

Development of High-durability and Low-cost UEA for Fuel Cell Stack

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ABSTRACT

Honda's newly structured fuel cell stack features a Unitized Electrode Assembly, which consists of a Membrane Electrode Assembly that generates electricity and an outer resin frame structure. Specifications of the Unitized Electrode Assembly were developed that have greater durability and lower cost than the previous fuel cell stack, while maintaining its output performance. Specifically, the amount of Pt was reduced to 1/5 that of the previous specifications due to increased activity by using a Pt alloy catalyst for the electrode catalyst, and the thickness of the electrolyte membrane was reduced to 3/5. For the outer resin structure, the cost was reduced to less than half that of the previous injection-molded product by using a general-purpose resin film.

As for the durability associated with the above-mentioned changes, the researchers developed degradation models for the various components of the Unitized Electrode Assembly and worked to increase the accuracy of those models. These efforts achieved both increased durability and lower costs by reflecting the material specification limit ranges in operation control based on the models obtained.

1. Introduction

Global warming has become so severe that in July 2023, UN Secretary-General António Guterres warned that the earth has entered the era of global boiling⁽¹⁾. At COP28, the 28th Conference of the Parties, in 2023, it became clear that humanity is falling short of achieving the 1.5°C target of the Paris Agreement adopted at COP21 in 2015⁽²⁾. Meanwhile, moves are underway to reduce and absorb greenhouse gas emissions, which are a factor in rising temperatures.

Hydrogen, in particular, is important to help achieve not only carbon neutrality but also energy resilience⁽³⁾.

Focusing on the potential of hydrogen energy, Honda began basic research on fuel cells in the late 1980s. Today, there is an urgent need to further increase durability and lower costs for a wide variety of applications, including not just passenger cars but also commercial vehicles and stationary power sources.

Against this background, Honda and General Motors have jointly developed a newly structured fuel cell stack that is more than twice as durable and has only 1/3 the cost

of the previous fuel cell stack, while maintaining its output performance⁽⁴⁾.

The Unitized Electrode Assembly (UEA) consists of a Membrane Electrode Assembly (MEA) that generates electricity and its outer resin frame structure. The previous UEA used a large amount of platinum (Pt) in the electrode catalyst and ionomer components, which are fluorinated polymer electrolytes that play a role in proton transport. These help to ensure durability, but also have a big impact on costs. Moreover, injection molding was used for the outer resin structure due to the configuration of the power generation cells. This requires a thickness of at least 250 μm to help ensure formability, and thus it was not easy to increase the power density by making the cells thinner.

2. Development Goals

The structure of the UEA in the previous stack is shown in Fig. 1. The power generation cell consists of two UEAs sandwiched within three stainless steel (SUS) separators⁽⁵⁾. A stack consists of several hundred of these power generation cells stacked on top of each other. The

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UEA consists of the MEA and an outer resin structure. The material cost breakdown of the previous UEA is shown in Fig. 2. As Fig. 2 shows, the cathode catalyst and electrolyte membrane make up a high percentage of the cost. Pt in the electrode catalyst, and ionomer, the main component of the electrolyte membrane, are expensive and not effective to mass-produce, and thus reducing use of these components is an effective way to lower the cost of the UEA.

Meanwhile, the gas diffusion layer (GDL) has the third greatest impact on cost, and in this case mass production seemed likely to reduce costs. Therefore, the researchers focused instead on simplifying the structure of the outer resin structure, which has the fourth greatest impact on cost. The outer resin structure can increase the power density.

Therefore, the goal of this development was to achieve the following while maintaining the same or greater output

performance and durability of the previous fuel cell stack.

- (1) Reduce the amount of Pt in the electrode catalyst
- (2) Reduce the amount of ionomer in the electrolyte membrane
- (3) Simplify the structure of the outer resin structure

In addition, the durability of the MEA is inversely related to the reduction in the amount of Pt and ionomer used⁽⁶⁾⁻⁽⁷⁾, and thus ensuring durability while reducing costs is a challenge. For each UEA component, a degradation model was developed that can quantitatively represent the impact on durability and material degradation factors. Based on these models, controlling the operating environment so it is appropriate for each material's specification limits helped to ensure durability.

