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Coulometric determination of dissolved hydrogen with a multielectrolytic modified carbon felt electrode-based sensor

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Abstract

A multielectrolytic modified carbon electrode (MEMCE) was fabricated by the electrolytic-oxidation/reduction processes. First, the functional groups containing nitrogen atoms such as amino group were introduced by the electrode oxidation of carbon felt electrode in an ammonium carbamate aqueous solution, and next, this electrode was electroreduced in sulfuric acid. The redox waves between hydrogen ion and hydrogen molecule at highly positive potential range appeared in the cyclic voltammogram obtained by MEMCE. A coulometric cell using MEMCE with a catalytic activity of electrooxidation of hydrogen molecule was constructed and was used for the measurement of dissolved hydrogen. The typical current vs. time curve was obtained by the repetitive measurement of the dissolved hydrogen. These curves indicated that the measurement of dissolved hydrogen was finished completely in a very short time (ca. 10 sec). A linear relationship was obtained between the electrical charge needed for the electrooxidation process of hydrogen molecule and dissolved hydrogen concentration. This indicates that the developed coulometric method can be used for the determination of the dissolved hydrogen concentration.

Key words: multielectrolytic modified carbon electrode (MEMCE); coulometric determination; dissolved hydrogen

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Introduction

Electrodes made of carbon materials have been widely used for electroanalysis because of a wide potential window, a low background current and good stability as compared with metal electrode (Jenkins and Kawamura, 1971; Swain, 1994; Xu et al., 1998; Yano et al., 1998; Hirono et al., 2002; Niwa et al., 2006; McCreery, 2008). In addition, the chemical modification of carbon surfaces has been of significant interest in numerous fields such as electroanalytical chemistry (Murray, 1984; McCreery, 1991). Recently, it has been found that the reduction potential of oxygen can be significantly moved to positive direction by using carbon alloy with nitrogen (Lyth et al., 2009) and graphite (Watanabe et al., 2009). Especially, it has been revealed that nitrogen atoms containing functional groups such as amino group can easily be introduced to the surfaces of the glassy carbon and carbon felt electrodes by the electrode oxidation of ammonium carbamate (Uchiyama et al., 2007), and the electron transfer rates of many inorganic and organic compounds are accelerated to be able to measure excellent redox waves (Wang and Uchiyama, 2008).

In this work, we described that the electrode reduction current of aminated carbon electrode increases with an increase of electroreduction time in sulfuric acid, and also with the increase of the evolution rate of hydrogen gas. This fact indicates that the catalytic sites of the hydrogen evolution increases owing to the electrochemical modification of carbon surfaces by the electrode reduction in sulfuric acid. Furthermore, we obtained the result that the large electrooxidation wave of hydrogen molecule appeared after the aminated carbon electrode was electroreduced in sulfuric acid for a long time. It is well known that bare (or unmodified) carbon electrode has no electrooxidation activity for hydrogen molecule. Thus, this phenomenon realizing the electrooxidation of hydrogen molecule has very important meaning in the field of electrochemistry.

A porous carbon felt material has been used as a suitable working electrode for a coulometric measurements (Uchiyama et al., 1988, 1990). In this coulometric cell, the electroactive species added to the working electrode surface is diffused into the inner part of the carbon felt quickly in a three-dimensional direction, while undergoing the electrode reaction. Therefore, we adopted this coulometric technique to perform the simple and rapid measurement for

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the dissolved hydrogen in aqueous solution.

In this study, a multielectrolytic modified carbon electrode (MEMCE) was fabricated and a coulometric cell using MEMCE with a catalytic activity of electrooxidation of hydrogen molecule was constructed for the determination of dissolved hydrogen. Since the dissolved hydrogen concentration is unstable, the whole-quantity measurement by the coulometric technique is believed to be useful compared with the other electrochemical methods which need calibration.

In general, the cleaning agents, such as sulfuric acid and hydrofluoric acid, are useful for semiconductor manufacturing. However, the great volume of water has been used in order to rinse them. In recent years, the hydrogen dissolved water has been used in the washing processes of wafers. Therefore, the coulometric determination of dissolved hydrogen is considered to be suitable for realizing the simple, rapid and accurate sensing technique of hydrogen dissolved in water.

