Scientific Research

An Investigation on the Electrochemical Characteristics of Ta₂O₅-IrO₂ Anodes for the Application of Electrolysis Process

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ABSTRACT

The electrochemical characteristics of Ta₂O₅-IrO₂ electrodes prepared from different chemical compositions and coating methods were observed by using cyclic voltammetry, potentiostatic polarization, galvanostatic polarization and scanning electron microscopy. The efficiency for chloride oxidation and oxygen evolution processes was not only influenced by the chemical composition but also by the surface morphology of the oxide electrode which was susceptible to the ratio of the two components and the coating method. Ta₂O₅(50)-IrO₂(50) electrodes revealed the highest catalytic activity for the chloride ion oxidation and oxygen evolution reaction because they had the largest effective surface area. The durability of the oxide electrodes in the accelerated life tests was improved as the thickness of the oxide layer increased and the ratio of [IrO₂] to [Ta₂O₅] approached 80/20.

Keywords: *Ta₂O₅-IrO₂ Anode, Chloride Oxidation, Oxygen Evolution, Accelerated Life Test, Corrosion Resistance*

1. Introduction

Mixed metal oxides have been developed as insoluble anodes to protect steel-concrete structures in corrosive atmospheres induced by chloride ions [1,2]. Oxide electrodes used in severe environments require a high chemical stability as well as electrocatalytic activity, whose properties are closely related with their surface exfoliation, electrochemical dissolution, and an increase of electrochemical impedance at the substrate/metal oxide interface [3,4]. For the fabrication of highly stable metal oxide electrodes, interfacial properties at the metal oxide/solution interface should be considered in addition to the chemical composition of the metal oxide itself.

Various types of metal oxides such as PtO_x, IrO_2, Ta_2O_5, SnO_2, Co_3O_4 have been tested to improve the corrosion resistance of mixed metal oxide anodes in recent times [3-14]. According to this previous research, it is generally accepted that a noble metal tends to increase the catalytic activity of the anode while a general metal is added to strengthen the electrochemical stability of the composite catalytic layer. IrO_2 was used as an oxygen evolution catalyst due to its low overpotential for the oxygen evolution reaction, high electrical conductivity and corrosion resistance. Furthermore, an IrO_2 catalyst

layer admixed with SnO_2 or Ta_2O_5 component was very effective to extend the service life in a corrosive environment [3]. On the other hand, Ta_2O_5 is chemically/electrochemically stable and is known to prohibit separation between the substrate and upper catalytic layer by hindering passivation of the Ti substrate metal [5,6]. For this reason, the importance of the mixed metal oxide prepared from IrO_2 and Ta_2O_5 was stressed for the industrial applications including electroplating, electrowinning, electrosynthesis, salt-splitting, descaling, cathodic protection and so on [15-28].

In this research, we investigated the relationship between the electrochemical properties of Ta_2O_5-IrO_2 mixed metal oxides and their surface morphology both in anodic and cathodic process. And, electrochemical characteristics of Ta_2O_5-IrO_2 electrodes prepared from different chemical compositions and coating methods were observed by using cyclic voltammetry, potentiostatic polarization, galvanostatic polarization and surface morphologies from scanning electron microscopy.

2. Experimental

2.1. Electrode Preparation

Commercial 99.5 wt% titanium plate (10 × 40 × 1.5 mm)

was used as a substrate for the electrode and its surface was pretreated by sand blasting and degreasing in a mixed solution of trichloroethylene and acetone prior to the formation of the oxide layer. Each layer of mixed metal oxide was prepared by successive oxide precursor coating, drying, and thermal decomposition. The precursor solution for mixed metal oxide coating consisted of 0.1 M H$_2$IrCl$_6$·4H$_2$O (Strem Chemicals, 99.9%), 0.1 M Ta(OC$_2$H$_5$)$_5$ (Aldrich, 99.98%) and 8.3 M isopropyl alcohol (Duksan Pure Chemicals, 99.8%) as a solvent which was stirred vigorously at room temperature before use. The application of the oxide precursor to the substrate was done by brushing or dipping method for comparison. The metal Ta:Ir mole ratio in the precursor solution varied from 20:80 to 80:20.

