. Cyclic voltammograms (CVs), sweep rate 20 mV s\(^{-1}\), for the cathode catalyst layer for (a) the SPK cell and (b) SPP cell (c-e) SPP-QP types 1, 2, and 3 cells operated with H\(_2\)-N\(_2\), humidified at 76% RH and 86% RH at \(T_{\text{cell}} = 90^\circ\text{C}\), the cathode N\(_2\)-flow halted just prior to the measurement. Current normalized to unit mass of Pt. The loading amount of Pt was 0.5 mg cm\(^{-2}\).

After the accelerated OCV stress evaluation, the cells were disassembled and checked by visual observation. We were able to observe a pinhole at the edge of the CL for the SPP-QP membrane using the type 1 cell (red circle in bottom right corner), as shown in Figure 4a. The helium leakage for the SPP-QP type 1 cell also showed a high leakage rate in the edge region of the CCM after the test (Figure 4b). Ishikawa et al. have reported that the stress was much higher at edge sites of CCMs due to the configuration of MEAs and cells, in which there is a large degree of membrane shrinkage and swelling, in comparison with other sites.\(^{37}\) They also reported that the use of soft GDLs was able to decrease the mechanical stress from fastening the cells, which improved the mechanical strength of the membranes in the cells\(^{37}\) (Figure 5).

From the viewpoint of the membranes themselves, the stress-strain curves differed significantly between the SPP-QP and SPP, SPK membranes (Figure 6a). SPP-QP exhibited higher stress strength, but, on the other hand, for strain aspect, it was much lower in comparison with the other membranes, which means that the SPP-QP membrane is more brittle. We consider that this difference is quite significant for the mechanical stability in the cells.

Since the soft GDL was chosen as an MEA component to reduce the stress at edge sites, we also considered modifying the gas-gaskets. Figure 6b shows stress-strain curves for a PTFE gasket and a Si/PEN/Si gasket, which exhibited very different Young’s moduli (0.3 GPa, 2.0 GPa, respectively) and elongation (strain at break point...
= 360%, 33%, respectively). These values indicate that the PTFE gasket is much softer and easier to stretch than the Si/PEN/Si gasket. Thus, we selected the PTFE gasket in place of the Si/PEN/Si gasket with the SGL GDL and expected that it would bring about nearly same effect as that of the soft GDL.

In order to examine for these effects, we constructed a new type of cell, designated a type 2 cell, using the soft GDL with the Si/PEN/Si gasket, and a type 3 cell, using the PTFE gasket with the SGL GDL, as listed in Table II.

During accelerated OCV stress evaluation, the voltages (Figure 2a), the amounts of emitted sulfate ion from the IC (Figure 2c), and the shapes of the CVs (Figures 3d, 3e) all indicated that both types 2 and 3 cells using SPP-QP possessed higher stability than those of others. The voltages were over 0.9 V during the evaluation period, the sulfate emissions were about one-third as high as that of the SPP/QP cell and one tenth as high as that of the SPK cell, and, significantly, the shapes of the CVs hardly changed. These results indicate high chemical stability for the SPP-QP membrane in the cells during accelerated OCV stress evaluation. The effect of the soft GDL in the cells decreased the mechanical stress during shrinkage and swelling of the membrane. The PTFE gasket also brought about decreased mechanical stress in the cells. After the durability evaluation, the PTFE gasket was able to peel away easily from the membrane, while the Si/PEN/Si gasket had adhered strongly to the membrane. In the case of the PTFE gasket, the mechanical stress might have decreased by slippage of the membrane. We also consider that there is a need to investigate the effect of the PTFE gasket on the mitigation of mechanical stress in more detail in future work.

| Membrane               | x: y (%) | IEC (meq g\(^{-1}\)) |
|------------------------|----------|----------------------|
| SPP-QP pristine        | 34:66    | 2.9                  |
| Type1 cell after tests | 34:66    | 2.9                  |
| Type2 cell after tests | 33:67    | 2.9                  |
| Type3 cell after tests | 33:67    | 2.9                  |

