

# Stainless Steel Foil for Bipolar Plates of PEFCs NSSMC-NAR-316BC

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## Abstract

*Fuel cell is an ultimate clean energy sources, because it utilizes hydrogen as a fuel and produces only water as a by-product. The system, therefore, can be one of the essential alternative generations in view of urgent global environmental issues. This paper presents a newly developed stainless steel foil "NSSMC-NAR-316BC" for bipolar plates of PEFCs (Polymer Electrolyte Fuel Cell). The developed one provides both excellent corrosion resistance as well as superior electrical conductivity.*

## 1. Introduction

Modern society is faced with global warming ascribable largely to the ever-expanding energy demand and the ever-increasing CO<sub>2</sub> emissions on a global basis. Fuel cells ("FCs") are a clean energy source that generate electricity from hydrogen and oxygen and produce water as only waste matter. As one of energy sources of the future, FCs will increase in importance.<sup>1)</sup> Of them, the polymer electrolyte fuel cell ("PEFC") offers a high output density despite the fact that its operating temperature is as low as 70-90°C. In addition, since PEFCs can be made lighter in weight and smaller in size, the application of PEFCs as power supplies for automobiles and as portable distributed power sources for home use is considered in earnest.

In the construction of a PEFC, the separators (or bipolar plates) not only form a passage for gases but also partition the adjoining cells (basic constituents of FC) stacked one on another and serve as the transmitter (connector) of an electric current to the outside. In the present study, we developed a new stainless steel foil, NSSMC-NAR-316BC ("316BC"), in which conducting metallic precipitates are finely dispersed so as to secure good corrosion resistance and high conductivity that are required of the PEFC separators.<sup>2)</sup>

## 2. Concept of Newly Developed Foil

### 2.1 Properties required of FC separators

The image of a PEFC is schematically shown in **Fig. 1**. The separators make contact with the anode/cathode gas diffusion layer ("GDL") of the membrane electrode assembly ("MEA"). Therefore, any material used for separators is required to have sufficiently low contact resistance with the carbon paper (CP) that makes up the GDL. On the other hand, since many sulfuric acid groups are coordinated in the polymer membrane of the MEA to secure adequate

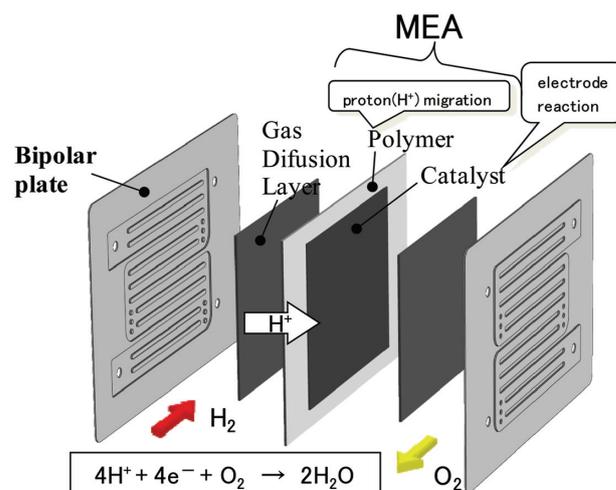


Fig. 1 Schematic image of PEFC

conductivity, there is a possibility that sulfuric acid ions dissolve in the water produced in the FC as the membrane deteriorates. Thus, depending on circumstances, the FC separators may be put in a severe sulfuric acid environment while the FC is in and out of operation. Therefore, it is necessary that they always display good corrosion resistance regardless of their operation conditions and environment.

