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Electrolytic dissolution characteristics of SIMFUEL in carbonate solutions of high concentration

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ABSTRACT

This work studied anodic dissolution characteristics of a SIMFUEL electrode at several potentials in carbonate solutions of a high concentration at several pHs. The electrolytic uranium dissolution of SIMFUEL was much affected by a corrosion product of UO_2CO_3 generated at the electrode during the dissolution in carbonate solution. The corrosion product distorted the voltammogram of SIMFUEL in the potential region of oxygen evolution and increased the overpotential of oxygen evolution at the electrode. The effective dissolution of the SIMFUEL electrode in a carbonate solution could be obtained at an applied potential such as +4 V (vs SSE) or more which had an overpotential of oxygen evolution high enough to rupture the corrosion product on the electrode surface. The corrosion potential of SIMFUEL decreased with pH in the carbonate solution, and the dissolution rate and current efficiency of the SIMFUEL increased with a decrease of pH in the carbonate solution.

1. Introduction

The dissolution of uranium-bearing oxide mixture waste generated in the front- and back-end of the fuel cycle such as during the stages of ore processing, refinement, fuel fabrication as well as spent nuclear fuel in carbonate media at high pH has recently attracted considerable attention because it is considered to have better inherent advantages with respects to nuclear safe guards, environmental friendliness, and operational safety in the dissolution than the conventional way using a strong and hot acid [1–6]. Uranium can be dissolved in the form of uranyl-carbonato complex with high solubility in carbonate solution at high pH, but most of metal oxides have very low solubilities in such a carbonate solution. Accordingly, uranium can be selectively leached from the uranium mixture waste or spent nuclear fuel in the carbonate solution. The study on the electrochemical dissolution of UO_2 or U-bearing oxide mixture such as spent nuclear fuel is helpful in understanding the dissolution mechanism of UO_2 or uranium-mixture oxide in the carbonate solution.

Many works on the anodic dissolution of UO_2 or SIMFUEL (SIMulated spent nuclear FUEL) have been carried out to date for understanding dissolution or corrosion characteristics of U or transuranium element oxides in aqueous solutions under geological conditions in the case of direct disposal of spent nuclear fuel [7–17]. So the previous works studied mainly the dissolution phenomena of the UO_2 or SIMFUEL electrode surfaces in the bicarbonate, NaCl or $NaClO_4$ solutions at pH 8–10 and near the corrosion equilibrium potential where the dissolution just begins to occur rather than at the high potentials where the dissolution occurs substantially and rapidly. In our previous work [18], intensive dissolution of UO_2 in a carbonate solution could be obtained at an applied potential such as +4 V (vs SSE) or more which was with an overpotential of oxygen evolution high enough to rupture the corrosion product at the electrode surface rather than near the potential for the oxidation of UO_2 grain as UO_2^{2+} before the oxygen evolution. However, the anodic dissolution of UO_2 mixed together with various other metal oxides such as SIMFUEL has to be further evaluated for a better understanding of the dissolution of spent nuclear fuel in strong carbonate solutions at several pHs.

In this work, the anodic dissolution characteristics of a SIMFUEL was studied with cyclic voltammograms in the wide potential ranges with various positive limits which were much higher than the corrosion potential, and their dissolution rates were evaluated at several applied potentials in carbonate solutions of a high concentration at several pHs.

2. Experimental

A SIMFUEL used in this work was fabricated with the metal oxides chosen on the basis of the main components of a PWR spent fuel calculated by the ORIGEN code. Table 1 shows the kinds of chosen elements, their amounts and chemical forms for the SIMFUEL used in this work. The anode used in this work was prepared by connecting a SIMFUEL pellet of 8.2 mm in diameter and 10 mm in length and a stainless steel rod of the same diameter as a terminal lead and inserting them into a Teflon sleeve with tight sealing. The open SIMFUEL pellet surface to be used as the working electrode

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was polished with several steps of emery paper (Nos. 400, 800, and 1200) and 0.5 μm gamma alumina, then cleaned in a sonicator for 15 min, and finally thoroughly washed with ultrapure water. The counter and reference electrodes were Pt wire and Ag/AgCl (Silver–Silver chloride electrode (SSE) in 3 M KCl), respectively. The working electrode was set into a cell of 50 mL without a membrane with the electrode surface being upward to prevent the oxygen gas generated at high potential from covering the surface. The measurements of voltammograms and dissolution rates at the potential range of −1.5 V to +4.5 V (vs SSE) were carried out with a potentiostat (Zahner IM6) in 0.5 M Na_2CO_3 solutions at several pHs controlled with HNO_3 and NaOH. All the voltammograms in this work were measured at a scan rate of 20 mV/s. In order to measure the dissolution rates of uranium at applied constant potentials, the solution was sampled at regular intervals. The concentrations of uranium and other metals in the solution were analyzed by an ICP (Induced Coupled Plasma spectroscopy; Jobiny von JY 38 Plus). The corrosion potential was measured as an open circuit potential after stabilization over 15 h, on switching to open circuit [14]. After the dissolution experiments, a 1 mm thickness of the open surface of the working electrode was cut off for *ex situ* analyses with an electron probe microanalyzer (EPMA, JEOL JXA 8600 with EDX detector) with Scanning Electron Microscope (SEM).

