

Empirical Model for Polymer Electrolyte Fuel Cell Electrocatalyst Degradation Using Platinum Stress

Masakuni YAMAMOTO*

Hiroshi MATSUMORI*

ABSTRACT

A method of predicting polymer electrolyte fuel cell performance loss due to electrocatalyst degradation was developed in order to reduce the performance loss caused by fuel cell startup and stop and by acceleration and deceleration during driving.

An electrochemical method was used to define a platinum stress index to express the influence on platinum (Pt) catalyst degradation from potential cycles in a fuel cell stack. It was found that Pt stress shows an exponential relationship with the rate of loss of catalyst activity, and an empirical prediction model for Pt catalyst degradation was developed.

When the developed model based on Pt stress was used to predict performance loss in a stack during startup and stop, the results showed that a prediction accuracy of approximately 90% was achieved and confirmed that the model could be applied as a catalyst degradation prediction model.

Operating conditions derived from the prediction model were incorporated into fuel cell system development. The results showed that the rate of performance loss during startup and stop was reduced by approximately 57 points compared to the conventional rate, thereby confirming the usefulness of the model.

1. Introduction

A fuel cell vehicle (FCV) that uses hydrogen for fuel has driving performance on a par with gasoline vehicles and also has high environmental performance with its clean emissions and high efficiency.

The motors also provide smooth driving and quiet operation. The FCV is therefore attracting attention as a method for reducing the environmental impact of the automobile.

Widespread popular adoption of the FCV will necessitate higher fuel cell (FC) stack performance and durability as well as lower cost. In previous development, one issue in FC durability has been performance loss, and performance loss is known to be greater at startup and stop and when the vehicle is driving (Fig. 1).

One factor in the acceleration of performance loss at startup and stop that has been proposed is the mechanism whereby the electrocatalyst of Pt nanoparticles supported on carbon black is ionized due to repeated operation of the cathode high potential during startup and stop, so that the Pt is dissolved and diffused in the polymer electrolyte membrane, after which the Pt is deposited due to reduction of Pt ions by crossover of hydrogen through the anode side ⁽¹⁾⁻⁽³⁾. The catalyst in a single cell was actually observed using a scanning transmission electron microscope (STEM)

after assumed startup and stop testing. Figure 2(a) shows the Pt nanoparticles dispersed on carbon black before the testing, whereas Fig. 2 (b) shows these are dissolved and deposited in the polymer electrolyte membrane (Fig. 2(b) area outlined in red). STEM observations shown in Fig. 2 match with the previously hypothesized mechanism.

The degradation of electrocatalyst is the main factor in performance loss, and establishment of a technique to inhibit this degradation is an urgent issue for enhancement of FC durability. It is important, therefore, to have an index representing the relationship of Pt catalyst degradation to

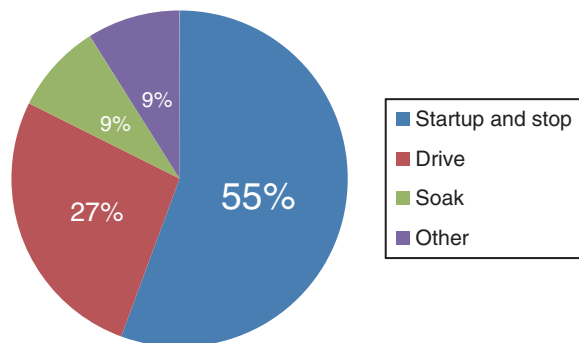


Fig. 1 Breakdown of performance loss in conventional FC stack

* Automobile R&D Center

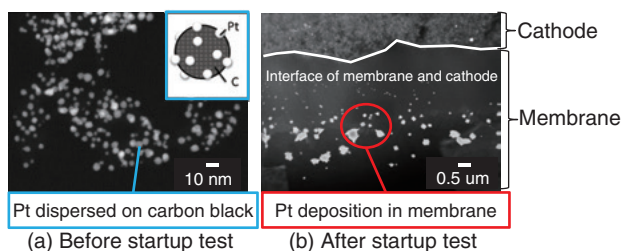


Fig. 2 STEM image of Pt catalyst on carbon black support (Pt/C) before (a) and after startup test at 1000 cycles (b)

FC system control, but there are virtually no instances of the relationship between control and Pt catalyst being clearly represented in the form of an index.

2. Development Goals

This paper will describe the development of a new index that represents the relationship between FC system control and Pt catalyst degradation, and the development of an empirical model, based on that index, for predicting Pt catalyst degradation.

It will also describe cases relating to the development of FC system control in order to mitigate catalyst degradation during startup and stop according to the prediction.

Issues involved in the development of an index representing the relationship between FC system control and Pt catalyst degradation and in development of a catalyst degradation prediction model include the following:

Issue 1: A method for quantifying and indexing the influence that potential cycle in the FC have on catalyst degradation; and

Issue 2: Setting the quantitative correlation between the index and FC stack performance loss.

First of all, resolving Issue 1 will require research on a method for detecting the changes that occur in Pt catalyst with respect to potential cycles.

Cell voltage in FC is expressed as the difference between the potential at the cathode and at the anode, and the anode potential (reversible hydrogen electrode, RHE) is the reference for the cathode potential (cathode potential [V vs. RHE] in Fig. 3).