### 2.2 Development of a solid metallic separator having high conductivity and good corrosion resistance

By replacing conventional carbon separators with those made from metal, especially corrosion-resisting stainless steel foil, it be-

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comes possible to make the PEFC stacks of multiple cells less costly, lighter in weight, and smaller in size at the same time. In addition, by implementing progressive press forming, it becomes possible to enhance the productivity and lower the manufacturing cost of separators markedly. Furthermore, cells and stacks of welded construction become practicable. As mentioned earlier, to secure the desired performance of PEFC stacks, it is necessary to lower the electrical contact resistance between the GDL and the separators. As is well known, stainless steel sheets are coated with a passive-state film on the surface, the electrical contact resistance of which is so high that these sheets cannot be used directly for separators. Ordinarily, therefore, the separators are plated with gold—a precious metal—on the surface. Even so, various problems have yet to be solved before stainless steel can be used for separators on a mass-production basis; these problems include the deterioration of separator corrosion resistance due to plating defects, the adverse effects of plating exfoliation on separator long-term durability, and the cost of recovery recycling expensive gold.

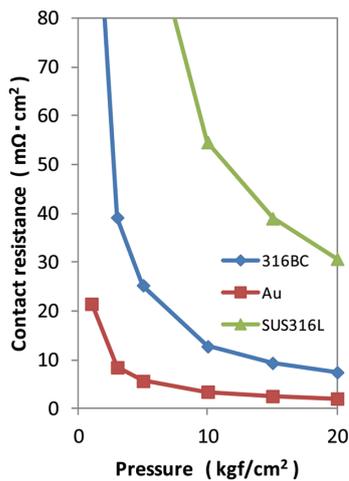
For the newly developed stainless steel foil, “NSSMC-NAR-316BC,” surface conductivity has been improved by exposing to the surface those fine conducting metallic precipitates (borides composed mainly of Cr: M<sub>2</sub>B) which are widely dispersed in the steel by

means of spray etching in ferric chloride solution. (The process of exposing M<sub>2</sub>B to the surface is called “surfacing.”) The corrosion resistance of the cell interior is secured by so designing the material that the mother phase composition becomes almost the same as that of SUS 316L. **Figure 2** shows the contact resistance between 316BC and CP. It can be seen that the higher the surface pressure (i.e., stack clamping pressure), the lower becomes the contact resistance. Although it cannot be said that 316BC is an equivalent of gold-plated stainless steel sheet, it is by far lower in contact resistance than SUS 316L sheet. The low contact resistance of 316BC is maintained even after it is exposed to the open air for a long time. **Figure 3** shows an scanning electron microscope (SEM) image of 316BC after ferric chloride etching. It is M<sub>2</sub>B that is dispersed in the surface layer of 316BC.

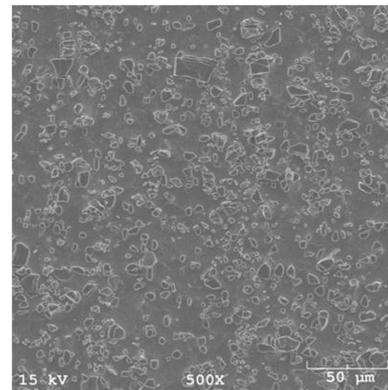
### 3. Properties of NSSMC-NAR-316BC<sup>3)</sup>

#### 3.1 Corrosion resistance and conductivity

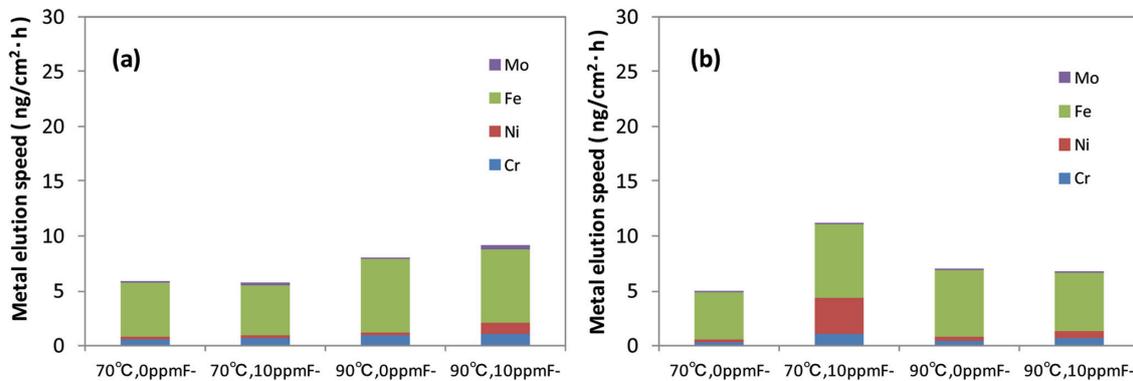
**Figure 4** shows the rate of elution of metal ions measured in a 200-h immersion test using a sulfuric acid solution simulating the PEFC operating environment. The rate of metal ion elution was appreciably small. From the condition of the specimen surface, the elution was judged to be uniform in a passive state, and no local corrosion was observed. Assuming the fuel cell life to be 10 years, if the rate of metal ion elution shown above is maintained for 10 years, the decrease in sheet thickness in the same period will be approximately 1 μm, which is negligibly small. Incidentally, with any of the