1 Materials and methods

1.1 Chemical and materials

A porous carbon felt (GF-20-5F, 20 mm diameter, 5 mm thickness) was obtained from Nippon Carbon Co., Ltd. (Tokyo, Japan). The test solution used was 0.1 mol/L phosphate buffer (pH 8.0) to which the bubbling of the hydrogen gas was carried out. Hydrogen gas was prepared using a hydrogen generator (A9150-100, Parker, USA). Potassium hexacyanoferrate(III) and sulfuric acid (H_2SO_4) was supplied by Wako Pure Chemical Industries, Ltd. (Osaka, Japan). Ammonium carbamate was purchased from Merck (Germany).

1.2 Preparation of multielectrolytic modified carbon electrode

In order to perform the constant potential electrolysis, a potentiogalvanostat (HA-151B, Hokuto Denko Co. Ltd., Japan) and a digital recorder (GR-3500, KEYENCE, Japan) were used. A MEMCE was fabricated as follows. The amination of the carbon felt (or glassy carbon) electrode was prepared as previously described (Uchiyama et al., 2007). The aminated carbon felt electrodes were electroreduced in 1 mol/L H_2SO_4 at -1.0 V vs. Ag/AgCl for 20 hr, immediately after the amination was finished, and it was found that the reduction current gradually increased as the electrolysis time increased. During this electroreduction, hydrogen gas was evolved in proportion to the reduction current value. This fact means that the electron transfer rate of hydrogen ion to hydrogen gas is accelerated by forming catalytic electron transfer site by the electrode reduction of aminated carbon felt electrode. This suggests that functional groups containing nitrogen atoms contributing the catalytic electron transfer of hydrogen ion are introduced.

1.3 Electrochemical measurements

Cyclic voltammetric measurements were carried out using an automation polarization system (HZ-3000, Hokuto Denko Co. Ltd., Japan) with a three-electrode cell consisting of a working carbon felt electrode, a Ag/AgCl (3 mol/L NaCl electrolyte) reference electrode, and Pt counter electrode. To observe the electrode oxidation wave of hydrogen molecule, we performed the cyclic voltammetry of hydrogen molecule using the electrolytic cell as shown in Fig. 1. The hydrogen gas was bubbled into H_2SO_4 electrolyte continuously during cyclic voltammetric measurement at a flow rate of 50 mL/min. By employing oxygen electrode, we confirmed that the dissolved oxygen was almost removed by bubbling into hydrogen gas continuously. Amperometric measurement of the dissolved hydrogen was carried out by using the glassy carbon electrode (3 mm diameter) activated by the electrolytic-oxidation/reduction processes described above. The hydrogen molecule was electrolyzed at constant potential (-0.5 V vs. Ag/AgCl) and the oxidation current generated by supplying of hydrogen gas into an electrolyte was monitored. In order to perform controlled potential coulometry, a potentiostat (NPOT-2501, Nikko Keisoku, Japan), a digital coulometer (NDCM-1, Nikko Keisoku, Japan), and a digital recorder (GR-3500, KEYENCE, Japan) were used. The cross section of the controlled potential coulometric cell fabricated is shown in Fig. 2. A multielectrolytic modified carbon electrode (MEMCE, 20 mm diameter, 5 mm thickness) was used as a working electrode. The unmodified carbon felt without

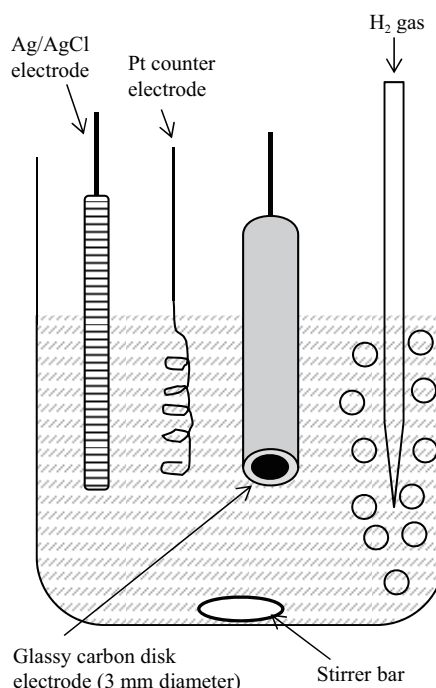


Fig. 1 Schematic diagram of the cyclic voltammetric cell for bubbling hydrogen gas.