Since hydrogen oxidation reaction (HOR) of the fuel occurs at the anode, the RHE in the case of pure hydrogen will generally show as around 0 V. Figure 3 shows an example of a typical pattern of the cathode potential cycle when the RHE is taken as the reference and the current that loads on the stack (the FC current in Fig. 3)⁽³⁾. By feeding hydrogen as fuel into the anode and air as an oxidizing agent into the cathode, the cathode will be in a high potential state (the “Start” portion in Fig. 3), just as in the case of open circuit voltage (OCV) in an ordinary battery. Low power operation with the FC in an idle state

causes a slight loss in the cathode potential (the “Idle” portion in Fig. 3). The subsequent patterns of the cathode potential cycle of the stack become complicated, with the sudden drop in the potential due to high power loaded (the “High load” portion in Fig. 3), the cathode potential cycles resulting from acceleration and deceleration during driving (the “Acceleration or deceleration” portion in Fig. 3), and the state of reduced cathode potential due to cathode oxygen concentration control when stopping (the “Stop” portion in Fig. 3).

Cyclic voltammetry (CV) was taken up as an electrochemical method for understanding changes that occur in the Pt catalyst with respect to these voltage cycles.

CV can provide information about Pt catalyst reactions with regard to potential in terms of the current caused by electrochemical reaction. Figure 4 shows a typical current-potential curve obtained by CV using carbon-supported Pt catalyst. The insets in Fig. 4 show a schematic representation of reactions on the Pt catalyst surface. At around 0.5 V, a current charged/discharged by

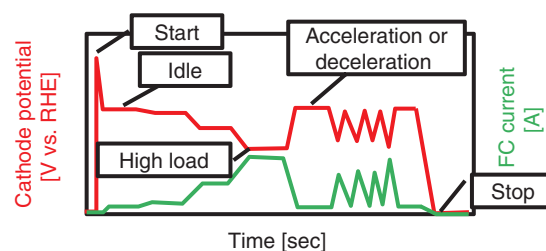


Fig. 3 Typical pattern of cathode potential cycle in FCV

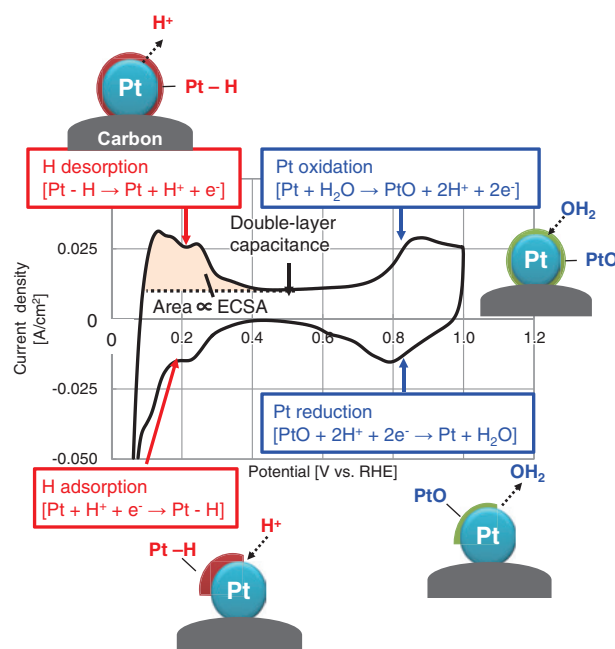


Fig. 4 General CV depicting features observed on the surface of Pt catalyst

electrochemical double-layer capacitance is observed in the FC, and from 0.6 to 1.0 V, formation of a Pt oxide layer is caused by an adsorption reaction of Pt and molecular water (Pt oxidation). The Pt oxide is reduced to bare Pt surface by scanning potentials from 1.0 to 0.6 V.

At an potential lower than 0.5 V (from 0 to around 0.45 V), however, hydrogen adsorption desorption (HAD) occurs on the Pt catalyst. The charge of HAD (q_{HAD}) and the electrochemical surface area (ECSA) of the catalyst are in the relation of proportionality shown in Eq. (1). Therefore, the effective Pt catalyst surface area can be obtained from q_{HAD} .

$$\text{ECSA} [\text{cm}^2] = \frac{q_{\text{HAD}} [\text{uC}]}{210 [\text{uC}/\text{cm}^2]} \quad (1)$$

In Eq. (1), 210 uC/cm^2 is the commonly used conversion ratio of q_{HAD} and electrochemically effective surface area of Pt catalyst.

Given the above characteristics, CV was adopted as the method for quantifying the influence that potential cycle in FC has on catalyst degradation.

Next, in order to resolve Issue 2, the index determined by CV had to be tied to FC performance. We therefore focused on the relationship between FC performance loss and catalyst degradation.

FC performance is commonly evaluated using the polarization curve (Pol. curve) shown in Fig. 5⁽⁴⁾. The factors in the voltage loss can largely be placed in three main categories, which include:

- (1) Ohmic loss (iR_{loss}) in the FC system;
- (2) Overvoltage (η_{ORR}) relating to the oxygen reduction reaction (ORR) in the cathode; and
- (3) Overvoltage (η_{tx}) relating to oxygen transport to the cathode catalyst.

The cell voltage in the FC stack is expressed as the difference between the voltage losses in (1) to (3) and the voltage loss due to anode HOR (HOR_{loss} in Fig. 5).