The coating layer was dried at 100°C in air for 10 minutes and then thermally decomposed at 450°C for 20 minutes. Also the outermost layer was annealed at the same temperature for 60 minutes for the complete oxidation of the coating. The thickness of oxide was controlled by the repetition of the above successive processes and the loading amount of the oxides weighed 1.07 ~ 1.35 mg/cm^2 after l0 layers were completed.

2.2. Electrochemical Measurements

Mixed metal oxide (area: 2 cm^2), platinum plate (area: 4 cm^2), and SCE (saturated caromel electrode) were used as working, counter, and reference electrodes respectively for the electrochemical analysis. Also the electrochemical characteristics of the mixed metal oxides were measured by using a Gamry PC4/FAS1 potentiostat (Gamry Instruments). Cyclic voltammograms of each mixed metal oxide anode were obtained in 5 M NaCl (Duksan Pure Chemicals, 99%) aqueous solution in the potential range of –0.15 ~ 1.00 V at 50 mV/s. Potentiostatic polarization was performed in 1 M HClO$_4$ (Samchun, 60% ~ 62%) aqueous solution from 1.20 V to 1.42 V with a potential step of 20 mV in the positive direction, where the potential was maintained for 30 seconds at each potential step and thereby steady state currents were recorded as a function of applied potential.

For the evaluation of durability of the mixed metal oxide anodes in real plant environments, galvanostatic polarization experiments were executed with a 2-electrodes configuration – a mixed metal oxide and a platinum plate as an anode and a cathode respectively. The corrosion resistance of each anode was assessed by the potential transition curve in a mixed solution composed of 1M NaCl and 2 M HClO$_4$ at 50°C at a constant current density, 2 A/cm^2, under the conditions that the concentration of chloride ions and overall conductivity of the solution were kept in the range of 32 ~ 38 g/l and 900 ~ 1000 mS

by periodic addition of the concentrated mixed solution.

2.3. Microscopic Observations

The surface morphology and surface composition of the mixed metal oxides were analyzed by scanning electron microscopy (SEM, JSM-5800, JEOL)) and energy dispersive x-ray spectroscopy (EDX).

3. Results and Discussion

3.1. Redox of Chloride Ions

Figure 1 shows cyclic voltammograms of chloride ions in a brine solution at the mixed metal oxide anodes prepared by **(a)** a dipping and **(b)** a brushing method. Redox peaks appearing at 0.3 ~ 0.6 V represent the oxidation/reduction by Ir$^{3+/4+}$ species on the anode surface, indicating that the IrO$_2$ component is the electrochemically ac-
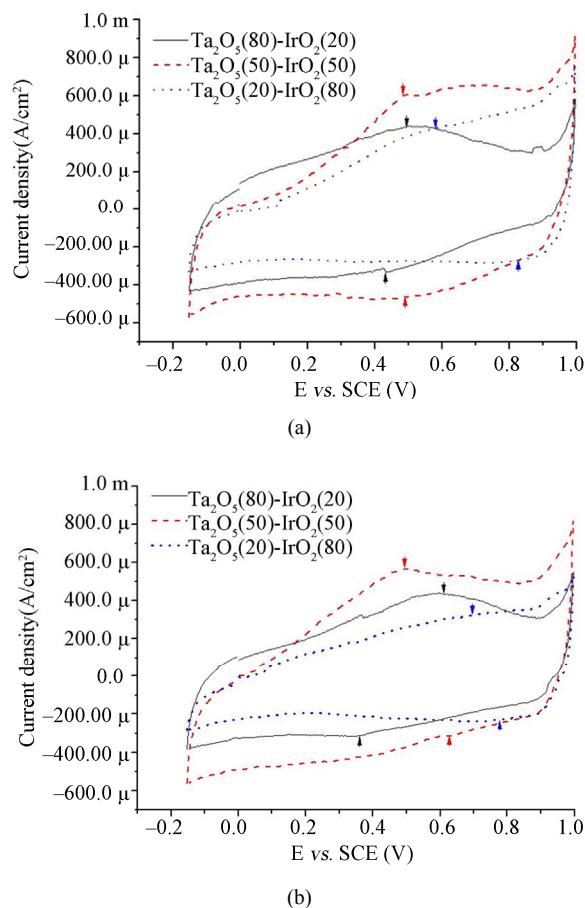