**Fig. 2** Contact resistance between CP and developed stainless steel (NSSMC-NAR-316BC), compare to conventional stainless steel or gold



**Fig. 3** SEM image of developed stainless steel (NSSMC-NAR-316BC) after ferric chloride etching



**Fig. 4** Metal elution amount immersing 200 hours in pH3 aqueous solution adjusting sulfuric acid adding fluoride ion, aqueous solution amount per 100 mm<sup>2</sup> : 12 mL  
(a) Developed stainless steel (NSSMC-NAR-316BC), (b) SUS316L

specimens of 316BC, the amount of boride elution was below the detection limit. **Figure 5** shows the contact resistance values measured before and after the immersion test. While the contact resistance of SUS 316L increased after the immersion test, that of 316BC remained low throughout the test. This is attributed to the fact that  $M_2B$  keeps a low contact resistance.

**Figure 6** shows the anodic polarization behavior in a sulfuric acid solution simulating the PEFC operating environment. In the excessively passive potential region of stainless steel nobler than 0.95 V vs. SHE, the dissolution of  $M_2B$  occurs and hence, the anodic current density of 316BC is higher than that of SUS 316L without the precipitation of  $M_2B$ . In the potential region less noble than 0.8 V vs. SHE, on the other hand, the passive behavior of 316BC is as stable as that of SUS 316L. When a stainless steel sheet is kept in a high-potential region exceeding 1 V vs. SHE for a long period, the excessively passive dissolution becomes conspicuous. Even the use of 316BC in such a region should preferably be avoided. Nevertheless, 316BC can safely be kept in the said region intermittently for a short period or used under the normal PEFC operation conditions even for a long period.

**3.2 Mechanical properties**

**Table 1** shows the principal mechanical properties of the newly developed foil (316BC) subjected to bright annealing in comparison

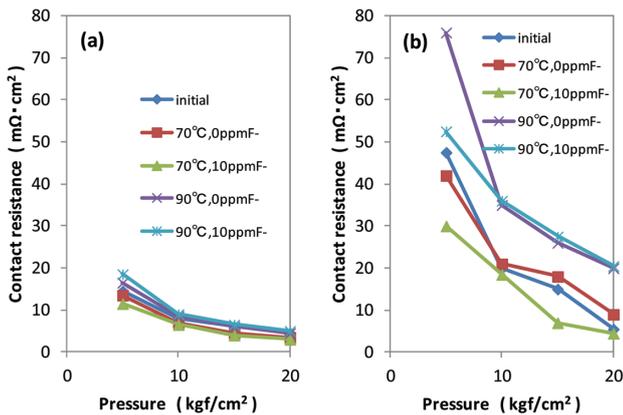
with those of SUS 316L. 316BC is superior in tensile strength to SUS 316L, although it is slightly inferior in yield strength. While the elongation of 316BC is smaller than that of SUS 316L, the yield strength, tensile strength, and elongation of 316BC all meet the standards for SUS 316L. It has been confirmed that in the actual press forming of separators, 316BC offers as good a groove form as does SUS 316L, although it depends more or less on the separator design.