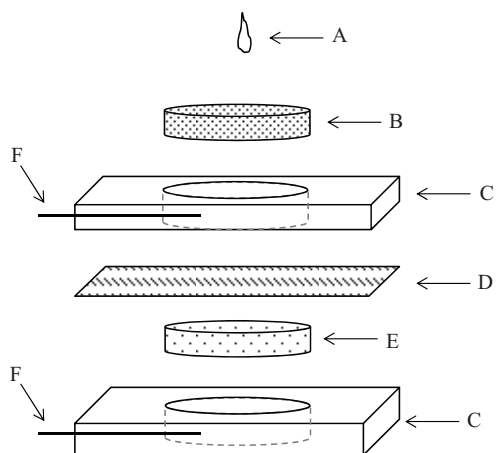


Fig. 2 Configuration of controlled potential coulometric cell. A: sample addition; B: multielectrolytic modified carbon felt; C: acrylic plate; D: ion exchange membrane; E: counter carbon felt; F: Pt lead wires.

any electrochemical treatments was employed as a counter electrode. The MEMCE was separated from counter carbon felt electrode by a cation exchange membrane (CMV, Asahi Glass Engineering, Japan). A platinum wire (0.5 mm diameter) was used as a lead wire. A working MEMCE, was impregnated with 0.1 mol/L phosphate buffer (pH 8.0) solution. Saturated aqueous solution of potassium hexacyanoferrate(III) was employed as a counter solution soaked in the counter carbon felt electrode. The procedure for the coulometric determination is described as follows. First, the electrode potential between a working electrode and a counter electrode was maintained at a constant value by a potentiogalvanostat. Next, an aliquot of the dissolved hydrogen water was added to the center of the working electrode using a micropipet after the residual current reached a constant steady-state value. The concentration of the dissolved hydrogen was calculated from total electrical charges measured by a coulometer.

2 Results and discussion

2.1 Electrochemical measurements for the electrooxidation of hydrogen molecule

A glassy carbon electrode was used to examine the electrocatalytic properties. **Figure 3** shows the monitoring of the electrode oxidation current of unmodified glassy carbon electrode with electrooxidation time in 0.1 mol/L ammonium carbamate aqueous solution. The oxidation current was remarkably increased by applying the electrode potential of +1.1 V (vs. Ag/AgCl). This result suggests that the functional groups containing nitrogen atoms was introduced to the glassy carbon electrode surface by the electrode oxidation of ammonium carbamate aqueous solution using glassy carbon electrode. This modified electrode was named as aminated glassy carbon electrode. The electrochemical performance of the aminated glassy carbon electrode was reported according to the method described

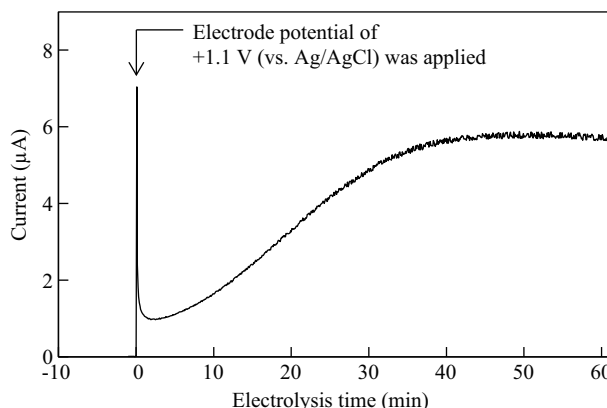


Fig. 3 Variation of the electrode oxidation current of glassy carbon electrode with time at +1.1 V (vs. Ag/AgCl) in 0.1 mol/L ammonium carbamate aqueous solution.

earlier (Uchiyama et al., 2007).

