A Novel Evaluation Method of Powder Electrocatalyst for Gas Evolution Reaction

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Graphical Abstract

Forced-Flow Method

A-A

Rib Channel

Membrane

Catalyst attached area

Flow direction

Time Zero Analysis (t=2-10 ms)

Pressure-bonded electrode by CIP
A novel evaluation method of powder electrocatalyst
for gas evolution reaction

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Abstract

The evaluation of the powder catalyst activity is essential for the development of high-performance electrocatalysts for water electrolysis. However, the gas generated therein interferes with the accurate measurement by covering the catalyst surface. In this study, we have developed a new evaluation method for a powder oxide electrocatalyst using an alkaline electrolyte. This measurement and analysis method can obtain the intrinsic catalyst activity with no influence from the generation of bubbles. To establish this method, the electrolyzer for evaluation adopts the interdigit flow field for the forced-flow method, and the two methods, namely time zero analysis and flow rate infinitization analysis, were demonstrated by the extrapolation of the measurement data. As a result, the combination of forced-flow method and time-zero analysis is the best evaluation method for the determination of kinetic current at time zero as the equivalent of the no-generation gas condition.

Keywords: water electrolysis, powder oxide catalyst, activity evaluation method, electrolyzer evaluation.
1. Introduction

Water electrolysis is one of the most important hydrogen production methods for the spread of power-to-gas systems because of zero CO₂ emissions from the renewable energy source, at present.¹,² Performance evaluation is an indispensable factor in the development of gas evolution reaction catalysts for water electrolysis. Many synthesized gas evolution reaction catalysts for alkaline water electrolysis (AWE), proton exchange membrane water electrolysis, and anion exchange membrane water electrolysis are powders that are used in the material development stage. However, accurate evaluation of the powder catalyst activity is difficult owing to the existence of certain problems.

In many cases, the performance of the powder catalyst has been evaluated using a rotating disc electrode (RDE).³–⁶ However, it is difficult to measure the intrinsic catalyst performance by RDE evaluation because gas generation due to the oxygen or hydrogen evolution reaction (OER or HER, respectively) continuously occurs inside the catalyst coating layer on the electrode. Furthermore, the generated gas remains at the center of the catalyst-coating disc area during rotation. The gas-covered catalyst loses OER or HER activity, and the effective catalyst utilization ratio of the catalyst decreases. This results in a decrease in performance or causes noise in the measurement data. In particular, in the catalyst durability test of antimony-doped tin oxide supported iridium powder using RDE,
El-Sayed et al. reported that the decrease in the performance in the potential cycle was caused not by catalyst degradation but by the bubbles remaining in the OER catalyst layer.\textsuperscript{7,8} On the other hand, the noise in the electrochemical data increases at a low rotation speed.\textsuperscript{9,10} It exists even upon using the ring of the rotating ring disc electrode (RRDE) to prevent the gas from remaining at the center of the electrode.\textsuperscript{10} In addition, the catalyst layer of RDE contains a binder, such as Nafion, and whose existence also causes a decrease in the performance.

Pressure-bonded electrodes using cold isostatic pressing (CIP) have been reported as another evaluation method for metal oxide powder catalysts.\textsuperscript{11} This method did not require any binder and no high catalyst–substrate adhesion occurred. Therefore, it showed a better performance compared with the RDE. However, this method too has some concerns. Firstly, in the fabrication process of this electrode, heat treatment at 800 °C was conducted for calcination. This treatment has the potential to change the electrocatalytic properties in some cases. Secondly, the CIP device is not general or common equipment for catalyst researchers, furthermore it is large and expensive in some cases. Hence, it is difficult to use this device widely. The catalyst evaluation method should instead be a simple process.

The aforementioned situations necessitate the development of a reliable and simple
evaluation method for the activity and durability measurement of gas generation catalysts.

In this study, we developed an evaluation method for a gas evolution powder catalyst in order to solve the abovementioned problems. This method used a 1 cm² component evaluation standard electrolyzer\textsuperscript{12} with an interdigit flow field for the \textit{forced-flow method}. The measurement and analysis using this method can eliminate the effect of bubbles and measure the intrinsic performance of the catalyst using a simple method.