**3.3 Durability of press forming dies**

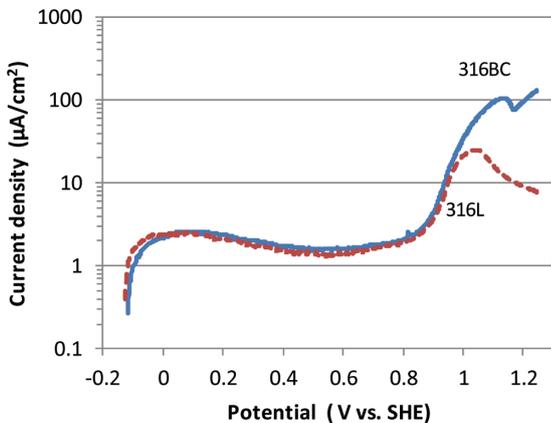
In progressive press forming of the newly developed foil (316BC), damage to the dies due to  $M_2B$  dispersed in the foil is most feared.  $M_2B$  is hard enough to accelerate the wear of the dies. Therefore, we evaluated the amounts of die wear at the part subject to the largest amount of wear using a cyclic punching test of various specimens. As shown in **Fig. 7**, although 316BC causes a larger amount of wear of the punching die than SUS 304 and SUS 316L, the amount of wear can be significantly reduced by selecting a suitable material for the punching die. Using a suitable die material, 316BC could be punched more than 200,000 times without reconditioning the tool tip. In addition, it was confirmed that even after 220,000 separators were subjected to stretch forming, the amount of die wear was negligible. By previously deciding the life of dies and implementing periodical repairs, it is fully possible to apply progressive press forming to 316BC on a mass-production basis.

**3.4 Weldability**

If separators can be formed by welding, the freedom of separator design increases dramatically. Therefore, using Yb laser, we evalu-



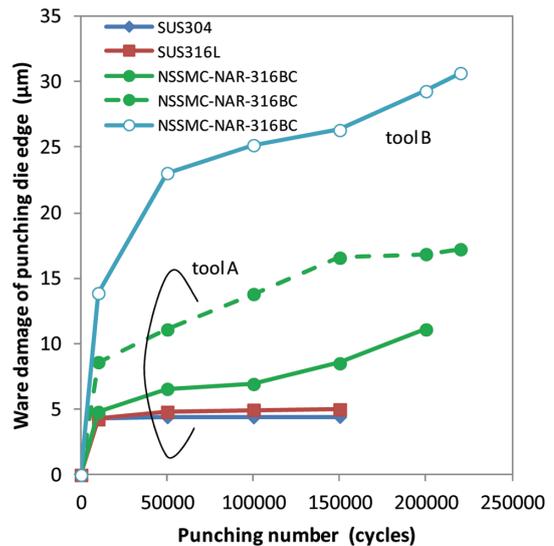
**Fig. 5** Contact resistance before and after immersing 200 hours in pH3 aqueous solution adjusting sulfuric acid adding fluoride ion (a) Developed stainless steel (NSSMC-NAR-316BC) (b) SUS316L



**Fig. 6** Anodic polarization curve in 90°C, air babbling, pH3 aqueous solution adjusting sulfuric acid

**Table 1 Characteristics of developed stainless steel (plate thickness 0.116 mm)**

Steel	Item	Yield strength (N/mm <sup>2</sup> )	Tensile strength (N/mm <sup>2</sup> )	Elongation (%)	Erichsen (mm)
NSSMC-NAR-316BC		279	660	47.6	9.7
SUS 316L	Representative	281	592	54.5	12.3
	Standard	≥ 175	≥ 480	≥ 40	-