(a)

(b)

Figure. 1. Cyclic voltammograms of Ta$_2$O$_5$-IrO$_2$ electrodes in a brine solution (5M NaCl, pH 2.3, temperature = 80°C) at 50 mV/s. (a) Curves: (solid) Ta$_2$O$_5$(80)-IrO$_2$(20), (dash) Ta$_2$O$_5$(50)-IrO$_2$(50), (dot) Ta$_2$O$_5$(20)-IrO$_2$(80) prepared by a di pp.ing method; (b) Curves: (solid) Ta$_2$O$_5$(80)-IrO$_2$(20), (dash) Ta$_2$O$_5$(50)-IrO$_2$(50), (dot) Ta$_2$O$_5$(20)-IrO$_2$(80) prepared by a brushing method. Number of oxide layers was 4.

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tive species participating in the redox process of chloride ions [29-31]. When the composition of a mixed metal oxide layer changes in the order of $Ta_2O_5(50)$-$IrO_2(50)$, $Ta_2O_5(80)$-$IrO_2(20)$ and $Ta_2O_5(20)$-$IrO_2(80)$, their peak potentials shifted in a positive direction and also their current peaks decreased. This means that the overpotential for chloride ion oxidation increases and the current efficiency decreases in the order of the above composition.

Table 1 summarizes the anodic charge densities at the mixed metal oxide anode prepared by a dipping method and a brushing method. From the change of the anodic charges at the electrodes prepared by different coating methods for the same chemical compositions, mixed metal oxide anodes coated by dipping showed a higher catalytic activity than those by brushing for all the chemical compositions. Comparing the variation of anodic charges among different chemical compositions for each preparation method, it was revealed that the Ta_2O_5-(50)-$IrO_2(50)$ electrode had the highest anodic charge density and the $Ta_2O_5(80)$-$IrO_2(20)$ electrode was comparable to or a little higher than the $Ta_2O_5(20)$-IrO_2-(80) electrode.

Since electric charge is proportional to the electrode surface area, we can say that the dipping method effectively increases the active area of the catalyst taking part in the electrochemical oxidation reaction of chloride ions more than by brushing. Therefore, it was estimated that the $Ta_2O_5(50)$-$IrO_2(50)$ electrode had the largest catalytic area and the $Ta_2O_5(80)$-$IrO_2(20)$ electrode and the $Ta_2O_5(20)$-$IrO_2(80)$ electrode had similar effective surface areas.

3.2. Oxygen Evolution Reaction

Figure 2 represents the steady state current densities at each applied step potential in 2 M $HClO_4$ to compare the oxygen evolution reactions of anodes as a function of the chemical composition. Because the oxygen evolution reaction competes with the oxidation of chloride ions in the saline solution due to their similar ovepotentials, it is necessary to differentiate the former from the latter for practical use.

The $Ta_2O_5(20)$-$IrO_2(80)$ electrode was expected to show the highest steady state current density due to the inherently low overpotential characteristics of IrO_2 for the oxygen evolution process. However, for the mixed metal oxide anodes prepared by dipping, the efficiencies of oxygen evolution reaction were $Ta_2O_5(50)$-$IrO_2(50)$ ≈ $Ta_2O_5(20)$-$IrO_2(80)$ > $Ta_2O_5(80)$-$IrO_2(20)$ in **Figure 2(a)**. The fact that the $Ta_2O_5(50)$-$IrO_2(50)$ electrode had a comparable efficiency to the $Ta_2O_5(20)$-$IrO_2(80)$ electrode was assumed to be due to the large active surface area of the former as shown in **Figure 3**. On the other hand,

Table 1. Superficial anodic charge densities of chloride oxidation reactions with the variation of [Ta₂O₅]/[IrO₂] of Ta₂O₅-IrO₂ electrodes prepared by a di pp.ing method and brushing method.