All the reagents used in this work were reagent grade, and they were dissolved, as received, in demineralized water of 18.2 MΩ prepared by a double distillation and one ion exchange (Milli-Q plus). All the experiments were carried out at room temperature.

3. Results and discussion

In order to know the dissolution characteristics of UO_2 of the major component in SIMFUEL, it is necessary to understand the general dissolution mechanism of UO_2. The dissolution of UO_2 begins with the oxidation of stochiometric UO_2 to $UO_{2.33}$, as shown in Eq. (1). Then, the oxidation and dissolution of $UO_{2.33}$ occurs as UO_2^{2+}. The dissolution can be accelerated or be blocked depending on the solution condition affecting the solubility of UO_2^{2+} species in the solution. In the neutral non-complexing solution, the dissolved UO_2^{2+} can change to a corrosion product of $UO_3{\cdot}xH_2O$ at the surface due to the low solubility of UO_2^{2+}, which results in the suppression of the dissolution. On the other hand, in a carbonate solution, UO_2^{2+} forms uranyl carbonate complex ions such as $UO_2(CO_3)_x^{y-}$ that have a much higher solubility in the solution, which accelerates the dissolution with preventing the formation of $UO_3{\cdot}xH_2O$ at the surface. However, in the carbonate solution of high concentration, other corrosion product of UO_2CO_3 can occur on the surface, which also blocks the dissolution [12–18].

$$\begin{array}{ccccc} & & (UO_3\cdot xH_2O)_{surface} & \text{or} & (UO_2(CO_3)\cdot xH_2O)_{surface} \\ & & \uparrow & & \uparrow \\ UO_2 \rightarrow UO_{2+x} \rightarrow UO_{2.33} \rightarrow & & (UO_2^{2+})_{surface\ and\ bulk} & \text{or} & (UO_2(CO_3)_x^{y-})_{surface\ and\ bulk} \end{array} \quad (1)$$

In order to compare the dissolution characteristics of UO_2 electrode itself and UO_2 in SIMFUEL electrode and to know the clear dissolution mechanism of UO_2 in the SIMFUEL electrode, the voltammogram of UO_2 electrode was first measured before using the SIMFUEL electrode. Fig. 1 shows the consecutive cyclic voltammograms in a 0.5 M Na_2CO_3 at pH 11.6 in the potential window of −1 to +1 V. Several peaks were observed, which were similar to the peaks found in the results of other workers [11–16]. The overall cyclic voltammogram shape was not apparently changed with the number of cycles. The peak 1 is ascribed to the oxidation of non-stochiometric species of UO_{2+x} at grain boundary, which is due to incomplete reduction during fuel fabrication, up to $UO_{2.33}$. The peak 2 is ascribed to the general oxidation of stochiometric UO_2 at grain up to limiting composition $UO_{2.33}$, where O^{2-} ions are incorporated into the interstitials sites, and the peak 3 is attributed to the oxidation of the UO_{2+x} layer at the surface made at peak 2 to soluble specie of UO_2^{2+}. Above this peak 3, a corrosion product deposit of $UO_3{\cdot}2H_2O$ can occurs in the neutral non-complexing solution. However, in the carbonate solution like in this work, such a corrosion product does not form. In the carbonate solution of over pH 11 where only CO_3^{2-} exists, the dissolved uranium ion, UO_2^{2+} complexes with the CO_3^{2-} ion to produce the uranyl tri-carbonato complex of $UO_2(CO_3)_3^{4-}$ with a high solubility like Eq. (2). The overall dissolution oxidation of UO_{2+x} in the CO_3^{2-} solution in the peak 3 region can be expressed as shown in Eq. (3). The peak 4 is attributed to the reduction of peak 2, which results in O^{2-} ions

Table 1
Elements chosen as major metal oxides in spent fuel, the chemical oxide form, and amounts of the elements used for the preparation of SIMFUEL.