**Degradation site analysis of the molecular structure**—These SPP-QP membranes used for all cell types were analyzed by NMR in order to clarify the molecular structural changes before and after the accelerated OCV stress evaluation. Figure 7 shows the initial NMR spectra of the SPP-QP membrane and those after the stress evaluation. The NMR spectral peaks for the SPP-QP membranes for all cells were hardly changed before and after the stress evaluation. The peak-area ratios of the hydrophilic moiety and hydrophobic moieties, and the IEC values calculated from them, were also hardly changed (Table IV). The initial ratio of the integrated areas for hydrophilic to hydrophobic moieties and IEC values for SPP and SPK membranes that were decreased significantly after the stress evaluation. From these NMR analyses, the chemical structure of the SPP-QP membrane was found to have a higher stability versus the accelerated OCV stress evaluation in comparison with those for the SPP and SPK membranes. The water-soluble oxidative degradation by-products were washed away during cell operation. In the IC results, sulfate ion was detected in the SPP-QP membrane cell, which means that the degradation of the SPP-QP membrane occurred to a measurable extent, but it was lower than that of the other HC membranes. The difference might be attributed to the simple hydrophilic structure and hydrophobic structure, made up by phenylene groups in the SPP-QP membrane whose chemical structure hardly underwent radical attack, resulting in a membrane that saw less chemical decomposition.

**Conclusions**

Our newly developed polyphenylene-based HC membrane, SPP-QP, was evaluated under accelerated stress conditions at OCV. The results for the cell using the SPP-QP membrane with soft GDL, type 2, and PTFE gaskets, type 3, showed the highest durability in comparison with other cells and exhibited high OCV values for more than 1000 h and the lowest emission rate of sulfate emission (30 \(\mu\)g cm\(^{-2}\) h\(^{-1}\), 2.6% loss) accumulated over 1000 h. On the other hand, with the normal MEA configuration, type 1, the SPP-QP membrane was ruptured at an edge site after 800 h in accelerated OCV evaluation due to the brittleness of the SPP-QP membrane. The simple hydrophilic and hydrophobic structure, made up solely of phenylene groups in the SPP-QP membrane, led to remarkably high chemical stability; however, it also made the membrane brittle. We conclude that the SPP-QP membrane has high intrinsic durability, based on the OCV stress evaluation, in comparison with the other HC membranes. The next stage of new development for HC membranes will require more flexible structures while maintaining the high chemical stability of the SPP-QP membrane. We also suggest that this negative aspect of the SPP-QP membrane is able to be mitigated by the use of modified cell components such as appropriate gaskets and GDLs.

**Acknowledgments**

This work was partially supported by funds for the “Superlative, Stable, and Scalable Performance Fuel Cell (SPer-FC)" Project from the New Energy and Industrial Technology Development
Organization (NEDO) of Japan, and the Ministry of Education, Culture, Sports, Science and Technology (MEXT) Japan through a Grant-in-Aid for Scientific Research (18H02030, 18H05515, 18K19111). The authors are grateful to Panasonic Corporation for kindly providing the experimental GDL as soft GDL. Also, the authors are grateful to Honda R&D Co., Ltd. for kindly providing the accelerated OCV stress-evaluation protocol.

ORCID
Kenji Miyatake https://orcid.org/0000-0001-5713-2635
Makoto Uchida https://orcid.org/0000-0002-7847-3727