Composition	Di pp.ing method	Brushing method
$Ta_2O_5(80)$-$IrO_2(20)$	7.07 mC/cm²	6.49 mC/cm²
$Ta_2O_5(50)$-$IrO_2(50)$	9.44 mC/cm²	8.51 mC/cm²
$Ta_2O_5(20)$-$IrO_2(80)$	6.87 mC/cm²	4.84 mC/cm²

the $Ta_2O_5(20)$-$IrO_2(80)$ electrode was more active than the $Ta_2O_5(80)$-$IrO_2(20)$ electrode owing to its higher
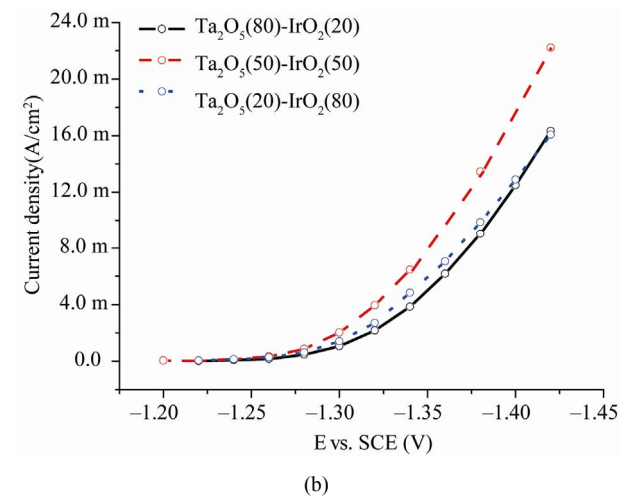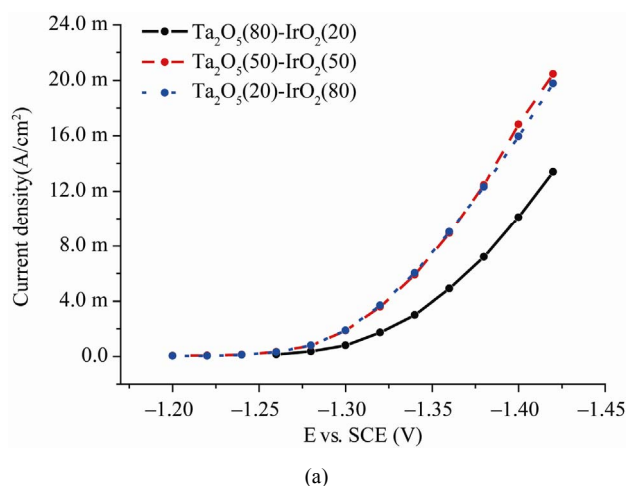

(a)

(b)

Figure 2. Steady state current densities of Ta₂O₅-IrO₂ electrodes in 1 M HClO₄ aqueous solution (20°C) from –1.20 V to –1.42 V with a potential step of +20 mV for 30 s. (a) Curves: (solid) Ta₂O₅(80)-IrO₂(20), (dash) Ta₂O₅(50)-IrO₂(50), (dot) Ta₂O₅(20)-IrO₂(80) prepared by a di pp.ing method; (b) Curves: (solid) Ta₂O₅(80)-IrO₂(20), (dash) Ta₂O₅(50)-IrO₂(50), (dot) Ta₂O₅(20)-IrO₂(80) prepared by a brushing method. Number of oxide layers was 4.

content of the IrO$_2$ component. Anodes formed by brushing also had a similar tendency to those formed by dipping in **Figure 2(b)**.

3.3. Microstructure Analysis

Figure 3 displays the surface morphologies of Ta$_2$O$_5$-IrO$_2$ mixed metal oxide layers prepared by **(a-c)** dipping and **(d-f)** brushing. Oxide layers formed by dipping showed smoother surfaces than those by brushing and the transition of surface microstructures with a molar ration of Ta: Ir was similar for both methods.

Oxide layers containing high Ta$_2$O$_5$ contents (**Figures 3 (a)** and **(d)**) formed a type of agglomerates of IrO$_2$ on an amorphous Ta$_2$O$_5$ background, while oxides with high IrO$_2$ contents showed a high crack density on the surface. Interestingly, the Ta$_2$O$_5$(50)-IrO$_2$(50) electrode had both agglomerates and many cracks on the surface, which may lead to a large active surface area and result in the highest steady state current densities in **Figure 1** and **Figure 2**.