Of the three, factor (2) accounts for the greatest part of the voltage loss. Since factor (2) depends on the kinetics of ORR, it has a determining influence on catalyst activity, and ECSA loss in the Pt catalyst contributes greatly to it.

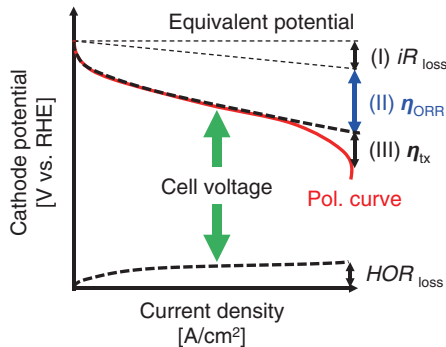


Fig. 5 Diagram of relationship between FC polarization curve and voltage loss

ORR occurs because molecular oxygen is adsorbed in and desorbed from the surface of the Pt catalyst in the FC, as with catalysts in general. In order to increase the ORR rate, therefore, the ECSA becomes an important physical property because it indicates the active sites of Pt catalyst reaction. The quantity η_{ORR} that is derived from the equation for the catalytic reaction rate is theoretically expressed by Tafel's equation in Eq. (2), and it is known to have a logarithmic relationship with the ECSA⁽⁵⁾.

$$\eta_{\text{ORR}} = \frac{2.303 \times RT}{\alpha F} \times \log \left(\frac{\text{ECSA} \times i_0}{i} \right) \quad (2)$$

R is the ideal gas constant, T is the absolute temperature, α is the cathode transfer coefficient, F is Faraday's constant, i is the reaction current density, and $\text{ECSA} \times i_0$ corresponds to the effective exchange current density of the catalyst.

It was reasoned that the development of a catalyst degradation model capable of predicting ECSA loss from the relationships in Fig. 5 and Eq. (2) would enable estimation of performance loss. To that end, the mechanism of presumed ECSA degradation was hypothesized in order to correlate the index describing catalyst degradation due to potential cycles in the FC with the ECSA, which is a catalyst property.

As indicated in Chapter 1, previous reports indicated that the coagulation of Pt nanoparticles caused by dissolution and deposition of Pt were the determining factor in ECSA loss⁽¹⁾. The view was therefore taken that the basic mechanism was loss of catalyst activity due to the potential cycle.

In order to address the potential cycles caused by startup and stop in FC and by acceleration and deceleration during driving, shown in Fig. 3, potential cycle was further subdivided into the three elements of the attained potential, the hold time at one, and scan rate.

Quantification of the influence on Pt catalyst was examined in terms of these three elements.

3. Quantification of the Influence of Potential Cycle on Catalyst Degradation

3.1. Experimental

In order to quantify the influence of potential cycle on Pt catalyst, CV measurement was carried out using a single cell.

The membrane electrode assembly (MEA) used for evaluation was fabricated by hot pressing commercially available catalyst layer decals onto polymer electrolyte membranes (catalyst coating membrane, CCM) and sandwiched by gas diffusion layers (GDL). The MEA is placed into a single cell, which has a 50 cm^2 active area (Fig. 6).

The single cell operating conditions are summarized in Table 1.

The basic conditions for cell operation were set with reference to previous reports⁽⁶⁾. The cell temperature was

Table 1 Operating conditions of single cell

Operating condition	Anode (RE/CE)	Cathode (WE)
Atmosphere (flow rate) [NLM]	H ₂ (0.3)	N ₂ (2)
Pressure [kPa]	230	200
Temperature [°C]	70	
RH [%]	50	73

controlled by thermocouple to maintain a level of 70°C, and the relative humidity (RH) at the anode and cathode was adjusted using a humidifier. Humidified pure hydrogen gas was fed into the anode flow field at 0.3 NLM, while an inert gas (N₂) was fed into the cathode flow field at 2 NLM, and total gas pressures were maintained at the anode of 230 kPa and at the cathode of 200 kPa.

The cathode was set as the working electrode (WE) and the anode as the reference and counter electrode (RE and CE). WE potential with respect to the RHE was controlled using a potentiostat-galvanostat (PGS) in order to measure the reaction current on the Pt catalyst. Figure 6 shows a schematic diagram of the measurement system.

The CV measurements were performed by setting from the potential elements (attained potential and hold time at one) at 50 mV s⁻¹ sweep rate.

3.2. Quantification of the Influence of Upper Level Potential Due to Startup and Stop Operation and Its Hold Time

First, the influence of the upper level potential (ULP) assumed by a high cathode potential during startup was confirmed. As an example of verification, Fig. 7 shows the cathode potential cycling profile and current-potential curves with ULP set at 0.9 V and 1.0 V.