Elements	wt.%	Oxide form
U	96.33	$UO_{2.18}$
Ce	31.03	CeO_2
Gd	0.42	Gd_2O_3
La	3.75	La_2O_3
Nd	12.35	Nd_2O_3
Pr	3.45	PrO_2
Sm	2.58	Sm_2O_3
Eu	0.48	Eu_2O_3
Y	1.41	Y_2O_3
Mo	12.86	MoO_2
Pd	4.41	PdO
Ru	6.56	RuO_2
Zr	11.15	ZrO_2
Ba	5.79	BaO
Sr	2.37	SrO
Te	1.41	TeO_2

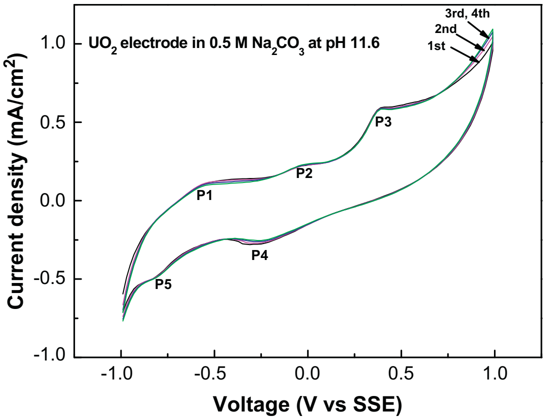

Fig. 1. Consecutive cyclic voltammograms at a UO_2 electrode in the potential window of −1 V to +1 V in a 0.5 M Na_2CO_3 solution.

coming out of the interstitials sites [13,16]. The oxidation–reduction reaction of peak 2 and peak 4 are not changed with consecutive cyclic scans. Peak 5 is attributed to the reduction of $UO_2(CO_3)_3^{4-}$ or the corrosion product at the surface generated after the peak 3 region [13,18].

$$UO_2^{2+} + 3CO_3^{2-} = UO_2(CO_3)_3^{4-}, \tag{2}$$

$$UO_{2+x} + 3CO_3^{2-} + xH_2O = UO_2(CO_3)_3^{-4} + 2xOH^- + (2-2x)e^-. \tag{3}$$

Fig. 2 shows cyclic voltammograms at UO_2 and SIMFUEL electrodes in a 0.5 M Na_2CO_3 solution at pH 11.6 in the potential window of −1.5 to +1.5 V. The peaks observed at the UO_2 electrode in Fig. 1 were not clear at the SIMFUEL electrode and the charging current of the voltammogram at the SIMFUEL electrode was much higher than that at the UO_2 electrode. A broad and relatively big peak was observed at the SIMFEUL electrode near the potential region where the peak due to the oxidation of the non-stochiometric species of UO_{2+x} at the grain boundary in Fig. 1 occurred. The oxidation current of the UO_{2+x} to a soluble specie of UO_2^{2+} after the peak increased with potential and showed a peak near +1.2 V. The reasons for the big charging current and unclear development of peaks at the SIMFUEL electrode can be explained as follows. The big charging current is considered to be ascribed to the presence of metallic particles (Ru, Pd, Mo) as known epsilon-particles and the trivalent rare earth elements acting as dopants in the UO_2 lattice, which result in an increase in electronic conductivity [13,19]. The unclear development of peaks, compared with those at the UO_2 electrode, is considered to be a result of an increase in the non-stochiometric metal oxides in the SIMFUEL matrix and the partial dissolution of the metal oxides of which the SIMFUEL consists such as Mo oxide. Among the metal oxides of the SIMFUEL, Mo oxide was confirmed to be partially dissolved (refer Table 3).