Figure 4 shows the variation of the electrode reduction current of aminated glassy carbon electrode with electroreduction time in 1 mol/L H_2SO_4 electrolyte. This result indicates that the reduction current increases with an increase of electroreduction time, and it can be recognized that the electrolytic modification of the electrode surface continuously proceeds during a long term electrolysis. During the studies of the electrode reduction of the aminated carbon electrode surface, we found that the electrode reduction current gradually increased in the sulfuric acid electrolyte as the electrolysis time increased, and this reaction was accompanied by a vigorous evolution of hydrogen gas. This fact indicates that the electrocatalytic site for the reduction of the hydrogen ion is produced during the electrode reduction in 1 mol/L H_2SO_4 electrolyte. This catalytic site must be generated by the electrode reduction of the nitrogen atoms containing functional groups introduced by the prior electrode oxidation of carbamic acid. Structural studies for the carbon surface are now in progress. The evolution potential of hydrogen gas gradually moved to positive direction until 10 hr reduction, and we

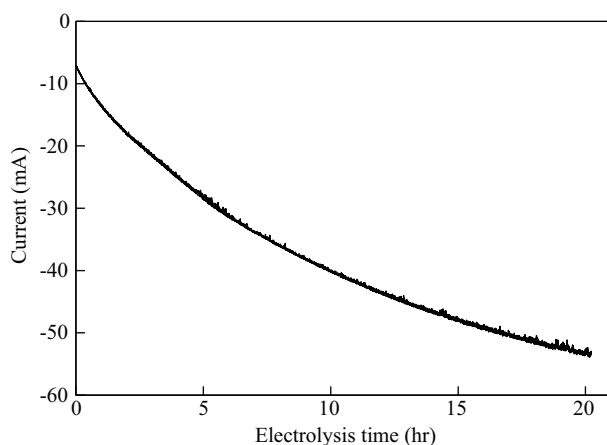


Fig. 4 Variation of the electrode reduction current of aminated glassy carbon electrode with time at -1.0 V (vs. Ag/AgCl) in 1 mol/L H_2SO_4 electrolyte.

chose 20 hr as electrolysis time of aminated glassy carbon electrode in 1 mol/L H_2SO_4 .

Figure 5 shows the cyclic voltammograms of bubbling H_2 in 0.1 mol/L phosphate buffer solution (pH 8.0) obtained by using the aminated glassy carbon electrode electroreduced for 20 hr. The electrode oxidation wave of hydrogen molecule appears in the potential range from -0.6 V to $+0.2$ V. We examined that no oxidation wave of hydrogen molecule was observed when the aminated glassy carbon electrode was used in place of MEMCE. Then, this result means that the catalytic electron transfer site for hydrogen molecule is produced by multielectrolyses of glassy carbon electrode surface. Therefore, it can be recognized that the active site acquires its activity by the electrode oxidation at -0.6 V and lose one by further electrode oxidation at $+0.2$ V.

The cyclic voltammograms of the unmodified carbon felt electrode and the MEMCE in 0.1 mol/L phosphate buffer (pH 8.0) are shown in **Fig. 6** lines A and B, respectively. It can be recognized that no redox waves were observed in a potential range from -1.5 V to $+1.0$ V vs. Ag/AgCl when the unmodified carbon felt electrode is used as a working electrode. However, the hydrogen evolution wave begins at -0.7 V vs. Ag/AgCl in 0.1

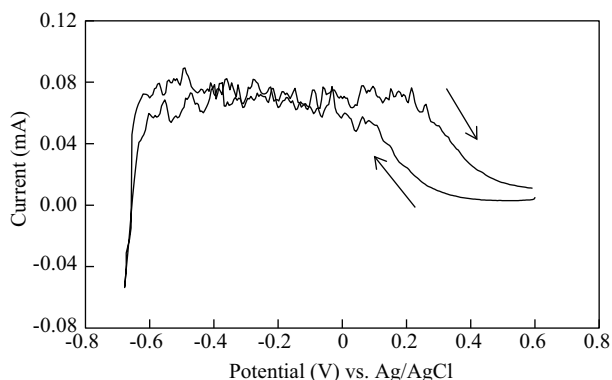


Fig. 5 Cyclic voltammogram of bubbling hydrogen gas in 1 mol/L H_2SO_4 electrolyte. Bubbling rate of H_2 : 50 mL/min; sweep rate: 50 mV/sec.