As shown in Fig. 4, the total current for HAD observed

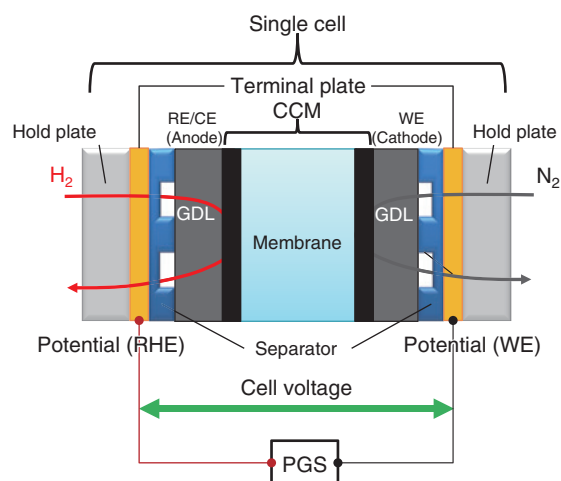


Fig. 6 Schematic drawing of electrochemical system using single cell

at the potentials from 0 to 0.45 V showed low dependence on ULP.

By contrast, it was verified that Pt reduction current obtained at the potential from 0.6 to 1.0 V was strongly dependent on ULP.

The higher the ULP, the greater the increase in the reduction current of the PtO layer formed by the rise in cathode potential, and this means that the electrochemical oxidation of Pt progressed due to molecular oxygen. It was found that ULP is the main factor that accelerates Pt catalyst oxidation, as reported in a previous study⁽⁷⁾.

Next, the ULP was fixed and CV measurements were made by the profile of cathode potential cycles with changed hold times (Fig. 8) in order to clarify the influence of Pt catalyst oxidation in terms of ULP hold time. From the CV measurement results in Fig. 8, it was found that a difference appears in the current from reduction of the Pt

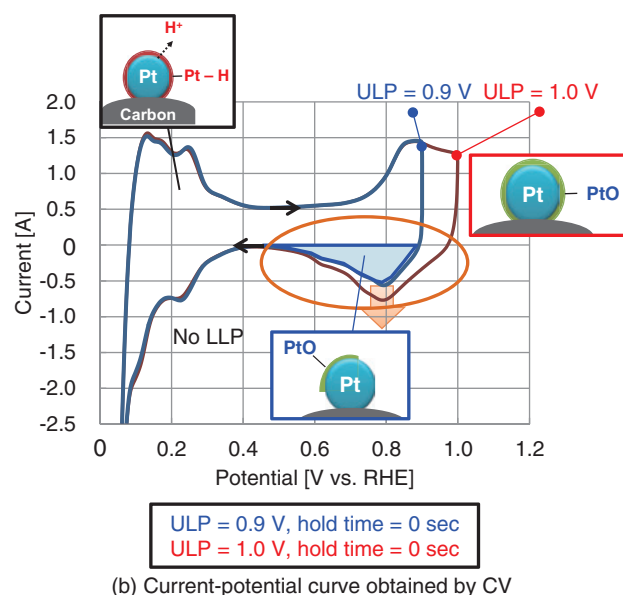
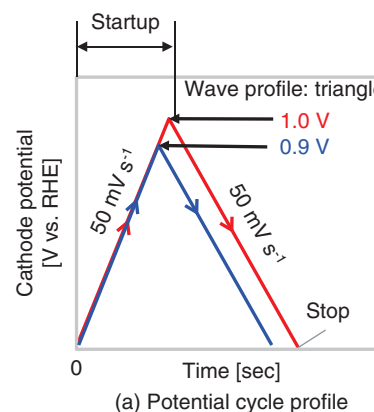


Fig. 7 Graph of WE potential trend, showing changed ULP levels using CV (a). CVs obtained at each ULP from 0.05 V up to 0.9 V (blue solid line), up to 1.0 V (red solid line) (b).

catalyst and molecular oxygen even when the ULP remains at the same 1.0 V as shown by the dotted curve in Fig. 8. Because the reduction current increases with respect to potential hold time, electrochemical oxidation of Pt surface was found to be dependent on these factors.

3.3. Quantification of the Influence of Lower Level Potential from Acceleration and Deceleration During Driving and Its Hold Time

Next, CV measurements were taken with the lower level potential during driving (LLP) due to potential cycles during acceleration set. The purpose was to quantify the influence of cathode potential cycles from acceleration and deceleration during driving. As an example of verification, Fig. 9 shows the cathode potential cycling profile and current-potential curve obtained by CV after ULP hold at 1.0 V for 100 sec and LLP hold at 0.8 V for 100 sec. The LLP in Fig. 9 assumes cathode potential drop due to the load applied during acceleration and deceleration. Comparison of results is shown in Fig. 8, which assumed startup and stop,