References
1. R. Borup, Chem. Rev., 107, 3904 (2007).
2. A. Kusoglu and A. Z. Weber, Chem. Rev., 117, 987 (2017).
3. M. P. Rodgers, L. J. Bonville, H. R. Kunz, D. K. Slattery, and J. M. Fenton, Chem. Rev., 112, 6075 (2012).
4. United States Department of Energy, Multi-Year Research, Development, and Demonstration Plan, Fuel Cells, 2016.
5. S. Shi, A. Z. Weber, and A. Kusoglu, Journal of Membrane Science, 516, 123 (2016).
6. R. Mukundan, A. M. Baker, A. Kusoglu, P. Beattie, S. Knights, A. Z. Weber, and R. L. Borup, J. Electrochem. Soc., 165, F3085 (2018).
7. M. Robert, A. E. Kaddouri, J.-C. Perrin, S. Leclere, and O. Lottin, J. Electrochem. Soc., 165, F3209 (2018).
8. Z. Zhang, S. Shi, Q. Lin, L. Wang, Z. Liu, P. Li, and X. Chen, Int. J. Hydrogen Energy, 43, 6579 (2018).
9. DuPont. Product information: Nafion XL PFSA membrane, (2010).
10. W. Liu, T. Suzuki, H. Mao, and T. Schmiedel, ECS Transactions, 50, 51 (2013).
11. E. Endoh, ECS Transactions, 16, 1229 (2008).
12. B. P. Pearman, N. Mohajeri, R. P. Brooker, M. P. Rodgers, D. K. Slattery, M. D. Hampton, D. A. Cullen, and S. Seal, J. Power Sources, 225, 75 (2013).
13. A. M. Baker, R. Mukundan, D. Spennaj, E. J. Judge, S. G. Advani, A. K. Prasad, and R. L. Borup, J. Electrochem. Soc., 163, F1023 (2016).
14. T. Weissbach, T. J. Peckham, and S. Holdcroft, Journal of Membrane Science, 498, 94 (2016).
15. L. Gubler, T. Nauser, F. D. Coms, Y.-H. Lai, and C. S. Gittleman, J. Electrochem. Soc., 165, F3100 (2018).
16. A. Roy, X. Yu, S. Dunn, and J. E. McGrath, Journal of Membrane Science, 327, 118 (2009).
17. B. Bae, T. Hoshi, K. Miyatake, and M. Watanabe, Macromolecules, 44, 3884 (2011).
18. Y. A. Elabd and M. A. Hickner, Macromolecules, 44, 1 (2011).
19. S. Takamatsu and P. Jannasch, Macromolecules, 45, 6538 (2012).
20. M. Adamski, T. J. G. Skalski, B. Britton, T. J. Peckham, L. Metzler, and S. Holdcroft, Angew. Chem., 129, 9186 (2017).
21. C. Perrot, L. Gonon, M. Bardet, C. Marest, A. Pierre-Baye, and G. Gebel, 50, 1671 (2009).
22. K. Si, R. Wycisk, D. Dong, K. Cooper, M. Rodgers, P. Brooker, D. Slattery, and M. Litt, Macromolecules, 46, 422 (2013).
23. T. Ko, K. Kim, B.-K. Jung, S.-H. Cha, S.-K. Kim, and J.-C. Lee, Macromolecules, 48, 1104 (2015).
24. T. Weissbach, A. G. Wright, T. J. Peckham, A. Sadeghi Alavijeh, V. Pan, E. Kjeang, and S. Holdcroft, Chem. Mater., 28, 8060 (2016).
25. J. Miyake, M. Kusakabe, A. Tsutsui, and K. Miyatake, ACS Applied Energy Materials, 1, 1233 (2018).
26. K. Miyatake, T. Yuda, and M. Watanabe, J. Polym. Sci., Part A: Polym. Chem., 46, 4469 (2008).
27. J. Saito, K. Miyatake, and M. Watanabe, Macromolecules, 41, 2415 (2008).
28. B. Bae, S. Kawamura, K. Miyatake, and M. Watanabe, Macromolecules, 49, 3863 (2016).
29. K. Miyatake, B. Bae, and M. Watanabe, Polymer Chemistry, 2, 1919 (2011).
30. B. Bae, K. Miyatake, and M. Watanabe, Macromolecules, 43, 2684 (2010).
31. B. Bae, T. Yoda, K. Miyatake, M. Uchida, H. Uchida, and M. Watanabe, The Journal of Physical Chemistry B, 114, 10481 (2010).
32. R. Shimizu, J. Tsuji, N. Sato, J. Takano, S. Itami, M. Kusakabe, K. Miyatake, A. Iiyama, and M. Uchida, J. Power Sources, 367, 63 (2017).
33. T. Miyahara, T. Hayano, S. Matsuno, M. Watanabe, and K. Miyatake, ACS applied materials & interfaces, 4, 2881 (2012).
34. J. Miyake, T. Kuchizuki, and K. Miyatake, ACS Macro Letters, 4, 750 (2015).
35. J. Miyake, R. Taki, T. Kuchizuki, R. Shimizu, R. Akiyama, M. Uchida, and K. Miyatake, Science Advances, 3, eaa0476 (2017).
36. M. Yamauchi and Y. Tsuji, Membrane-electrode-assembly and fuel cell, US Pat., 0076,392 A1, (2011).
37. H. Ishikawa, T. Teramoto, Y. Ueyama, Y. Sugawara, Y. Sakiyama, M. Kusakabe, K. Miyatake, M. Uchida, J. Power Sources, 325, 35 (2016).
38. A. Kabasawa, J. Saito, K. Miyatake, H. Uchida, and M. Watanabe, Electrochim. Acta, 24, 2754 (2009).