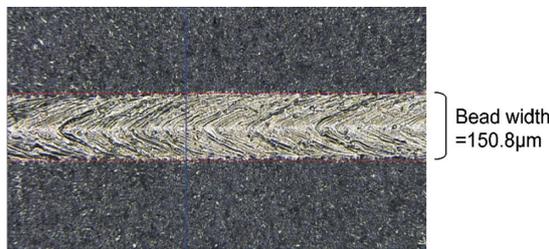
**Fig. 7** Wear damage difference between steel and punching die material Evaluating material thickness: 0.126 mm

ated the weldability of 316BC separators which were press-formed and subjected to the etching for surfacing conductive metallic precipitates in ferric chloride solution. **Figure 8** shows the appearance of a weld bead and the profile of a cross section. Because of a small heat input, the bead is only approximately 150 μm in width and 5 μm or less in height and has a smooth surface. In addition, from the results of an intergranular corrosion resistance test carried out in accordance with JIS G 0575, it was confirmed that the corrosion resistance of the weld zones was good and that the newly developed foil could be subjected to laser welding.

#### 4. Operation of FC Using NSSMC-NAR-316BC

##### 4.1 Characteristics of power generating operation of single-cell FC

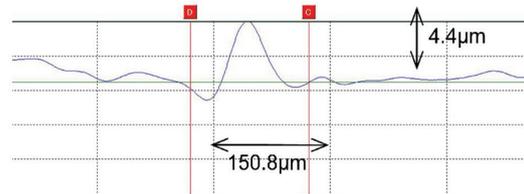
We evaluated the performance of the newly developed foil applied to an actual FC system. **Figure 9** shows an example of long-period operation of a single-cell FC. The FC was operated with the current kept constant under these conditions: temperature 70°C, current density 0.5 A/cm<sup>2</sup> (separator effective reaction area: 70 cm<sup>2</sup>).



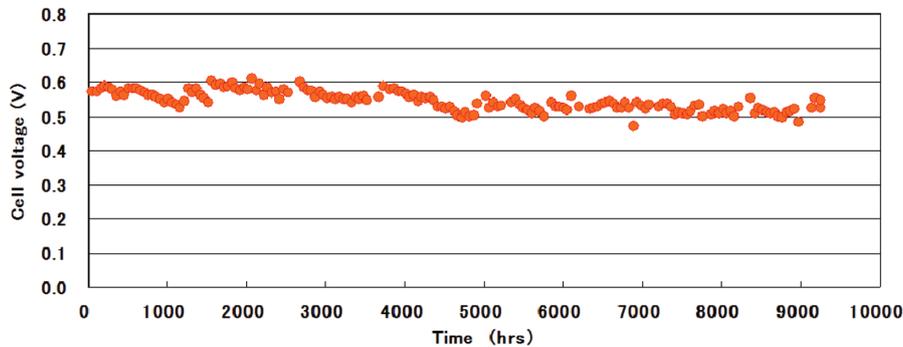
Because of a long-period operation, the FC operation was interrupted several times by an unexpected external factor. Even so, the FC achieved a cumulative operating time of 9,300 h. Although there was a small fluctuation of output due to the start-stop operation, the output could be kept appreciably stable. The drop of output during the FC operation was approximately 5.4 μV/h. **Figure 10** shows the results of observation of the separator surfaces after the FC operation. The surfaces were completely free from discernible local corrosion, including pitting. In addition, it was confirmed that M<sub>2</sub>B had been finely dispersed in the surface layer of separators.