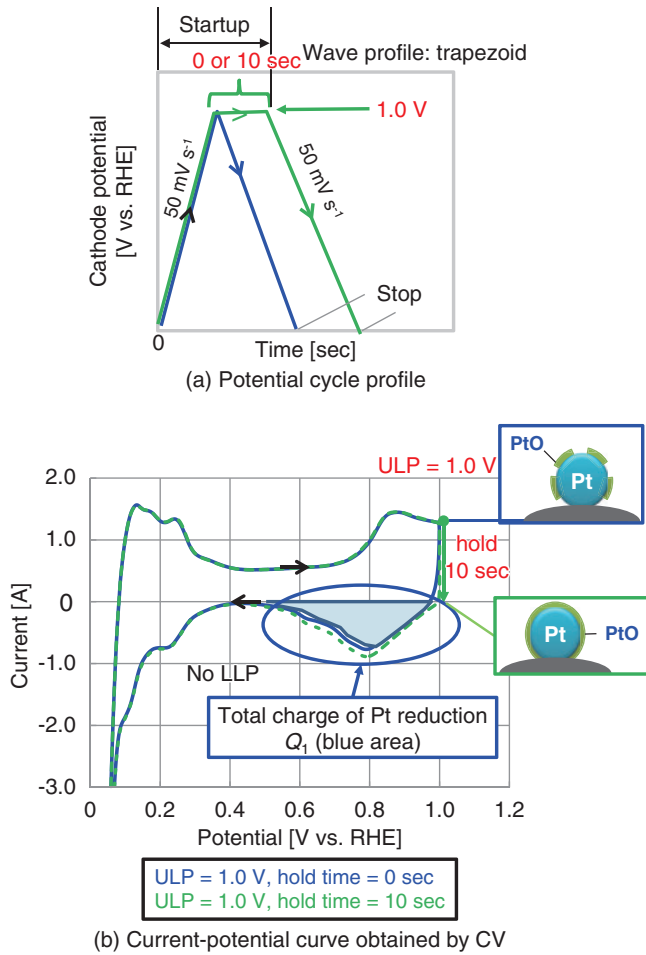


Fig. 8 Potential trend at cathode in CV (a). CVs obtained at same ULP from 0.05 V up to 1.0 V (blue solid line), and hold at 1.0 V for 10 sec (green dotted line) (b).

and in Fig. 9, which simulates cathode potential cycles due to acceleration and deceleration during driving. The total reaction charge of the Pt oxidation reaction due to ULP on the assumption of startup in Fig. 8, whose potential profile has no LLP operation, could be obtained as the charge of the PtO layer reduction reaction (Q_1 in Fig. 8).

On the other hand, when LLP operation at 0.8 V occurs on the assumption of cathode potential cycles due to acceleration and deceleration during driving, the Q_1 from ULP is reduced from a to b in Fig. 9 due to the hold time at LLP. It is thought that the charge of the reduction of PtO due to ULP is decreased by LLP holding caused by the load of acceleration and deceleration.

From these verifications that CV was used to quantify the influence of potential cycles in the FC on Pt catalyst, examination of the CV results shows that the charge caused from the PtO layer (the Pt reduction reaction current in Fig. 4) demonstrates strong dependence, and can clearly be seen to increase and decrease according to the ULP, LLP and their hold times.

Based on the above results, Pt stress was defined as the index of catalyst degradation as shown in Eq. (3) below. The purpose of this is to differentiate the influence of

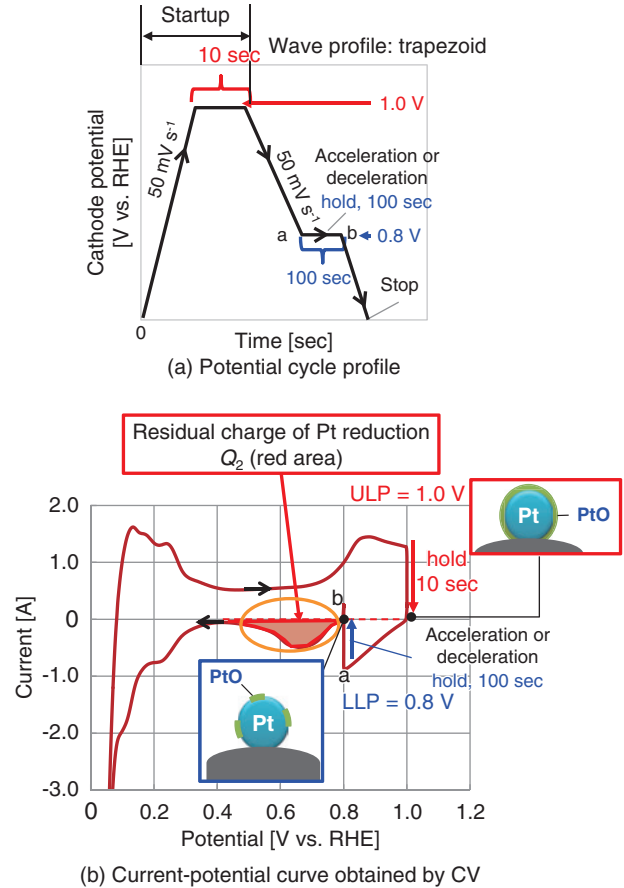


Fig. 9 Potential trend at cathode in CV (a). CV measured by 10 sec hold at ULP = 1.0 V, and 100 sec hold at LLP = 0.8 V (b)

cathode potential cycles resulting from startup and stop and from acceleration and deceleration during driving.

$$\text{Pt stress} \equiv \Delta Q = Q_1 - Q_2 \quad (3)$$

Q_1 is the total charge of the PtO layer when cathode high potential is assumed to occur during startup and stop. Q_2 is the residual charge of the PtO layer after reduction by load cycles during driving (Q_2 in Fig. 9). The Pt stress is determined from these charges that are based on electrochemical reactions. The influence of potential cycle resulting from mode of operation (ULP, LLP, and their hold times) is expressed by the variation of change (ΔQ). As distinct from results previously reports⁽⁷⁾, it becomes possible to determine the influence of LLP that arises from load cycle by Pt stress defined as variation of ΔQ .

In light of the above, the use of Pt stress as an index enabled the influence that potential cycles in the FC operation have on catalyst degradation to be expressed quantitatively. In the following, Pt stress will be termed ΔQ .