2. Experimental

2.1 Membrane electrode assembly (MEA) fabrication and operation condition

The anode catalyst evaluated as reference catalyst was LaNiO₃ (LNO, (Synthesis method: Supplementary information S1)).\textsuperscript{13, 14} The cathode catalyst and membrane were Pt/C (TEC10E50E, Tanaka Kikinzoku Kogyo, Japan) and Zirfon® (Perl UTP 500, Agfa, Belgium), respectively. The anode catalyst ink was prepared by mixing 20 mg catalyst and 1000 μL ethanol with a homogenizer for 3 min. A masking tape with five rectangular holes (1 × 8 mm² each) was applied to the membrane, and 100 mL anode catalyst ink was dropped on the masking tape. After drying at 25 °C for 30 min, the masking tape was peeled off. The loading of LNO was 1.2 mg cm⁻². The cathode catalyst ink was prepared by mixing 0.345 g Pt/C, 0.410 g ultra-pure water, 2.92 g Nafion solution (5 %, Sigma
Aldrich, US), and 2.42 g 1-propanol using a ball mill (LP-1, Ito Seisakusho Co. Ltd, Japan) at 200 rpm for 30 min. The catalyst-coated substrate as the cathode was fabricated by applying the ink on carbon paper (Sigracet 39BC, SGL carbon, Germany) using a bar coater at 0.5 mg Pt cm$^{-2}$ loading.

Figure 1(a) shows the component evaluation test electrolyzer$^{12}$ used in this study. This electrolyzer with 10 × 10 mm$^2$ geometric electrode area was equipped with the reference electrode. The reference electrode was connected to the membrane on the side of the electrode, as shown in Fig. 1(b). This electrolyzer had a precise pressure control system for the electrode area.

In the cell composition, the interdigit and parallel types of flow fields were applied to the anode and cathode, respectively. For comparison, a pattern using the parallel type for the anode as the evaluation side was also fabricated. The clamping pressure to the membrane was set at 2.5 MPa. Ag/AgCl (electrolyte: saturated KCl solution) was used as the reference electrode. The anode and cathode chambers were circulated in a 7 M KOH solution at 30 °C. The flow rate of the anode was set at 10, 30, 50, and 100 mL min$^{-1}$, and that of the cathode was fixed at 5 mL min$^{-1}$.

2.2 Forced-flow method
The flow field applied to the evaluation electrode side in the electrolyzer was of interdigit type, in order to harness its remarkable feature of eliminating the gas generated inside the catalyst layer. Figures 1(c) and (d) show the catalyst-applied area and the solution-flow direction in the interdigit flow field. The catalyst layer is only applied to the rib area of the interdigit flow field. In this structure, the solution to be introduced into the flow field flows into the catalyst layer by force because the catalyst layer is held tightly between the membrane and the rib of the flow field. The forced solution that flows into the catalyst layer extrudes the gas generated from the catalyst.

2.3 Electrochemical measurements

Electrochemical measurements were conducted using electrochemical impedance spectroscopy (EIS, frequency: 0.1–100 kHz, amplitude: 10 mV) and double potential step (1.5–1.9 V vs. RHE). The MEA pretreatment impressed a constant current of 2 A cm$^{-2}$ for 2 h. Figure 2 shows the double potential step pattern used in this study. The constant potential step measurement, which is the direction of increase, was applied for 10 s per potential. The interval time of chronoamperometry (CA) for each potential was set to 360 s to remove the gas generated inside the catalyst layer. The potential at the interval time was set to 1.50 V vs. RHE to avoid the phase transition of the catalyst by applying a lower
3. Results and Discussion

To eliminate the effect of the generated bubble, a millisecond timescale was used in the analysis after applying the potential, excluding the period of the non-Faradic current caused by the electric double layer charging. The time constant $\tau_{dl}$ to charge the electric double layer can be calculated using Equation (1).\

$$\tau_{dl} = R_s C_{dl}$$ (1)

where $R_s$ and $C_{dl}$ are the solution resistance and double layer capacitance, respectively. In the potential step measurement, it is estimated that a $5\tau_{dl}$ time is required to completely eliminate the effects of the charging current. In this study, $R_s$ and $C_{dl}$ were obtained from EIS measurements. $C_{dl}$ was calculated using Equation (2).\

$$C_{dl} = \frac{1}{2\pi f_{max} R_{ct}}$$ (2)

where $f_{max}$ and $R_{ct}$ are the frequency at the top of the semicircle in the Nyquist plot and the charge transfer resistance, respectively. In the powder oxide catalysts used as reference in this study, the value of $5\tau_{dl}$ was calculated to be between 0.2 and 1.1 ms. Hence, the time used for the analysis of CA was applied to the region above 2 ms in this study.
Figures 3(a) and (b) shows the comparison of chronoamperometry curves at 1.6 and 1.7 V until 100 ms and $i$–$E$ curves at 100 ms for the interdigit and parallel flow fields for LNO, respectively. In Fig. 3(a), the current density in the interdigit flow field is higher in the all-time region until 100 ms than that in the parallel flow field, and the curve is slightly gentler at approximately 10 ms. The tendency of the current density to increase in the interdigit flow field is the same for the other potential, as shown in Fig. 3(b). These two characteristics are considered to be the effects of the *forced-flow method* using the interdigit flow field. In the case of the parallel flow field, the gas generated from the catalyst surface remained in the catalyst layer because almost all of the solution flowed in the channel. On the other hand, in the interdigit flow field, the generated gas was removed by the forced flow of the solution in the catalyst layer. The differences in the absolute value and the curve for current density between the two flow fields originate from the effective utilization ratio of the catalyst.