The structure of the UEA of the newly structured stack is shown in Fig. 3. The power generation cell consists of a single UEA sandwiched between two bipolar plates (BPPs)⁽⁴⁾. The technology that increased the UEA's durability and reduced its costs is discussed below.

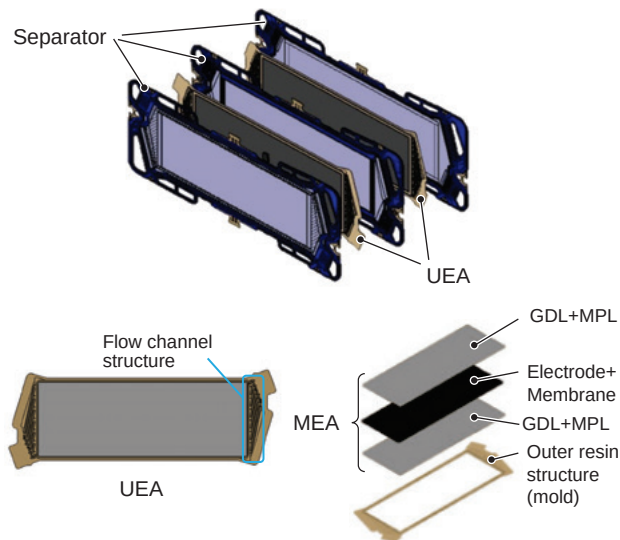


Fig. 1 Structure of previous power generation cell and UEA

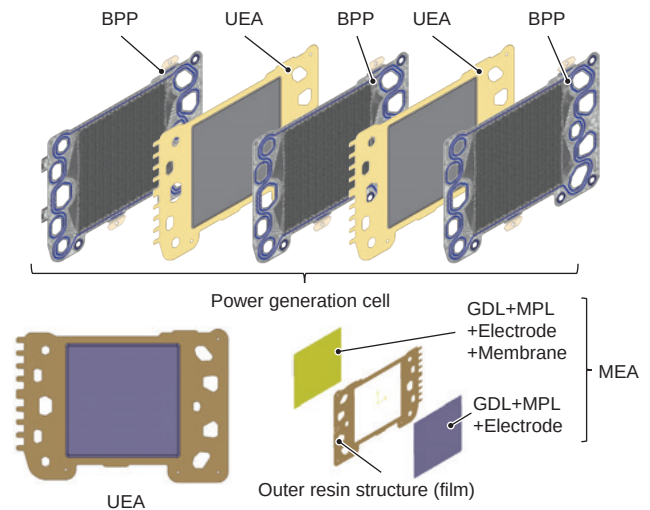


Fig. 3 Structure of new power generation cell and UEA

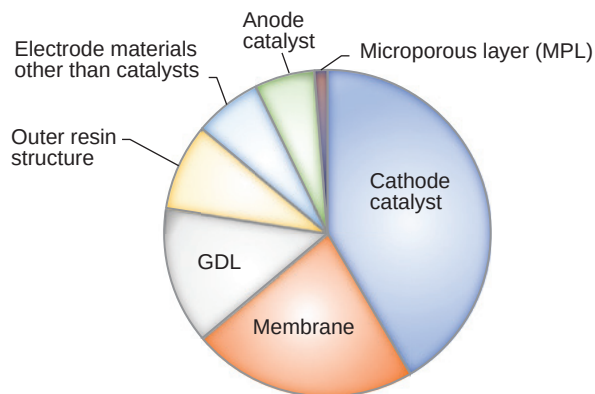


Fig. 2 Material cost breakdown in previous UEA

3. Reducing the Amount of Pt in the Electrode Catalyst

3.1. Pt Alloy Catalyst

Pt, a precious metal, is used in electrode catalysts. Therefore, reducing the amount of Pt is an effective way to reduce costs. However, reducing the amount of Pt also reduces the effective Pt surface area for generating power, resulting in lower power generation performance. Power generation performance is determined by multiplying the effective Pt surface area for generating power by the catalytic activity⁽⁸⁾. For that reason, a Pt alloy catalyst with high catalytic activity was adopted to increase activity as