The durability of an anode is influenced by its surface state, which also may be controlled by the molar ratio between constitutional chemical compositions. Therefore, the adjustment of the compositional ratio of the IrO$_2$-Ta$_2$O$_5$ system with a suitable preparation method should make it possible to enhance the corrosion resistance of an anode by inducing compact surface structures and so can be effectively used for the fabrication of the mixed metal oxide layer durable in severe environments.

3.4. Accelerated Life Tests

For the evaluation of the durability of individual mixed metal oxide anodes, accelerated life tests were executed in a mixed solution of 1 M NaCl and 2 M HClO$_4$ (50°C) at a constant current density of 2 A/cm^2. **Figure 4** compares potential transition curves among the mixed metal oxide anodes prepared from different molar ratios of [Ta$_2$O$_5$]/[IrO$_2$] and with different coating methods. For each anode, an abrupt potential increase appears before its breakdown, which means complete deactivation of the electrode surface. The mixed metal oxide anodes prepared by the brushing method showed relatively better corrosion resistance than those coated by the dipping method. Correlating the effect of constituent chemical compositions of anodes with a coating method, the coating method made little differences for the oxide layers of low IrO$_2$ contents, but the brushing method proved its superiority to the dipping method as the IrO$_2$ content became higher.

Figure 5 shows the effect of a coating repetition number-that is, oxide thickness - of the Ta$_2$O$_5$(20)-IrO$_2$- (80) layer on the electrode's serviceable life. The continuous process of coating, drying, and thermal decomposition was
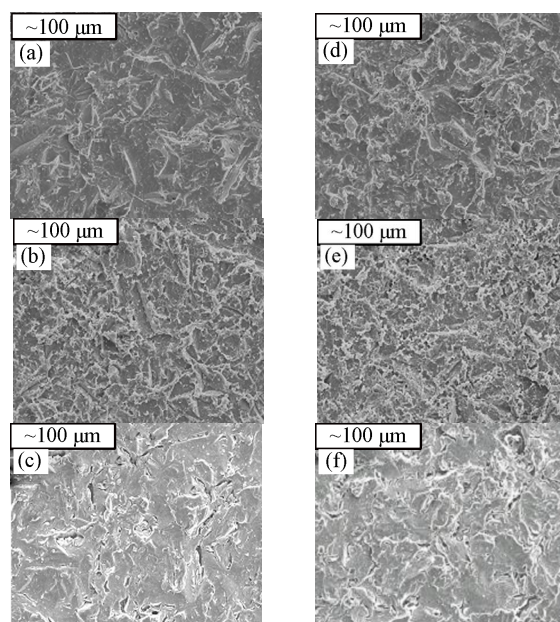

Figure 3. SEM photographs of Ta$_2$O$_5$-IrO$_2$ electrodes prepared by a di pp.ing method (a-c) and by a brushing method (d-f). (a,d) Ta$_2$O$_5$(80)-IrO$_2$(20); (b,e) Ta$_2$O$_5$(50)-IrO$_2$(50); (c,f) Ta$_2$O$_5$(20)-IrO$_2$(80), whose coating number was 4.

repeated three times, six times, and ten times respectively, which were confirmed as 0.3 μm, 0.7 μm, 1.2 μm thick, to achieve the comparative thickness of oxides. We could get the linear relationship between the oxide thickness and anode span in curve in a very aggressive solution (1 M NaCl and 2 M HClO$_4$, 50°C, at a constant current density, 2 A/cm^2) as below.

Service life (hr) = −12.4 + 28 X (coating number)   (1)

Since the oxide layer was formed in proportional to the coating number, around 0.1 ~ 0.15 μm/1 coat, this fact facilitates us to calculate the necessary oxide thickness for a certain service time.

Figure 6 shows typical potential–time curves representing the change of durability of anodes with different chemical compositions, whose layers were prepared by a brushing method with ten repetitions. Judging from the fact that the serviceable time of the electrodes has been extended with an increase of IrO$_2$ content in the IrO$_2$-Ta$_2$O$_5$ system, it is concluded that the IrO$_2$ content is one of the critical factors dominating the corrosion resistance of this type of mixed metal oxide anode. However, it is noticeable that a mixed metal oxide anode, Ta$_2$O$_5$(20)-IrO$_2$(80), showed a superior durability to the single component pure IrO$_2$ electrode.