The amount of gas in the catalyst layer is not zero, even upon using the interdigit flow path. However, the smaller gas volume in the catalyst layer occupies in the all-time region; the longer the transient time is quite effective for accurate measurement and analysis. The purpose of this measurement is to obtain the kinetic current in the absence of bubbles in the catalyst layer by analysis, as mentioned later. These features improve
the accuracy of the values obtained by measurement and analysis.

Figure 4(a) shows the flow rate dependence of CA for LNO at 1.9 V. The increase in flow rate from 10 to 50 mL min$^{-1}$ in the anode chamber affects the increase in the current density. However, the CAs between 50 and 100 mL min$^{-1}$ almost show the same curves.

In the *forced-flow method* using an interdigit flow field, the increase in flow velocity affects the removal of bubbles in the catalyst layer. This result indicates that the flow rate (> 50 mL min$^{-1}$) is sufficient to eliminate the bubbles in the catalyst layer. In this study, the flow rate of the standard measurement was set to 100 mL min$^{-1}$. Figure 4(b) shows the anode potential dependence of CA for LNO at 100 mL min$^{-1}$. As the anode potential increases from 1.6 to 1.9 V, the current density also increases, and the time required for the current density to be constant increases. These tendencies are due to the increase in the amount of generated bubbles with the increase in anode potential.

In the evaluation of the activity of OER or HER catalysts used in electrolysis, the generated bubbles covering the catalyst surface decrease the effective utilization ratio of the catalyst. Therefore, to accurately evaluate the intrinsic activity of the catalyst, it is necessary to extrapolate the measurement data to obtain the activity in the absence of bubbles. In this study, two analytical methods were investigated to determine the kinetic activity.
3.1.1 Time zero analysis

One method to determine the catalytic activity in the absence of the generated gas in the catalyst layer is to extrapolate from the initial time of gas generation because the charging of the electric double layer on the catalyst surface occurs immediately after applying the voltage. In this method, to eliminate the effect of the double layer charging current on the measurement data, the 2–10 and 10–50 ms time regions were used for analysis.

In this analysis method, the attenuation of current due to the gas generated in the catalyst layer is considered as the change in the diffusion dominant current. The reciprocal of current density \( i \) represents the sum of the reciprocals of the kinetic current density \( i_k \) and the diffusion-limited current density \( i_d \), as shown in Equation (3).

\[
\frac{1}{i} = \frac{1}{i_k} + \frac{1}{i_d} \tag{3}
\]

In this case, Cottrell equation (Equation (4)) can be applied to obtain the diffusion-limited current density as a function of time \( i_d(t) \) at time \( t \).

\[
i_d(t) = \frac{nFDC}{\pi^{1/2} t^{1/2}} \tag{4}
\]

where \( n, F, D, \) and \( C \) are the reaction electron number, Faraday constant, diffusion coefficient, and concentration, respectively. Hence, the reciprocal of the current density is represented by Equation (5).
\[ \frac{1}{i} = \frac{1}{i_k} + \frac{1}{nF \frac{1}{2} C} \left( \frac{t^2}{2} \right) \quad (5) \]

Figure 5 shows the plots of the square root of time versus the reciprocal of the current density for the time-zero analysis of LNO. The potentials in these plots were treated with \( iR \) correction. In this process, the current densities corresponding to the potentials in Fig. 5 were obtained using the interpolation method (Supplementary information S2). In this plot, the y-intercept, which is extrapolated using the approximately straight line between 2 and 10 ms, is equivalent to the reciprocal of the kinetic current density, as shown in Eq. 5. The value at a constant potential is not affected by the change in flow rate from 50 to 100 mL min\(^{-1}\). The kinetic currents at each voltage indicate intrinsic activity on the catalyst surface with neither gas generation nor an effect on the electric double layer charging.

3.1.2 Flow rate infinitization analysis

Another method to obtain the kinetic current is to extrapolate the flow rate condition to infinity. The infinite flow rate, at which all of the electrolyte flows into the catalyst layer owing to the interdigit flow field, means that there are effectively no bubbles in the catalyst layer because all the generated bubbles flow in infinitesimal time. The analysis process of the flow rate infinitization analysis can refer to the method used for the channel.
The diffusion limit current density $i_{\text{L(CFDE)}}$ of the CFDE cell is shown in Equation (6).\textsuperscript{15}

$$i_{\text{L(CFDE)}} = 1.165nFCwA^{-1}(\nu_mD^2x^2/h)^{\frac{1}{3}}$$ (6)

where $w, A, \nu_m, x,$ and $h$ are the width of the working electrode, electrode area, mean flow rate, length of the working electrode, and half of the channel height, respectively.