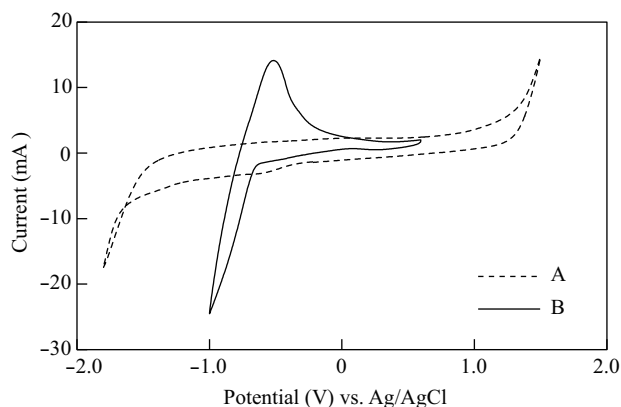


Fig. 6 Cyclic voltammograms of unmodified carbon felt (line A) and MEMCF (line B) electrode recorded in 0.1 mol/L PBS solution (pH 8.0) at sweep rate of 50 mV/sec.

mol/L phosphate buffer (pH 8.0), and the oxidation wave of hydrogen molecule surprisingly appears when MEMCE is used as a working electrode. The redox waves of hydrogen increased by performing twice electrolytic-oxidation/reduction processes. It was confirmed that the catalytic activity of redox waves of hydrogen decreased by adding maleimide which react with hydrazine rapidly in a strong acid. At present, unfortunately, it is unclear what kind of catalytic site for the redox reaction of hydrogen is produced at the carbon felt surface. However, it is confirmed that the electrode oxidation of hydrogen molecule begins at approximately -0.8 V vs. Ag/AgCl, then we tried to measure the electrode oxidation current of hydrogen gas by flowing into an electrolyte.

As shown in **Fig. 7**, the hydrogen gas aeration in a electrolyte solution resulted in the increase of electrode oxidation current of hydrogen molecule and this oxidation current decreased to original baseline by stopping the supply of hydrogen gas because the dissolved hydrogen is gradually escaped from the solution to air. This result shows that the concentration of the dissolved hydrogen can be easily monitored by using MEMCE, and it can be expected that the multielectrolytic modification of carbon electrode can open the way for convenient determination method of hydrogen molecule dissolved in water.

2.2 Effect of applied potential on the sensitivity

To achieve the rapid determination of dissolved hydrogen with high sensitivity, we examined the effect of applied potential to the MEMCE at coulometric cell (-0.4 to $+0.3$ V) on the total electrical charge. **Figure 8** shows the relationship between the applied potential and the electrical charge for the electrooxidation of hydrogen molecule in water. The electrical charge dramatically increased with increasing electrode potential from -0.4 to -0.1 V. On the other hand, the electrical charge decreased sharply when the electrode potential exceeded 0 V. In such an extremely positive potential region and this result indicates that the

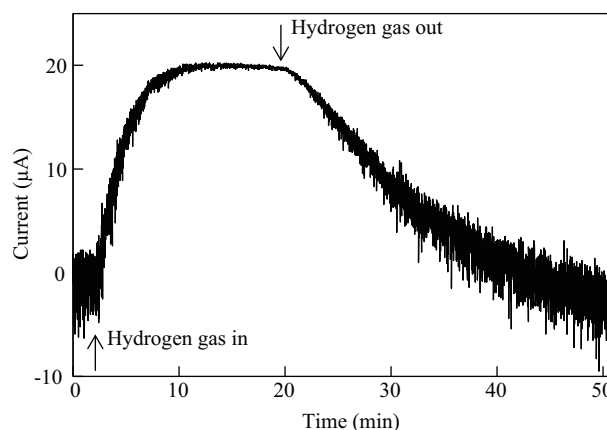


Fig. 7 Typical current response obtained by amperometric monitoring of dissolved hydrogen with a multielectrolytic modified glassy carbon electrode. Applied potential: -0.5 V (vs. Ag/AgCl), aeration rate of hydrogen gas: 50 mL/min.