compared to the Pt catalyst used in the previous UEA. The effect of using Pt alloy catalyst to increase performance is shown in Fig. 4. Porous carbon was used to support the Pt alloy, and Pt alloy particles were also supported in the carbon pores where ionomer could not penetrate. This suppresses catalyst poisoning by the ionomer⁽⁹⁾, while the use of Pt alloy increases the efficiency of the oxygen reduction reaction⁽¹⁰⁾, and as a result, activity is increased to more than double that of the previous Pt catalyst. The Pt alloy catalyst achieved the same catalytic activity as the previous catalyst, and thus the Pt surface area needed to obtain the same cell voltage as the Pt catalyst was reduced to about 1/4 that of the previous catalyst. In addition, to help ensure output, the voltage drop that may occur when the current density is increased needs to be suppressed. To suppress the voltage drop, the Pt alloy supporting rate was reduced to 3/5, which helped to ensure the oxygen reduction reaction at high current density and achieve the target cell output. Furthermore, a degradation model was developed as described in the next section to control the operating environment appropriately and suppress performance degradation.

As a result of the above, the amount of Pt per stack was reduced to 1/5, while the same power generation performance as before was maintained.

3.2. Suppression of Electrode Catalyst Degradation

Reducing the amount of Pt has a great impact on the cell voltage drop due to the decrease in effective Pt surface area for generating power. This makes it necessary to suppress the decrease in effective Pt surface area for generating power due to electrode degradation.

The mechanism of electrode catalyst degradation is shown in Fig. 5. In the power generation range of a fuel cell (0.6 to 0.8 V), the Pt surface is oxidized or reduced due to voltage fluctuations caused by load changes, and the Pt surface area decreases due to Ostwald growth, in which Pt repeatedly elutes and precipitates. When the

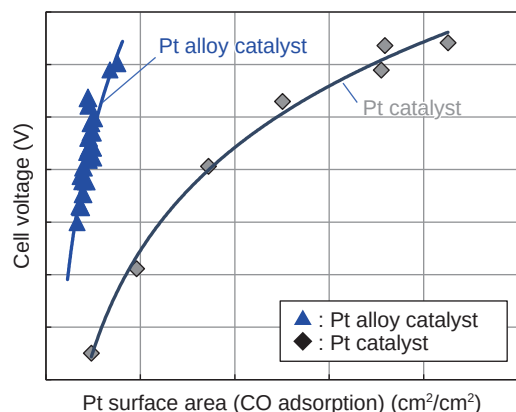


Fig. 4 Performance comparison of Pt catalyst and Pt alloy catalyst

voltage exceeds 0.85 V during startup or when power is being generated close to the open-circuit voltage, the amount of elution due to Pt oxidation increases. Furthermore, it is known that the presence of oxygen at the anode electrode during startup raises the cathode potential to about 1.5 V, which increases the Pt particle size and in addition degrades the carbon carrier⁽¹¹⁾. Therefore, controls were implemented to help prevent the anode electrode from having an air environment during shutdown, which suppresses the rise in cathode potential. In addition, a degradation model was developed for the degradation that can happen during operation. Then, degradation was suppressed by setting certain operating conditions, such as lowering the temperature in conjunction with the electrolyte membrane as described below.

The electrode degradation model was developed by using a single cell with a power generation area of approximately 50 cm² and conducting durability tests using the experimental design method to confirm the degradation factors and their effect. Results of checking the impact of potential and hold time on degradation are shown in Fig. 6.

The effects of temperature and humidity were also confirmed by the experimental design method, similar to Fig. 6. A model consisting of temperature, humidity, and potential was developed as shown in Eq. (1). The higher the temperature, humidity, and potential, the greater the acceleration of degradation⁽¹²⁾.