In most papers on the electrochemical dissolution of UO_2 fuel or simulated spent fuel to date, the potential ranges used for cyclic voltammogram measurement have been focused only within about −1 to +0.5 V, because the chosen potential ranges were enough to study their corrosion behavior in the geological disposal environment. Fig. 3 shows cyclic voltammogram in the potential range of −1.5 to +1.0 V in 0.1 M NaCl at the same SIMFUEL electrode used in Fig. 2. The voltammogram was similar to the results obtained by other workers [16]. A peak for oxidation of UO_2 grain to limiting composition $UO_{2.33}$, and a dissolution current of $UO_{2.33}$ to UO_2^{2+}, and a oxygen evolution current following after the peak were observed. An inset box of Fig. 3 shows the voltammogram with an expansion of the positive end of the potential range up to +4.0 V. The oxygen evolution current starting from about +1.0 V linearly increased with the potential without any meaningful change in the cyclic voltammogram up to +4.0 V. However, the currents due to dissolution of UO_2 and oxygen evolution in the carbonate solution at high pH were significantly distorted in the voltammogram at the SIMFUEL electrode. Fig. 4 shows consecutive cyclic voltammograms at the SIMFUEL electrode in a 0.5 M Na_2CO_3 solution at pH 11.6 in the potential window of −1.5 to +4.0 V. In contrast to the results in Fig. 2, a very big peak appeared in the potential between +1.5 and +2.5 V where the oxygen evolution normally vigorously occurred, and the current after the peak decreased and then rose again. The current over +1 V is ascribed to the oxygen evolution with the water split reaction of Eq. (4). The appearance of the big peak and the current decrease with cyclic scan are considered to be a result of the deposition of corrosion product such as UO_2CO_3 which is accumulated to block the electrode surface [10–16,18], resulting in suppression of the oxygen evolution and an increase in the overpotential of the oxygen evolution reaction. It makes the peak in the potential region of the oxygen evolution and makes the potential of oxygen evolution shifted more in the positive direction. Actually, oxygen bubbles were experimentally observed to begin from about +1 V and then decrease after the peak, finally to occur again at the potential range showing the re-rising current after +3 V. The deposition of UO_2CO_3 at the electrode is known to occur according to Eqs. (5)–(7) [8,12,15,17]. The UO_2 is first oxidized into UO_2HCO_3 with the help of bicarbonate of HCO_3^-. Carbonate species are interchangeable among the carbonic acid of H_2CO_3, bicarbonate of HCO_3^-, and carbonate of CO_3^{2-} species depending on the solution pH. Only the car-

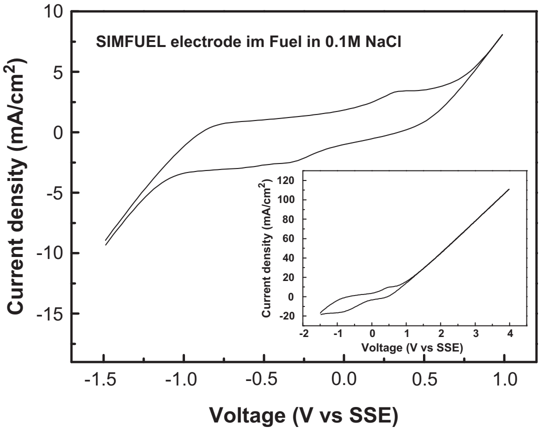

Fig. 3. Cyclic voltammogram at a SIMFUEL electrode in 0.1 M NaCl.

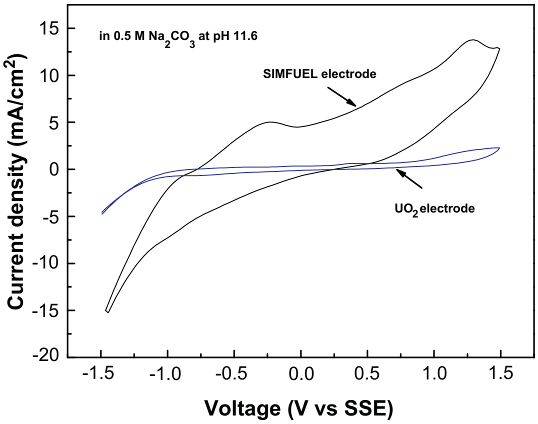

Fig. 2. Cyclic voltammograms at a UO_2 and SIMFUEL electrodes in the potential window of −1.5 V to +1.5 V in a 0.5 M Na_2CO_3 solution.

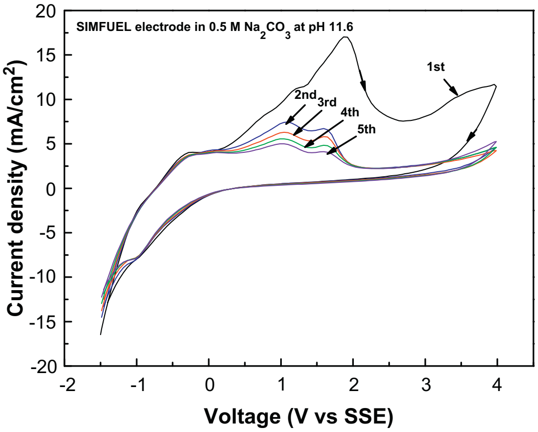

Fig. 4. Consecutive cyclic voltammograms at a SIMFUEL electrode in the potential window of −1.5 V to +4.5 V in a 0.5 M Na_2CO_3 solution.