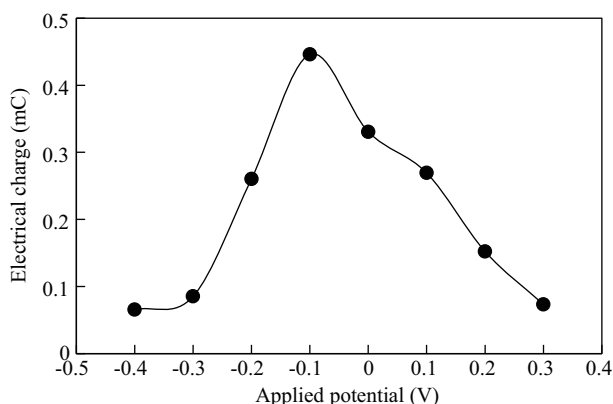


Fig. 8 Effect of the applied potential on electrical charge obtained for the measurement of dissolved hydrogen. Sample volume: 10 μL .

catalytic site on the electrode surface was destroyed by the electrode oxidation. Therefore, the optimal potential to measure the electrical charge was determined to be -0.1 V.

2.3 Coulometric determination of dissolved hydrogen

Figure 9 shows the typical current vs. time curve obtained by the repetitive coulometric measurement of the dissolved hydrogen. The sample solution of hydrogen molecule was prepared by bubbling hydrogen gas into the distilled water. The volume of the added sample was changed from 5 to 20 μL . The oxidation current was actually increased as sample solution volume increases, this fact indicates that current response was generated by the electrooxidation of hydrogen molecule in the sample solution. **Figure 10** shows the relationship between the volume of sample solution and the charge passed through the electrooxidation process of hydrogen molecule. This result shows that the electrical charge was proportional to the added sample

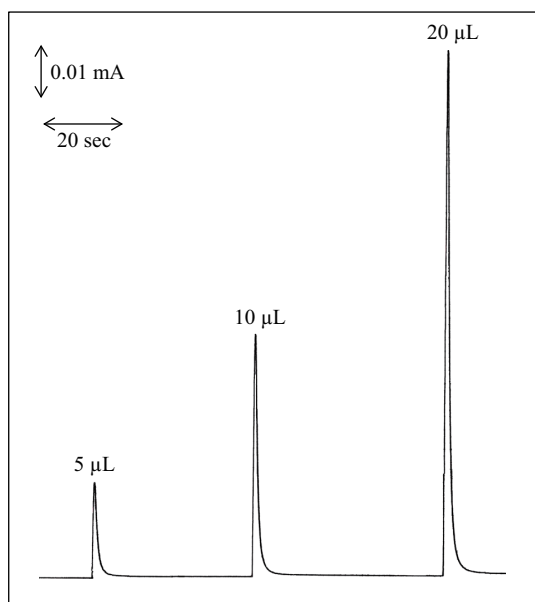


Fig. 9 Typical current vs. time curve of successive addition of each sample volume of dissolved hydrogen.

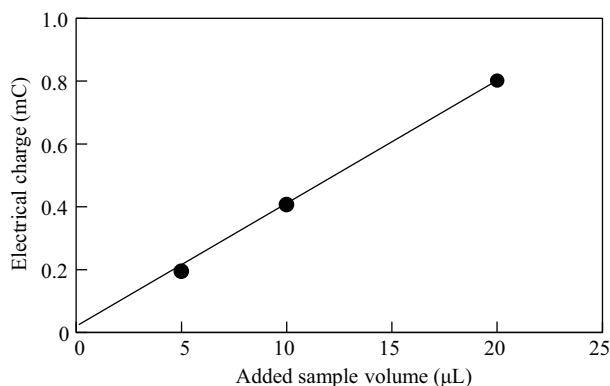


Fig. 10 Relationship between the electrical charge and added sample volume.

volume in the range from 5 to 20 μL with good linearity. Thus, the electrolysis efficiency for the hydrogen molecule is expected to be nearly 100% because extremely rapid determination is realized, compared with the escaping rate of the dissolved hydrogen molecule from the sample solution. Typical current vs. time curve obtained by the repetitive determination of the dissolved hydrogen are shown in **Fig. 11**.