##### 4.2 Metal elution during FC operation

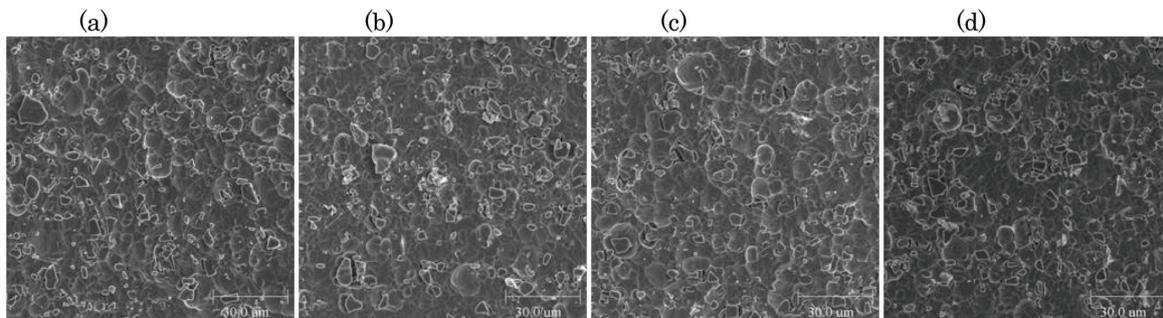
It is often pointed out that metallic separators have the problem of elution of metallic ions. For 316BC separators, we measured the concentration of metallic ions eluted into the water produced during the FC operation. A single-cell FC with 316BC separators was operated at 70°C with the current density kept at 0.5 A/cm<sup>2</sup> and the time-serial change in metallic ion concentration of the anode-side water and cathode side, respectively. The measurement results are shown in **Fig. 11**. Right after the start of FC operation, tens of ppb levels of



**Fig. 8** Ytterbium laser welding surface and cross sectional morphology  
Shielding gas: Nitrogen, voltage: 3.6 V, speed: 12 m/min



**Fig. 9** Single PEFC cell voltage applied the developed stainless steel bipolar plates  
Operating conditions: 70°C, 0.5 A/cm<sup>2</sup>, reaction area 70 cm<sup>2</sup>



**Fig. 10** Developed stainless steel (NSSMC-NAR-316BC) surface after operating 9,300 hours  
(a) Anode side MEA contacting area, (b) Anode side hydrogen flowing area, (c) Cathode side MEA contacting area, (d) Cathode side air flowing area

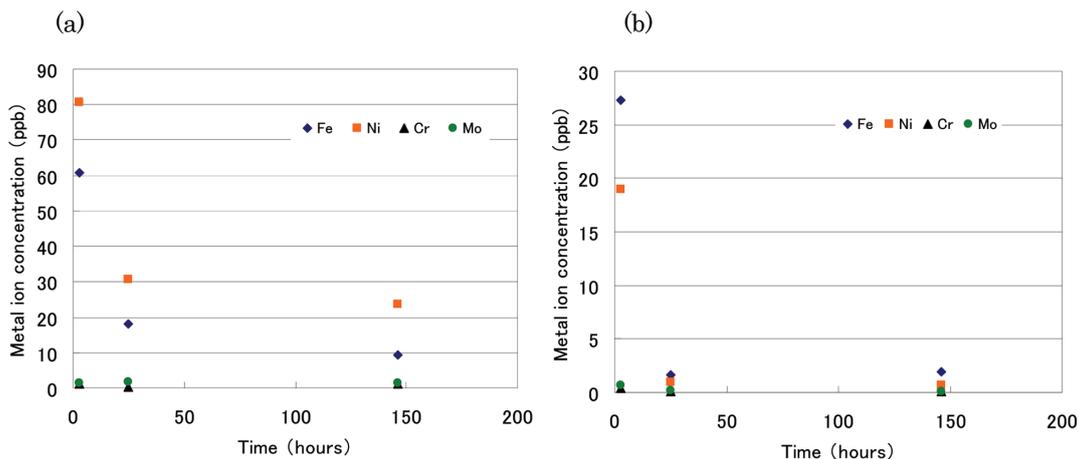


Fig. 11 Metallic ion concentration in drain water  
 (a) Anode side water, (b) Cathode side water, Operating conditions: 70°C, 0.5 A/cm<sup>2</sup>, Reacting area 70 cm<sup>2</sup>