bonate ion of CO_3^{2-} exists in the 0.5 M Na_2CO_3 solution with pH of more than 11 used in this work. However, proton ions are generated at the potential of over +1 V by the water spilt reaction according to Eq. (4), which makes the CO_3^{2-} ions near the electrode surface acidified, resulting in HCO_3^- ions coexisting together with CO_3^{2-} ions near the electrode surface. Accordingly, $UO_2CO_3{\cdot}xH_2O$ can be deposited at the electrode surface and it succeeds in partially dissolving as $UO_2(CO_3)_3^{4-}$ through Eqs. (5)–(7). In our previous work, $UO_2CO_3{\cdot}xH_2O$ ($x = 0.2$) was confirmed at a UO_2 electrode at +4.5 V in a 0.5 M Na_2CO_3 solution [18]. A peak around −1.0 V in the backward scan in Fig. 4, which was not observed in Figs. 2 and 3 where the corrosion product was not generated at the electrode, is ascribed to the reduction of the corrosion products to UO_{2+x} [13,16,18].

$$2H_2O = O_2 + 4H^+ + 4e^-, \tag{4}$$

$$UO_2 + HCO_3^- = U^V(HCO_3)_{ads} + e^-, \tag{5}$$

$$U^V(HCO_3)_{ads} + OH^- = (UO_2CO_3)_{ads} + e^- + H_2O, \tag{6}$$

$$(UO_2CO_3)_{ads} + 2CO_3^{2-} = UO_2(CO_3)_3^{4-}. \tag{7}$$

Fig. 5 shows the dissolution rate of the uranium of the SIMFUEL electrode at several applied potentials in a 0.5 M Na_2CO_3 solution and the changes of uranium concentration during the dissolutions with time (inset box). The dissolution almost linearly increased with time at each applied potential. *The dissolution rate at 0.0 V was almost zero. In the applied potential range of +0.5 V to +4 V*, the dissolution rate was the highest at +1.0 V and steadily increased with applied potential. Finally, it drastically increased at +4.5 V. The dissolution behavior can be explained with the results of the cyclic voltammogram in Fig. 4. The decrease in the dissolution rate and the sluggish increase after +1 V is considered to be due to the corrosion product generated at the electrode surface. The dissolution rate rapidly increased after +4 V where the oxygen evolution current rose again, as shown in Fig. 4. Table 2 shows the current efficiency of the UO_2 dissolution of SIMFUEL evaluated with the total supplied currents measured at constant applied potentials and the total amounts of dissolved uranium in Fig. 5. The current efficiencies at the potentials where only the dissolution of UO_2 occurs without oxygen evolution were less than 30%, and they slowly increased but they steeply increased up to about 67% at the potential +4.5 V with a sufficiently high overpotential of oxygen evolution. These results mean that an overpotential of the oxygen evolution high enough to break the UO_2CO_3 deposit layer hindering the dissolution on the electrode surface can accelerate greatly the dissolution of UO_2 of SIMFUEL electrode. Fig. 6 shows SEM photographs of the SIMFUEL electrode surfaces after applying constant potentials of +0.5 V (vs SSE) (upper photographs) and +4.5 V (vs SSE) (lower photographs) for 1 h in 0.5 M Na_2CO_3 solution. The electrode surface after applying +0.5 V was almost the same as the fresh electrode without apparent etched marks. The electrode surfaces after applying potentials of until +3.5 V were similar to that after applying +0.5 V, although their photographs are not present in this paper. On the other hand, the electrode surface after applying +4.5 V was severely etched with dissolved trenches. The ion concentrations of the metal oxides doped in the SIMFUEL except Mo were not detected in the solution after the dissolution experiments by ICP. Table 3 shows the change of concentrations of Mo ion in the 0.5 M Na_2CO_3 solution with applying several potentials at the SIMFUEL electrode. MoO_2 is known to be dissolved as MoO_2^{2-} of +6 oxidation state in alkaline solution [1,20]. The Mo ions were detected in the solution in the cases of the corrosion product being not generated or the corrosion product being ruptured at the electrode at a potential with a high overpotential of oxygen evolution, as described in Fig. 4. The co-dissolution of MoO_2 together with UO_2 in the SIMFUEL electrode is considered to affect the unclear development of the oxidation peaks of UO_2 in Fig. 2, as mentioned above.