The volume of the added sample is 10 μL . This curve indicates that the determination of dissolved hydrogen is finished completely in a very short time (ca. 10 sec), and no detectable residual current fluctuation appears after the electrolysis is completed. This typical current vs. time curve also indicates that the diffusion of the sample solution takes place very quickly (below 0.5 sec). The relative standard deviation for five successive measurements was 1.9%. **Figure 12** shows the relationship between the electrical charge and the dilution ratio (R_a) of the dissolved hydrogen. The dilution ratio was calculated according to: $R_a = C/C_0$, where, C_0 denotes the concentration of dissolved hydrogen water prepared by bubbling of hydrogen gas and C denotes the concentration of dissolved hydrogen water diluted with arbitrary rates. As shown in **Fig. 12**, the electrical charge was proportional to the

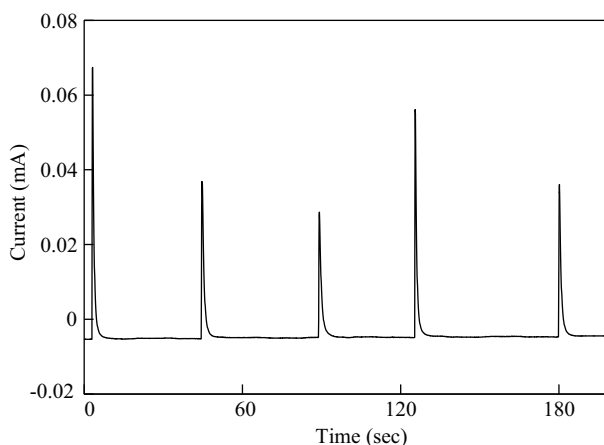


Fig. 11 Current vs. time curve obtained for the successive addition of dissolved hydrogen. Sample volume: 10 μL .

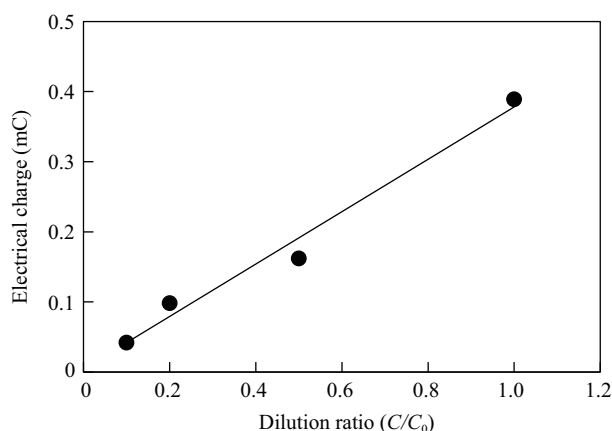


Fig. 12 Relationship between the electrical charge and the dilution ratio of the dissolved hydrogen. The ratio of 1 corresponds to the concentration of the prepared dissolved hydrogen without dilution.

dilution ratio in the range from 0.1 to 1.0 with a good linearity, and this fact supports the idea that the electrolysis efficiency for the hydrogen molecule is nearly 100%. Then, coulometry described here must be very useful analytical method because the accurate and rapid determination of the dissolved hydrogen concentration in water is easily performed.