In order to apply to potential cycles during FC operation of the kind shown in Fig. 3 with ΔQ as an index, the ULP, the LLP, and these hold times were varied while obtaining CV measurements, and the dependence of ΔQ on potential was mapped (Fig. 10). By using Fig. 10, it became possible to quantify the influence of potential cycles in actual vehicle of the kind as shown in Fig. 3.

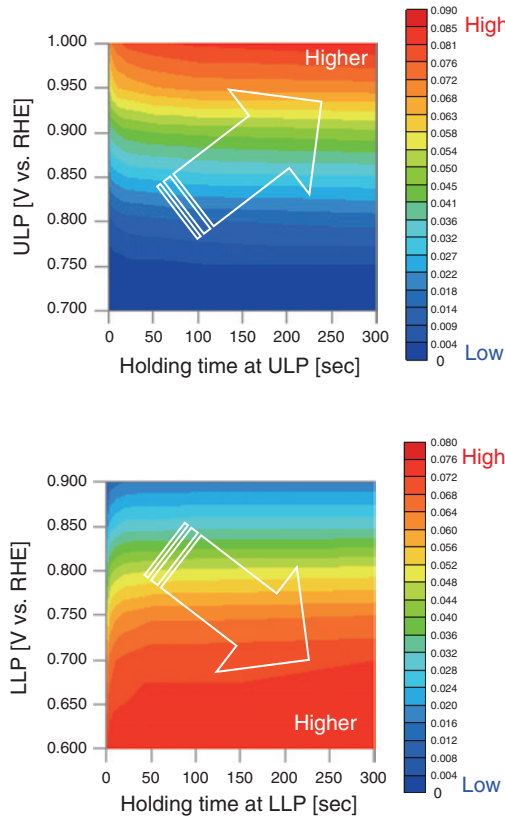


Fig. 10 Dependence of ΔQ on ULP, LLP and holding time at each potential. CVs obtained by the same conditions as Table 1

Table 2 List of accelerated stress test (AST) by potential cycling mode using single cell

Mode	Assumption	Cell temperature [°C]	RH [%]		Potential [V vs. RHE]		Wave profile	Holding time [sec]
			An.	Ca.	ULP	LLP		
Load cycle-I	Drive low RH	70	50	40	0.90	0.70	Rectangle	4
Load cycle-II	Drive middle RH	70	50	73	0.90	0.70	Rectangle	4
Load cycle-III	Drive high RH	70	50	100	0.90	0.70	Rectangle	4
Potential cycle-IV	Start up middle ULP	45	100	100	0.95	0.80	Trapezoid	4
Potential cycle-V	Start up high ULP-1	45	100	100	1.20	0.80	Trapezoid	4
Potential cycle-VI	Air Start up	45	100	100	1.40	0.80	Trapezoid	5
Potential cycle-VII	Fuel starvation	45	100	100	1.60	0.80	Trapezoid	5
Potential cycle-VIII	High load	70	50	73	0.95	0.65	Trapezoid	4
Potential cycle-IX	Middle load	70	50	73	0.90	0.70	Triangle	0

4. Setting the Correlation Between ΔQ and FC Stack Performance

The dependence between ΔQ and ECSA, which determines the voltage drop due to ORR, was confirmed in order to clarify the correlation between FC stack performance loss and ΔQ , quantified above through Chapter 3.

In order to confirm the dependence of ECSA on ΔQ , a single cell was used to conduct potential cycle testing, and the dependence between the rate of ECSA loss and ΔQ was verified. CV measurement was implemented under the same measurement conditions as described in Section 3.1. In the potential cycle test, ΔQ was varied by the conditions in Table 2, which simulates the potential cycles in actual vehicle. Modes IV to VII reflect cathode potential and temperature that assume startup and stop operation, and the rest of the modes were taken to assume acceleration and deceleration during driving.

Figure 11 shows the results from comparison of modes VIII and IX, which assume acceleration and deceleration during driving.

According to the ULP, LLP, and their hold times, the ΔQ per unit cycle can vary by a factor of approximately 2.5 even in the same acceleration and deceleration mode.

Potential cycle testing on the assumption of acceleration and deceleration in which ΔQ differs by a factor of approximately 2.5 was conducted for the designated number

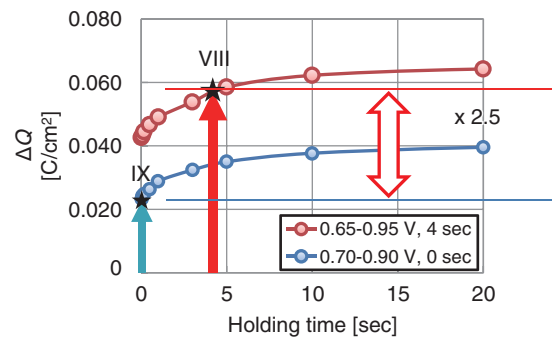


Fig. 11 Comparison of ΔQ under operating conditions of modes VIII and IX

of repetitions. Transitions in the rate of ECSA loss were confirmed.

Figure 12 shows the relationship between ΔQ and the rate of ECSA loss.