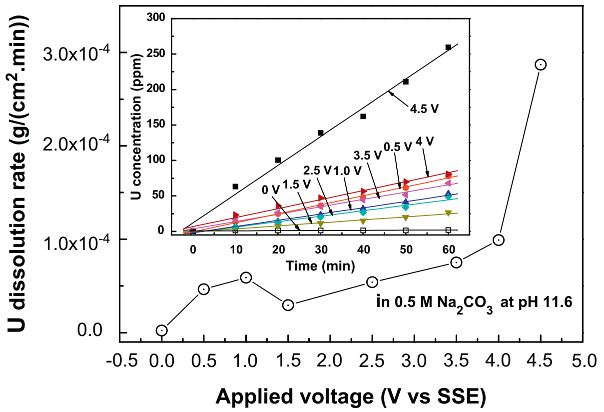

Fig. 5. Dissolution rate of uranium of SIMFUEL at several applied potentials in 0.5 M Na_2CO_3 solution.

Table 2
Calculated current efficiencies of uranium dissolutions of a SIMFUEL electrode at several applied voltages.

Applied potential (V vs SSE)	Current efficiency (%)
+0.5	29.2
+1.5	30.1
+2.5	36.8
+3.5	43.6
+4.0	46.8
+4.5	67.3

Table 3
Change of the concentrations of Mo ion in the carbonate solution with applying several potentials at the SIMFUEL electrode.

Applied potential (V vs SSE)	Mo (ppm)
+1.0	2.3
+2.5	<1.0
+4.0	<1.0
+4.5	2.6

Fig. 7 shows the consecutive cyclic voltammograms at the SIMFUEL electrode in several carbonate solutions at a constant carbonate concentration of 0.5 M with pH 11.7 (A), 10.4 (B), 9.4 (C), and 8.7 (D), respectively. Generally, the currents of UO_2 dissolution and oxygen evolution became less hindered and the repeatability of the cyclic voltammogram with cycles got better with a decrease of pH in the carbonate solution. The dissolution current of UO_2 starting from +0.5 V was gradually distinctly developed up to about +2.2 V, as the solution pH decreased. These results mean that the electrode surface is less blocked due to the corrosion product of UO_2CO_3 with a decrease of pH in the carbonate solution. According to Eqs. (6) and (7), the intermediate species of $UO_2(HCO_3)$ at the electrode increases with a decrease of pH but the generation of UO_2CO_3 in Eq. (6) diminishes because the ratio of HCO_3^- to CO_3^{2-} in the carbonate solution increases with a decrease of solution pH. One interesting thing in Fig. 7C and D is that the reduction peak in the backward scan looks like an oxidation peak. The phenomena

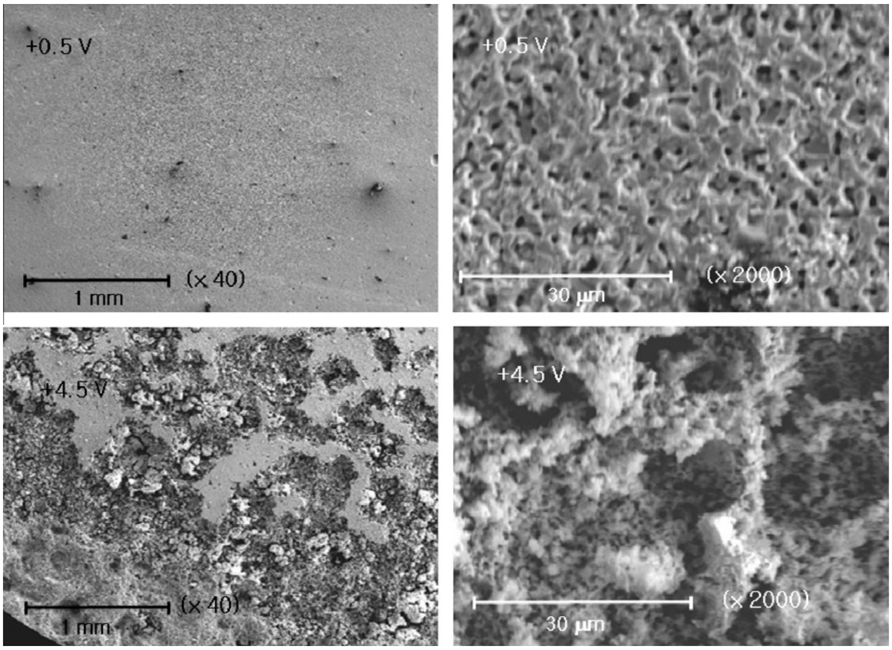

Fig. 6. SEM photographs of SIMFUEL electrodes after applying constant potentials of +0.5 V (vs SSE) (upper photographs) and +4.5 V (vs SSE) (lower photographs) in 0.5 M Na_2CO_3 solution.