3 Conclusions

We have succeeded in preparing the MEMCE with chemical activation based on the electrolytic-oxidation/reduction processes for novel kind electrode for the dissolved hydrogen gas determination. The hydrogen evolution wave appears at -0.65 V vs. Ag/AgCl and it is very surprising that the oxidation wave of hydrogen molecule appears by using MEMCE. When the MEMCE was used as the working electrode of coulometric cell, the electrode oxidation current abruptly increased after the dissolved hydrogen water was added to the center of the working MEMCE. The current vs. time curves indicate that the determination of dissolved hydrogen is finished completely in a very short time (ca. 10 sec) and the MEMCE has been proved to be suitable for detecting the dissolved hydrogen. No standardization method for the dissolved hydrogen concentration has been realized until now because hydrogen molecule does not react with the usual oxidizing agent, and needs appropriate catalysts such as platinum for its titration. Moreover, the concentration of the dissolved hydrogen is very changeable and a very rapid method is dispensable for accurate determination. In this respect, the proposed method needs below 10 sec for complete electrolysis of the dissolved hydrogen and is considered to be very useful for the determination of dissolved hydrogen concentration.

References

- Jenkins G M, Kawamura K, 1971. Structure of glassy carbon. *Nature*, 231(5299): 175–176.
- Swain G M, 1994. The susceptibility to surface corrosion in acidic fluoride media: A comparison of diamond, HOPG, and glassy carbon electrodes. *Journal of the Electrochemical Society*, 141(12): 3382–3393.
- Xu J S, Chen Q Y, Swain S M, 1998. Anthraquinonedisulfonate electrochemistry: A comparison of glassy carbon, hydrogenated glassy carbon, highly oriented pyrolytic graphite, and diamond electrodes. *Analytical Chemistry*, 70(15): 3146–3154.
- Yano T, Tryk D A, Hashimoto K, Fujishima A, 1998. Electrochemical behavior of highly conductive boron-doped diamond electrodes for oxygen reduction in alkaline solution. *Journal of the Electrochemical Society*, 145(6): 1870–1876.
- Hirono S, Umemura S, Tomita M, Kaneko R, 2002. Superhard conductive carbon nanocrystallite films. *Applied Physics Letters*, 80(3): 425–427.
- Niwa O, Jia J B, Sato Y, Kato D, Kurita R, Maruyama K et al., 2006. Electrochemical performance of angstrom level flat sputtered carbon film consisting of sp^2 and sp^3 . *Journal of the American Chemical Society*, 128(22): 7144–7145.
- McCreery R L, 2008. Advanced carbon electrode materials for molecular electrochemistry. *Chemical Reviews*, 108(7): 2646–2687.
- Murray R W, 1984. *Electroanalytical Chemistry* (Bard A J, ed.). Marcel Dekker, New York. Vol. 13, 191–368.
- McCreery R L, 1991. *Electroanalytical Chemistry* (Bard A J, ed.). Marcel Dekker, New York. Vol. 17, 221–374.
- Lyth S M, Nabae Y, Morita S, Kuroki S, Kakimoto M, Ozaki J et al., 2009. Carbon nitride as a nonprecious catalyst for electrochemical oxygen reduction. *The Journal of Physical Chemistry C*, 113(47): 20148–20151.
- Uchiyama S, Watanabe H, Yamazaki H, Kanazawa A, Hamana H, Okabe Y, 2007. Electrochemical introduction of amino group to a glassy carbon surface by the electrolysis of carbamic acid. *Journal of the Electrochemical Society*, 154(2): F31–F35.
- Wang X Y, Uchiyama S, 2008. Amperometric glucose sensor fabricated by combining glucose oxidase micelle membrane and aminated glassy carbon electrode. *Analytical Letters*, 41(7): 1173–1183.
- Watanabe H, Yamazaki H, Wang X Y, Uchiyama S, 2009. Oxygen and hydrogen peroxide reduction catalyses in neutral aqueous media using copper ion loaded glassy carbon electrode electrolyzed in ammonium carbamate solution. *Electrochimica Acta*, 54(4): 1362–1367.
- Uchiyama S, Ono M, Suzuki S, Hamamoto O, 1988. Coulometric cell using porous carbon felt. *Analytical Chemistry*, 60(17): 1835–1836.
- Uchiyama S, Umesato F, Suzuki S, 1990. Flow-coulometric detector for uric acid in human urine. *Analytica Chimica Acta*, 230: 195–198.

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