4. Conclusions

1) The efficiency for the chloride oxidation and the oxygen evolution process of the Ta$_2$O$_5$-IrO$_2$ mixed metal

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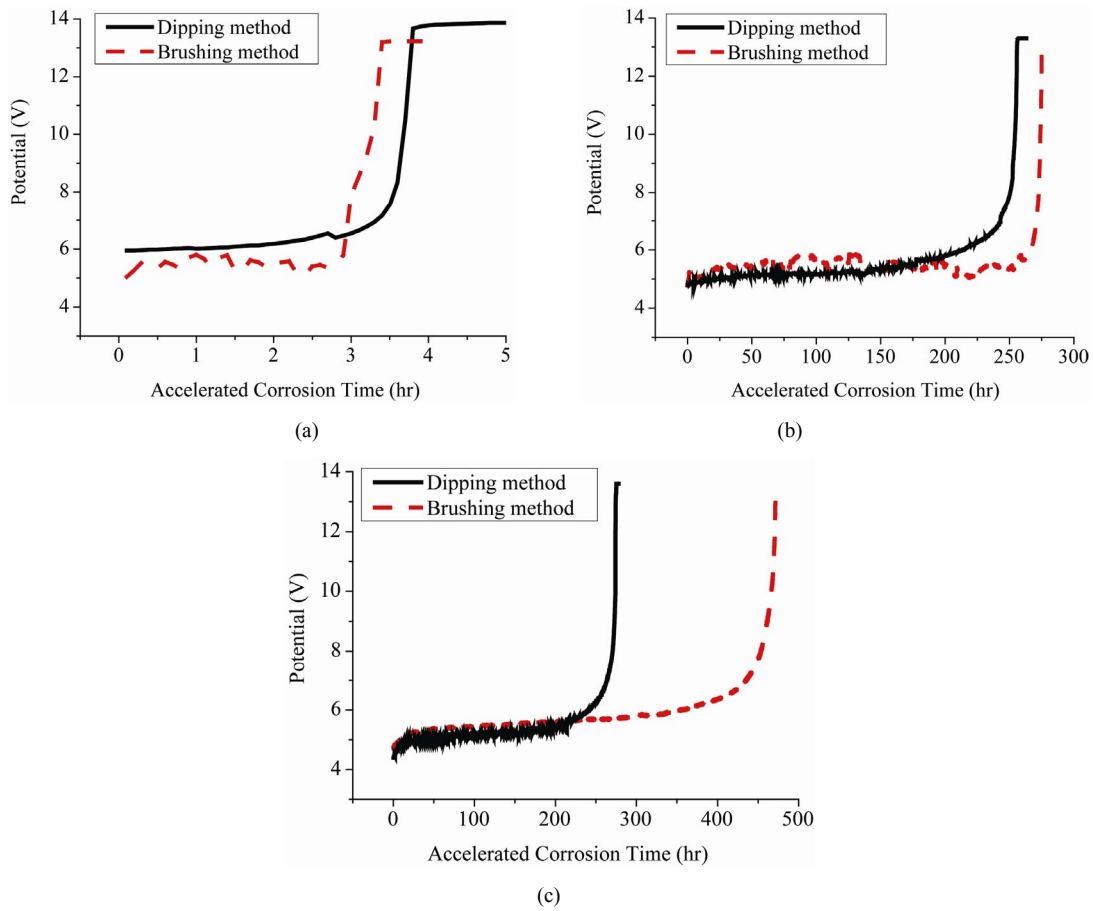Application of Electrolysis Process

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(a)

(b)

(c)