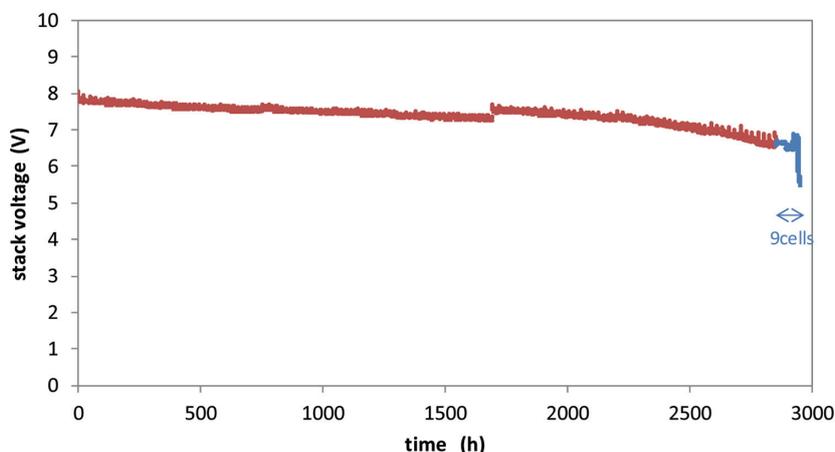


Fig. 12 10 cells PEFC stack voltage applied the developed stainless steel bipolar plates  
 Operating conditions: 70°C, 0.1 A/cm<sup>2</sup>, Reacting area 100 cm<sup>2</sup>

iron and nickel ions were detected. In tens of hours, however, the levels dropped to approximately one-half. After that, they dropped still more. The metallic ion concentration at the cathode side is lower than that at the anode side, which is attributed to the effect of dilution of the concentration by the water as a product of the cell reaction. The concentration of metallic ions in the water discharged from the anode was also measured with a cell operated for more than 3,000 h. The concentration was found appreciably low. From that fact, it was confirmed that the 316BC separators maintain their passive state stably during the FC operation.

**4.3 Long-time FC operation using FC stack**

With an FC stack composed of multistage cells, a decline in FC performance due to a certain factor can occur as described later. With the aim of clarifying the presence or absence of any correlation between the cause of cell deterioration and the decline in FC performance that can hardly be evaluated with single cells, we repeatedly carried out a long-time performance evaluation test of 10-cell stacks on a laboratory scale. The main causes of a decline in stack performance observed with multistage cell stacks are: (1) difference in temperature between cells and uneven temperature within cell plane, (2) uneven gas distribution between cells, (3) local abnormal rise in potential within cell plane due to local fuel gas shortage during start or stop of FC operation, (4) local wetting phenomenon (flooding)

caused by water produced by cell reaction, and (5) blocking of gas passage. All those are observed as a decline in performance of the catalyst, catalyst carrier, and/or polymer membrane. In many cases, they progress irreversibly.

Figure 12 shows the time-serial change in voltage of a 10-cell FC stack using separators formed from 316BC. The FC was operated at 70°C with the current density kept at 0.1 A/cm<sup>2</sup>. The operation was repeated for cumulative 2,000 to 3,000 h. Although any drop of voltage due to metallic ion elution from the separators did not occur, a voltage drop due to a decline in MEA performance did occur, which has much to do with the FC operation conditions.

In applying stainless steel separators, perforations in the separators caused by corrosion and accelerated deterioration of the MEA caused by elution of metallic ions are feared most. As long as stainless steel is used for separators, the elution of metallic ions in passive state is unavoidable. On the other hand, the accelerated deterioration of MEA is extremely difficult to check. However, we intend to make continued efforts to be able to present evaluation results that will eliminate the above fears.

**5. Conclusion**

As a material for PEFC separators, we developed a new stainless steel foil, MSSMC-NAR-316BC, having good corrosion resistance

and high conductivity. In addition, on the basis of results of an evaluation of press forming and a study of dies, we established a press forming system for mass production of 316BC separators. The newly developed 316BC is the world's first metallic material for PEFC separators that need not require any costly and sophisticated surface treatment. In addition, it has good weldability. Thus, it will increase the freedom of design of FC separators.

#### Acknowledgments

The present development was partly entrusted to us by NEDO. We wish to express our heartfelt thanks to all the people concerned.

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