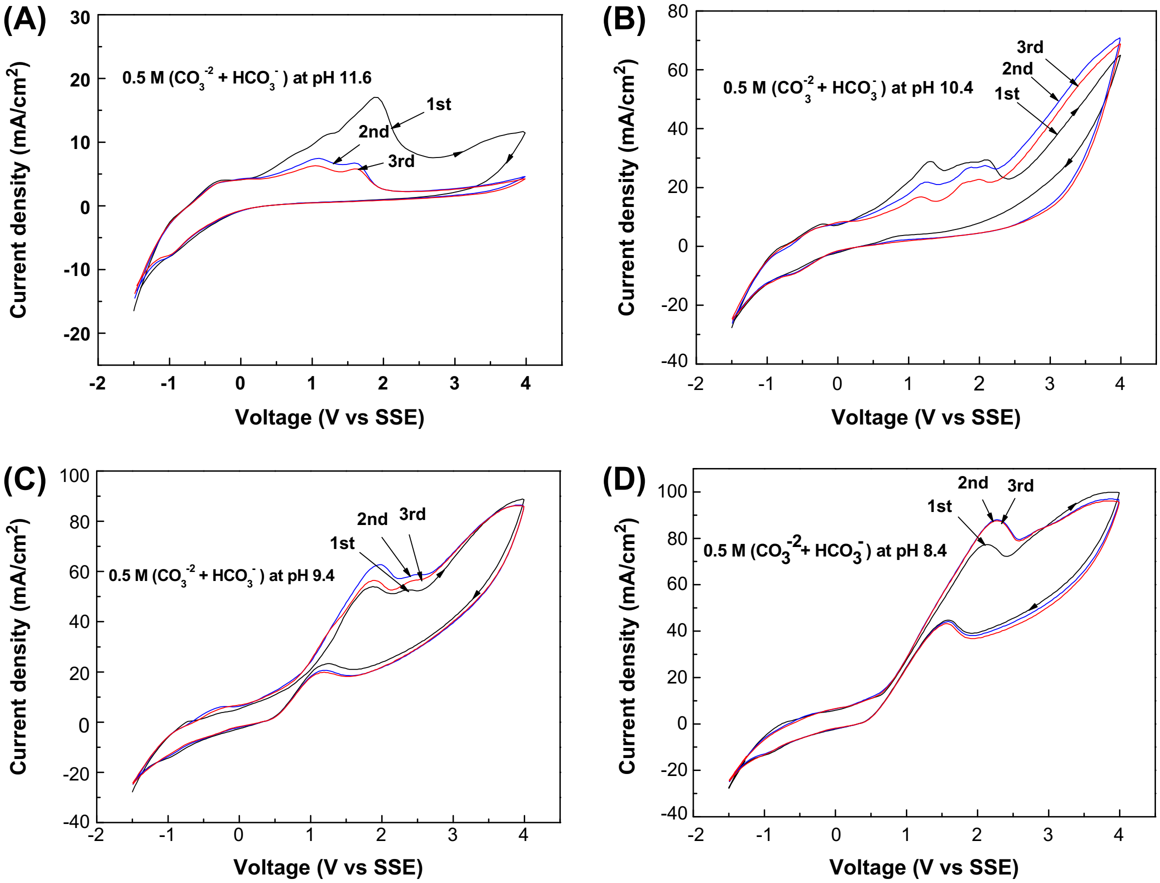

Fig. 7. Consecutive cyclic voltammograms at a SIMFUEL electrode in a 0.5 M carbonate solutions at several pHs.

is due to a visual illusion due to a reduction peak of $UO_2(HCO_3)$ adsorbed in the forward scan at the electrode surface around +2 V in the backward scan, because UO_2CO_3 is hard generated at the electrode in the pH range between 8 and 9 where the carbonate ion exists almost only in the form of HCO_3^- and the concentration of OH^- is much lower, compared with the cases of Fig. 7A and B. The explanation is shown more clearly in Fig. 8 where the positive end of the cyclic voltammogram is +2.5 V. The peaks of oxidation and reduction in the dotted circle in Fig. 8 are almost symmetric.

Fig. 9 shows the corrosion potentials of the SIMFUEL electrode in several carbonate solutions the pH of which were controlled with HNO_3 and NaOH at a constant carbonate concentration of 0.5 M. The corrosion potentials were determined as the values stabilized after about 15 h, on switching to the open circuit [14]. The corrosion potential in the 0.5 M Na_2CO_3 solution at pH 11.6 used in Fig. 2 was about −0.05 V which corresponded to the potential where the dissolution current of UO_2 to UO_2^{2+} began in Fig. 2. The corrosion potential increased with a decrease of pH in the carbonate solution. Fig. 10 shows the dissolution rate of the SIMFEUL electrode at 1.0 V (vs SSE) in 0.5 M carbonate solutions at several pHs. The dissolution rate of UO_2 of the SIMFUEL increased with a decrease of pH in the carbonate solution, and rapidly and linearly increased in the solutions at pH of less than 10 where there was less effect of the deposition of the corrosion product of UO_2CO_3 on the electrode. Table 4 shows the current efficiency of the dissolution calculated with the total supplied currents and the measured amount of dissolved uranium in Fig. 10. The current efficiency decreased with an increase of pH in the carbonate solution because the corrosion product of UO_2CO_3 at the surface suppressed the dissolution more in a higher pH solution.