The rate of ECSA loss during potential cycles in which ΔQ varies by a factor of approximately 2.5 was found to change by two orders of magnitude or more. Similarly, when the levels of ΔQ were varied according to Table 2, the results were found to show an exponential relationship with the rate of ECSA loss. However, although the correlation accuracy (correlation coefficient; $R^2 = 0.975$) in Fig. 12 is high, there is a difference from the actual measurement value. It was thought that the difference from the correlation occurs because ECSA loss operated under the conditions set to assume startup and stop (IV to VII) in Table 2 include the influence of carbon corrosion as well as the expected Pt degradation.

The influence of carbon corrosion on ECSA loss has not yet been defined. However, the results in Fig. 12 enable management of the rate of ECSA loss in each operating mode by means of ΔQ . This made it possible to clarify the difference between the catalyst degradation that occurs in startup and stop operation and in driving operation, in both of which it results in large performance losses.

It also becomes possible to estimate the rate of ECSA loss by calculating ΔQ from the control elements (ULP, LLP, and their hold times) for an actual vehicle when the map in Fig. 10 is used.

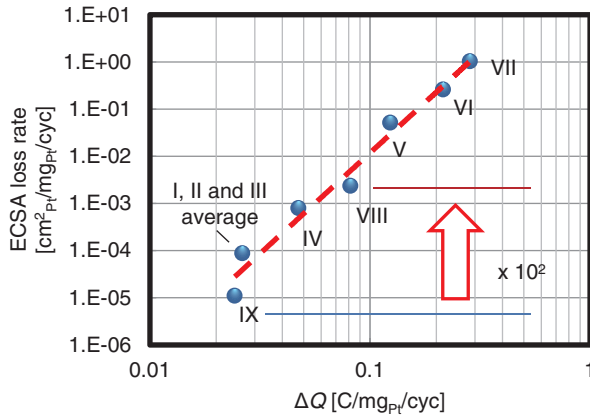


Fig. 12 Correlation of ΔQ and ECSA loss rate

5. FC System Control with Platinum Stress as Index

5.1. Verification of the Catalyst Degradation Prediction Model

In order to confirm the accuracy of the catalyst degradation prediction model that was developed as described up through the previous chapter, the performance loss was verified in tests assuming startup and stop

operation with a short stack.

Figure 13 shows the average voltage profile for the stack in assumed startup and stop testing.

The stack starts up at around 0 V due to control of the gas environment at both electrodes; then, when the air as oxidizing agent feeds into the cathode during startup, stack voltage rises. The cathode of the stack is held at OCV for the designated times, and when the operation of the stack stops it returns to around 0 V due to the control of oxygen concentration. Consequently, the Pt catalyst undergoes repeated potential cycles from 0 V to 1 V.

Catalyst degradation in startup and stop simulation testing was predicted according to the flow chart in Fig. 14 in order to estimate ΔQ according to the potential cycle in Fig. 13.

ULP and ULP hold time during startup and stop are identified from the average voltage profile for the short stack shown in Fig. 13. (In Fig. 14, ULP = 1.0 V and holding time = 100 sec.) The ΔQ during startup and stop is calculated from these potential elements and the ΔQ map in Fig. 10. The calculated ΔQ is converted into numerical values per amount of Pt loading in the cathode, and the exponential relationship in Fig. 12 is used to estimate the rate of ECSA loss during startup and stop in the short stacks.

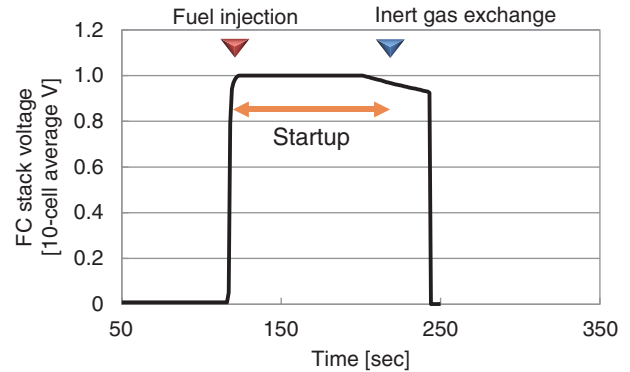


Fig. 13 Average voltage profile of FC stack at startup

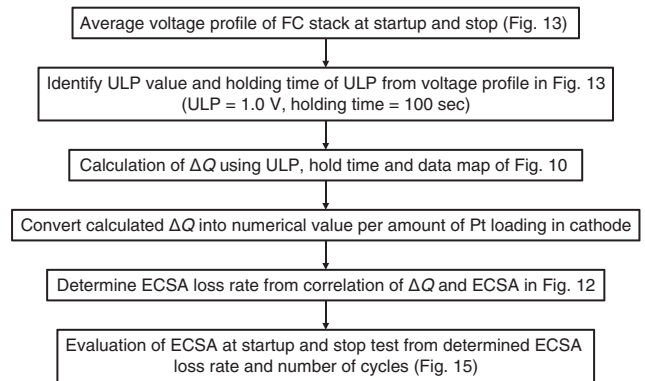


Fig. 14 Prediction flow of catalyst degradation in FC stack using empirical model based on ΔQ

The rate of ECSA loss estimated in Fig. 14 is multiplied by the number of cycles of the startup and stop simulation test, and the ECSA loss during startup and stop was predicted. Figure 15 shows the results from comparison of the estimated curve obtained by the catalyst degradation model (the dotted line in Fig. 15) with the actual ECSA values obtained from using the short stacks by CV measurement (the filled circle in Fig. 15).