Substituting $i_{\text{L(CFDE)}}$ for $i_L$ in Equation (1), the kinetic current density can be obtained from Equation (7).

$$\frac{1}{i} = \frac{1}{i_k} + \frac{\nu_m^{-\frac{1}{3}}}{\frac{1.165nFCwA^{-1}(D^2x^2/h)^{\frac{1}{3}}}{1}}$$ (7)

Figure 6 shows the plots of the inverse of the cube root of the flow rate versus the inverse of the current density in the flow rate infinitization analysis of LaNiO$_3$. This plot was also treated the same $iR$ corrected and interpolated, as shown in Fig. 5. In this plot, the 50 to 100 mL min$^{-1}$ region of flow rate was extrapolated. The intercept of the approximate straight line to the axis of the reciprocal of the current density indicates the reciprocal of the kinetic current density in Equation (7). This intercept implies that the flow rate is infinite, and it converges to a single point for different sampling times of CA. This tendency is the same for the different anode potentials.

Figure 7 shows the specific activity with time zero analysis and flow rate infinitization.
analysis of LNO. The sampling times are 2–10 and 10–50 ms for the time zero, and 2 and
10 ms for flow rate infinitization analysis. For each analytical method and time condition
in this study, the time zero method with a sampling time of 2–10 ms shows the highest
performance. In addition, this method exhibits a better performance than other evaluation
methods such as the RDE$^{21-29}$ and the pressure-bonded electrode by CIP.$^{11}$ The evaluation
range of current density in the conventional RDE method was from $10^{-2}$ to $10^{0}$ mA cm$^{-2}$.
On the other hand, the time zero analysis is possible to evaluate up to several tens of mA.
At the same potential, it is possible to obtain the current density from several to several
tens of times higher than the RDE, this different means that the intrinsic activity
measurement with the RDE is underestimated due to the bubbles. In comparison with the
pressure-bonded electrode by CIP, the time zero analysis (2–10 ms) has the advantage of
higher specific activity in the region below 3 mA. Due to this improvement of precision
measurement at low current density region, the time zero analysis can determine the Tafel
region and gradient, which was difficult to decide in the pressure-bonded electrode by
CIP. From these results, the most suitable analytical method and condition for the
evaluation of powder oxide catalysts for AWE is the time zero method with a sampling
time of 2–10 ms. The activity evaluation by combining the forced-flow method and the
time zero analysis is considered to demonstrate the intrinsic performance of the catalyst
under bubble-free conditions. This means that the specific performance metric of catalyst achieves the ideal maximum, similar to the RDE measurement in the evaluation of the OER for polymer electrolyte fuel cells, although the practical operating conditions are different. This method is most useful for comparing the performance of gas evolution powder catalysts and is a powerful tool for developing catalysts using water electrolysis.

4. Conclusions

For an accurate activity evaluation of the gas evaluation powder catalysts, the forced-flow method was developed by comparing the cell and MEA structures, and the time zero and flow rate infinitization analyses were investigated. In the forced-flow method, the electrolyte solution flowed only into the catalyst layer to remove the generated gas. For the time zero analysis and flow rate infinitization analysis, the kinetic activity was obtained by extrapolation. For the evaluation of the intrinsic activity under the condition of no gas generation in the catalyst layer, the time zero analysis using a sampling time of 2–10 ms with the forced-flow method was the most suitable. The obtained activity was specific to the catalyst, and the evaluation method was as simple as RDE. Hence, this evaluation method has the potential to be a common evaluation method.
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Figure 1 Schematic of (a) side view of electrolyzer, (b) front view of anode block, (c) interdigit flow field and (d) cross section of anode center block.
Figure 2 The pattern of double potential step.
Figure 3 (a) The comparison of chronoamperometry at 1.6 and 1.7 V until 100 ms and (b) $i$-$E$ curves at 100 ms for the interdigit and parallel flow fields for LNO.
Figure 4 (a) Flow rate and (b) anode potential dependences of chronoamperometry for LNO.
Figure 5 The plots of the square root of time versus the reciprocal of the current density for the time zero analysis of LNO.
Figure 6 The plots of the inverse of the cube root of the flow rate versus the inverse of the current density in the flow rate infinitization analysis of LNO.
Figure 7 Specific activity with the time zero analysis and flow rate infinitization analysis of LNO.