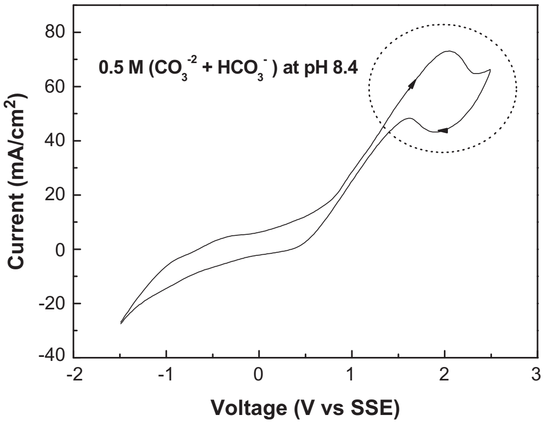

Fig. 8. Consecutive cyclic voltammograms at a UO_2 electrode in a 0.5 M carbonate solution at pH 10.4 .

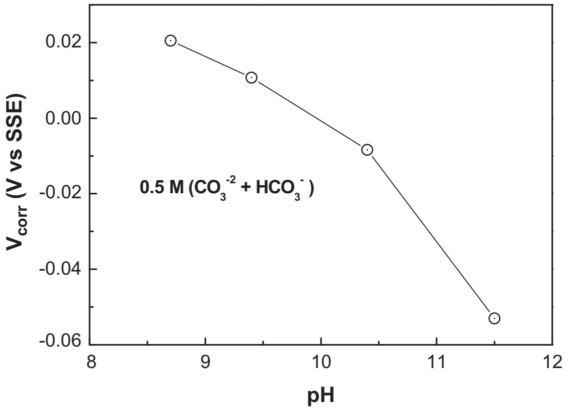

Fig. 9. Change of corrosion potential of a SIMFUEL electrode in carbonate solutions with different pHs.

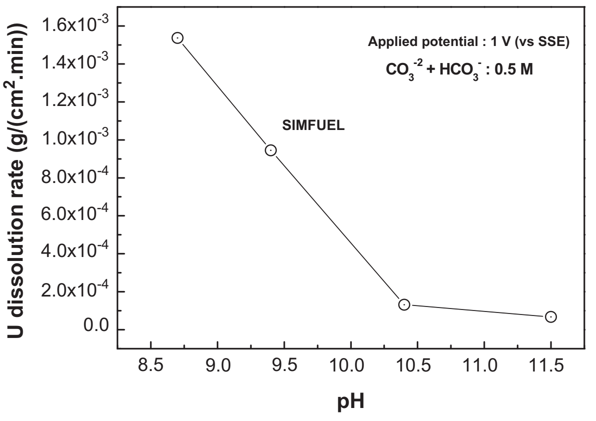

Fig. 10. Dissolution rate of UO_2 of a SIMFUEL electrode at +1.0 V (vs SSE) in carbonate solutions at several pH at a constant carbonate concentration of 0.5 M.

Table 4
Evaluated current efficiencies of uranium dissolution at SIMFUEL electrode in carbonate solutions with constant carbonate concentration of 0.5 M at several pHs.

pH	Current efficiency (%)
9.0	86.7
10.0	88.5
11.0	64.7
11.5	46.1

4. Conclusions

Electrolytic uranium dissolution of SIMFUEL was much affected by a corrosion product of UO_2CO_3 generated at the electrode surface during the dissolution in the carbonate solution. The corrosion product distorted the voltammogram at the SIMFUEL electrode in the potential region of oxygen evolution and increased the overpotential of oxygen evolution at the electrode. Effective dissolution of the SIMFUEL electrode in the carbonate solution could be obtained at an applied potential such as +4 V (vs SSE) or more which had an overpotential of oxygen evolution enough high to rupture the corrosion product on the electrode surface. The corrosion potential of the SIMFUEL decreased with pH in the carbonate solution, and the dissolution rate and current efficiency of the SIMFUEL increased with a decrease of pH in the carbonate solution.

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