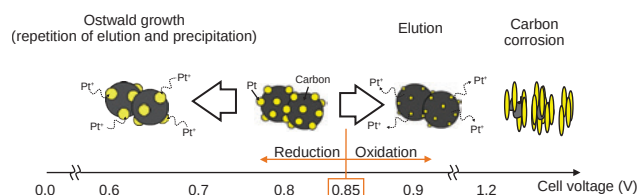


Fig. 5 Mechanism of electrode catalyst degradation

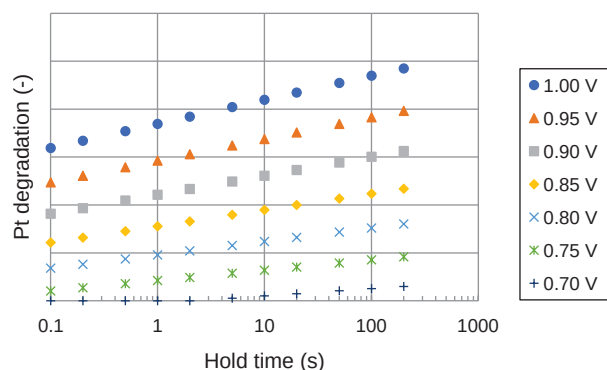


Fig. 6 Effect of potential and time on Pt degradation at 80°C

$$\theta_i = \frac{k_i(V, t) \times k_{\text{Temp}}(V, \text{Temp}) \times k_{\text{RH}}(V, \text{RH}) - r_i(V, t) \times r_{\text{Temp}}(V, \text{Temp}) \times r_{\text{RH}}(V, \text{RH})}{r_{\text{Temp}}(V, \text{Temp}) \times r_{\text{RH}}(V, \text{RH})} \quad (1)$$

θ_i : Amount of decrease of effective Pt surface area for generating power

t : Time

Temp : Temperature

RH : Humidity

V : Potential

k : Coefficient on high-voltage side

r : Coefficient on low-voltage side

Figure 7 compares the effective Pt surface area for generating power as calculated in Eq. (1) based on operating history during FC system durability testing with actual values measured after durability testing was completed. Although the target performance was achieved, the measured results showed degradation had progressed more than predicted by the model. This may be because the temperature used in the calculations from the operating history is the temperature of the cooling water. This is because, due to the heat generated by overvoltage, the temperature near the catalyst is higher than the cooling water temperature. Using this discrepancy between actual and predicted values as the performance yields a difference of approximately 5 kW, but it was confirmed that the necessary durability was achieved. In future, a further increase in the accuracy of predictions of remaining life will be necessary.

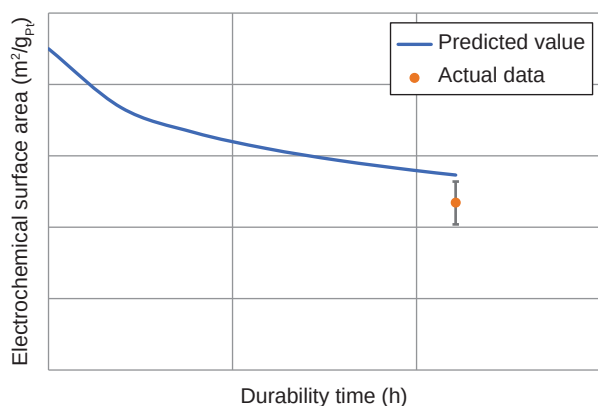


Fig. 7 Comparison of actual and predicted electrode degradation during FC system durability testing

4. Reducing the Amount of Ionomer in the Electrolyte Membrane

4.1. Thinner Electrolyte Membrane

Aside from reducing costs, a thinner electrolyte membrane would be an effective way to increase power generation performance by increasing proton conductivity

and to increase water transfer from the cathode to the anode, thereby helping to ensure membrane water retention and suppress flooding in the power generation cell. However, as the electrolyte membrane needs to function as a gas barrier between the cathode and anode, a thinner membrane increases the amount of gas permeation, which could reduce fuel economy. For that reason, after considering power generation performance and the impact to fuel consumption, the thickness of the electrolyte membrane was reduced to 3/5 that of the previous stack by addressing the chemical and mechanical degradation associated with thinner membranes and by employing a new proprietary MEA coating process and formulation.