Comparison of the ECSA loss rates in the final test cycle (end of test, EOT) showed that the model had prediction accuracy yielding approximately 90% conformity. These results demonstrate that the model developed with a single cell can be used to estimate catalyst degradation in the FC stack of an actual vehicle.

5.2. Verification of the Effectiveness of FC System Control with Pt Stress as an Index

Given the confirmation that the developed model is applicable to stacks, the empirical catalyst degradation model is complete. This model was used to examine operation modes that would inhibit catalyst degradation in order to reduce the performance loss during startup and stop that has been an issue with conventional stacks.

As shown in Fig. 13, cathode high potential cycle occurs at potentials from 0 V to 1 V during startup and stop. Reduction of ΔQ during startup and stop will be an important approach to inhibiting catalyst degradation. It was reasoned, therefore, that it would be necessary to have

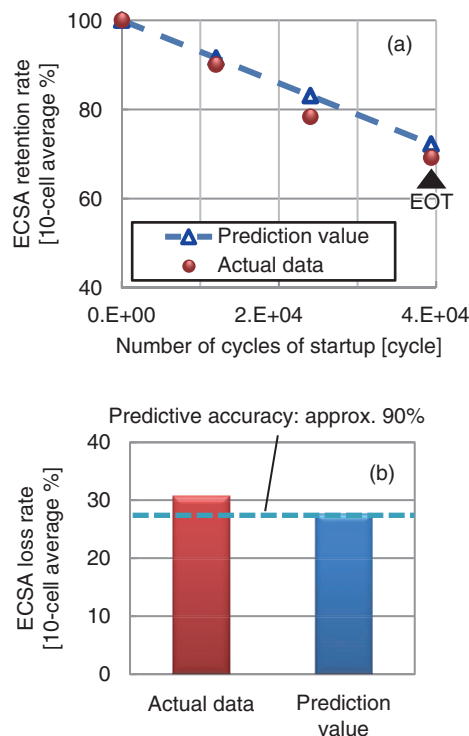


Fig. 15 Actual data of ECSA during startup cycle and prediction line (a). Comparison of actual data and predicted data at EOT (b)

control that lowers ULP due to OCV during startup.

When the conventional FC stacks stop, the stack voltage falls instantly to 0 V, which causes ΔQ to increase. An attempt was made to address this by reducing the margin of voltage loss after the cathode potential is held at startup. In other words, control to hold LLP at a higher level was implemented in order to reduce ΔQ .

Given the direction shown by estimation using the model and the feasibility of FC system control, voltage control during startup and stop was introduced to the stack to realize these goals. As a result, ΔQ was successfully reduced. The use of voltage control cut such high cathode potential as the OCV during startup, reducing the influence on ΔQ , which is an index of Pt catalyst degradation caused by ULP. Control of the voltage drop when stopping was also implemented to optimize LLP.

Control measures reduced ΔQ by approximately 10% at startup and approximately 20% during stopping relative to conventional FC figures. Catalyst degradation was inhibited, reducing the rate of stack performance loss during startup and stop by approximately 57 points (Fig. 16).

Following the above research, control to reduce performance loss based on the newly developed Pt catalyst degradation prediction model was demonstrated in FC system development and its usefulness was confirmed.

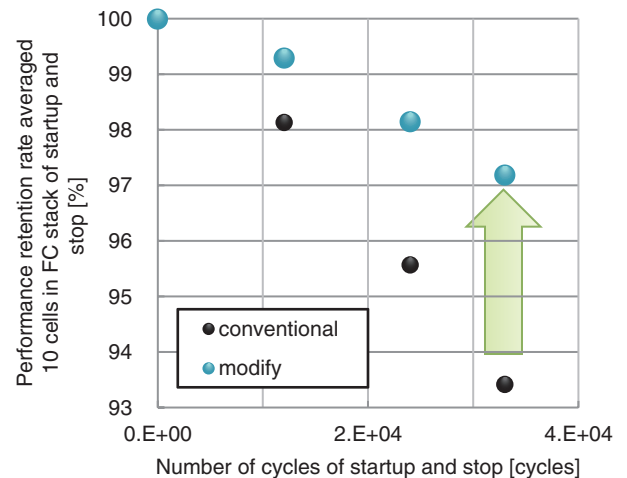


Fig. 16 Verification result of effect to performance loss in FC stack by modify mode at startup and stop

6. Conclusion

- (1) Pt stress was newly developed as a quantification of the influence that potential cycles in the FC have on catalyst degradation. This enabled quantification of the extent of influence of each operating mode.
- (2) The correlation of the rate of ECSA loss with Pt stress was newly discovered. By use of this correlation, Pt

stress could be calculated from operating conditions, making it possible to rapidly estimate ECSA loss trend.

- (3) Use of the developed empirical catalyst degradation prediction model enabled estimation of catalyst degradation in the FC stack in an actual vehicle and confirmed that the model is highly accurate. The model links the catalyst with operating conditions, and therefore enables estimation of conditions that inhibit degradation from the catalyst perspective, and the fact that the model can be used in development of a method for FC system control has confirmed its usefulness.

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■ Author ■



Masakuni YAMAMOTO



Hiroshi MATSUMORI