Figure 4. Typical potential-time curves of Ta₂O₅-IrO₂ electrodes prepared by (solid) di pp.ing method and (dot) brushing method with coating number of 10. (a) [Ta₂O₅] : [IrO₂] = 80:20; (b) [Ta₂O₅] : [IrO₂] = 50:50; (c) [Ta₂O₅] : [IrO₂] = 20:80. Accelerated life tests were executed at constant current densities of 2 A/cm² in a (1 M NaCl + 2 M HClO₄) mixed solution (50˚C).
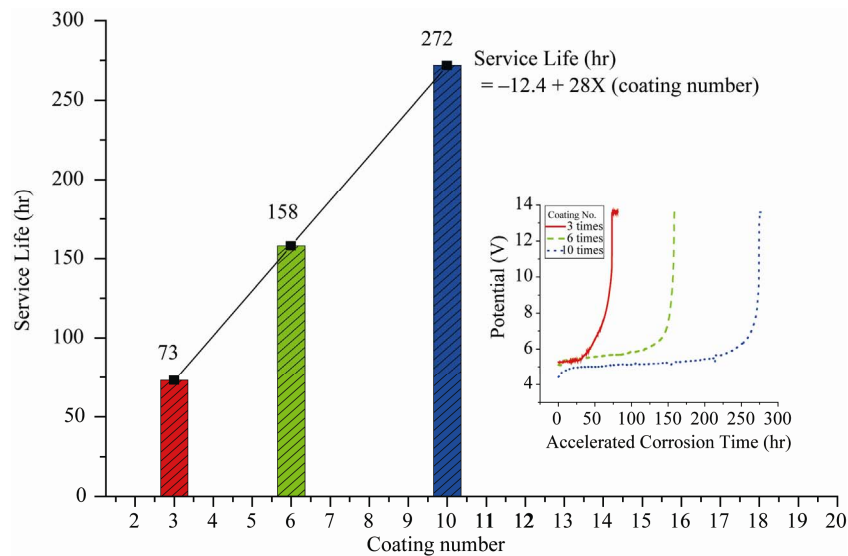

Figure 5. Typical potential-time curves of Ta₂O₅(20)-IrO₂(80) electrodes with different coating numbers, which were prepared by the brushing method and recorded under the same acceleration life test conditions of Figure 3.
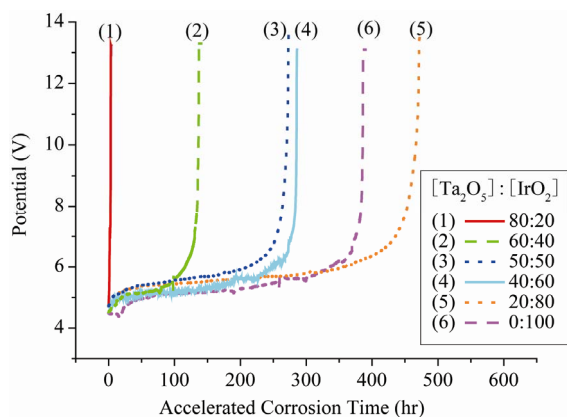
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Figure 6. Typical potential-time curves for Ta$_2$O$_5$-IrO$_2$ electrodes with a variation of [Ta$_2$O$_5$]/[IrO$_2$], which were prepared by the brushing method and recorded under the same acceleration life test conditions of Figure. 3. Curves: (solid) [Ta$_2$O$_5$] : [IrO$_2$] = 80:20; (dash) 60:40; (dot) 50:50; (bold solid) 40:60; (bold dot) 20:80; (bold dash) 0:100.

oxide anodes were not only influenced by the chemical composition but also by the surface morphology of oxide electrodes, which was susceptible to the molar ratios of the two components and the coating method. The Ta$_2$O$_5$(50)-IrO$_2$(50) electrode revealed the highest catalytic activity for the chloride ion oxidation and the oxygen evolution reaction due to the combinational effects of its chemical and surface characteristics.

2) Coating methods to prepare the mixed metal oxides did not make noticeable differences to the catalytic activity of the anodes, but the dipping method was relatively advantageous over the brushing method for the chloride ion oxidation reaction, while the brushing method showed better catalytic activity for the oxygen evolution process. When the effect of chemical composition of the Ta$_2$O$_5$-IrO$_2$ mixed metal oxides was considered separately, low contents of Ir$_2$O$_2$ in the anode facilitated the chloride ion oxidation reaction and higher IrO$_2$ promoted the oxygen evolution process.

3) The durability of the oxide electrode improved as the thickness of the oxide layer increased and the molar ratio of [IrO$_2$] to [Ta$_2$O$_5$] approached 80/20, which was superior to 100/0.

5. Acknowledgements

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