4.2. Dealing with Chemical Degradation of Electrolyte Membranes

There is both chemical and mechanical degradation of electrolyte membranes⁽¹³⁾. The mechanism of chemical degradation of the electrolyte membrane is shown in Fig. 8. OH radicals are formed by a reaction of H_2O_2 , which is a byproduct of the power generation reaction, with Fe ions, etc. OH radicals can cause an unzipping reaction, in which the main chain ends of the electrolyte membrane are attacked, and main-chain scission, leading to a decline in the molecular weight. When main-chain scission occurs, the number of terminal groups increases, which promotes degradation at an accelerated rate⁽¹⁴⁾. Degradation is known to accelerate the higher the temperature and lower the humidification⁽¹⁵⁾. The formation of H_2O_2 , which is how degradation begins, is also thought to be greatly affected by the amount of oxygen that permeates from the cathode side to the anode side. A reduction in membrane thickness increases the amount of gas permeation, which may accelerate degradation.

From the degradation mechanism shown in Fig. 8, the effects of temperature and humidity, as well as current and O_2 partial pressure, were confirmed using the experimental

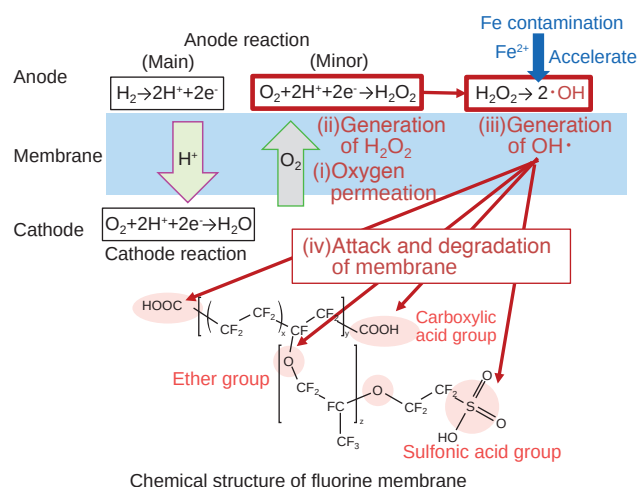


Fig. 8 Mechanism of membrane chemical degradation

design method in a single cell, similar to electrode degradation, and the electrolyte membrane degradation model shown in Eq. (2) was developed. Equation (2) shows the amount of chemical degradation stress, while Eq. (3) shows the relationship between the amount of stress and molecular weight retention rate. As with the electrodes, a degradation model was developed for durability, and the operating environment was controlled to help ensure it.

$$S_C = \exp(C_{Temp} \times Temp + C_{RH} \times RH + C_J \times J + C_{P_{O_2}} \times P_{O_2}) \times C_{P_{O_2}} \times P_{O_2} \quad (2)$$

$$Mw = \exp(C_{Sc} \times S_C \times t) + C_2 \quad (3)$$

- S_C : Chemical degradation stress
 Mw : Molecular weight retention rate
 t : Time
 $Temp$: Temperature
 RH : Humidity
 J : Current density
 P_{O_2} : O_2 partial pressure
 C : Coefficient for each degradation factor

As Eq. (2) shows, the degradation model consists of temperature, humidity, current density, and oxygen partial pressure. In particular, as low humidity accelerates membrane degradation all at once, operation is controlled so that the humidity input to the stack is kept above a certain level to help ensure durability⁽¹⁶⁾.

Figure 9 compares the electrolyte membrane's molecular weight retention rate as calculated in Eqs. (2) and (3) based on operating history during FC system durability testing with actual values measured after durability testing was completed. The range of measured values indicates variation among measured values at multiple locations by their stacking position and within the cell plane. The measured values show that degradation progressed more

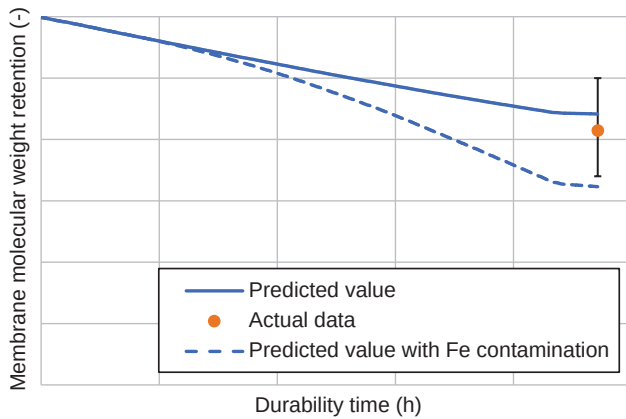


Fig. 9 Comparison of actual and predicted membrane degradation during FC system durability testing

than the predicted values suggested. Although it was confirmed that the necessary durability was maintained, this discrepancy is presumed to be because of accelerated degradation caused by Fe contamination. It has been confirmed that when deterioration acceleration caused by the maximum amount of Fe mixed in during durability testing is taken into account, it nearly matches the lower limit of variation of actual measured values.

4.3. Dealing with Mechanical Degradation of Electrolyte Membranes

Mechanical degradation of the electrolyte membrane caused by repeated swelling and shrinking of the electrolyte membrane due to changes in humidity is accelerated by cracks in the electrode⁽¹⁷⁾.

The frequency of electrode cracks themselves was suppressed by using a more suitable electrode ink composition and by quality control of the surface properties. In addition, the use of high-strength membrane reinforcement increased the toughness of the electrolyte membrane against degradation stresses that may occur at cracks, thereby suppressing the impact of cracks.

5. Simplifying the Structure of the Outer Resin Structure

5.1. Resin Film

To simplify the structure of the outer resin structure, the flow channel structure was integrated with the separator, helping to eliminate the need for a flow channel structure in the resin frame of the UEA. In this UEA, the outer structure was simplified by switching to punch-molded general-purpose resin film⁽¹⁸⁾. Because film could be used, a structure was adopted in which a general-purpose resin film with a linear expansion coefficient close to that of SUS, the material used for the BPPs, is laminated together to form the UEA structure shown in Fig. 3.

Previously, precision injection molding was used, with expensive engineering plastic. Because of its molding properties, injection molding required a thickness of 250 μm or more, but using general-purpose resin film made it possible to reduce this thickness by more than 50%, which helped increase the volumetric output density. It also cut the cost per piece in half compared with the previous cost.

5.2. Strength Durability

As Fig. 3 shows, the resin film is sandwiched between BPPs. When power is being generated, stress may occur in the resin film due to the gas pressure differential between the cathode and anode gases flowing in the BPP flow channel. The resin film retention interval distance by BPPs was set to relieve the stress. Moreover, in fuel cells, where water is generated by the process of power generation, hydrolysis causes chemical degradation. For that reason, the mechanical strength was designed for the gas pressure differential between the cathode and anode, taking into

account the reduction in resin film strength due to chemical degradation.

Finite element method simulation was performed on the gas pressure differential. As Fig. 10 shows, the simulation confirmed the relationship between stress and the retention interval distance by BPPs. The stress was suppressed by controlling the distance between the restraints with BPPs. As a result, sufficient strength durability was achieved.

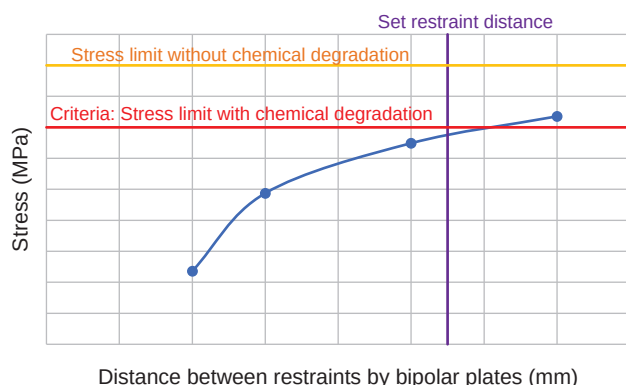


Fig. 10 Relationship between stress and retention interval distance by BPP

6. Conclusion

The low-cost UEA used in the newly structured fuel cell stack was developed in collaboration with General Motors and achieved the following compared with the previous UEA. It is more than twice as durable and has only 1/3 the cost of the previous fuel cell stack, while maintaining the previous fuel cell stack's output performance⁽⁴⁾.

- (1) By using Pt alloy catalyst, porous carbon, and a low supporting rate, which were predicted to inhibit degradation, the amount of Pt in electrodes was reduced to 1/5 that of the previous UEA.
- (2) The thickness of the electrolyte membrane was reduced to 3/5 that of the previous fuel cell stack by addressing chemical and mechanical degradation and by using a proprietary coating process.
- (3) By simplifying the structure of the outer resin structure, a resin film with a thickness reduced by half could be used.

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