

# ENHANCED STABILITY AND ELECTROCHEMICAL PERFORMANCE OF A BaTiO<sub>3</sub>/PbO<sub>2</sub> ELECTRODE VIA A LAYER OBTAINED WITH LAYER ELECTRODEPOSITION

## IZBOLJŠANA STABILNOST IN ELEKTROKEMIJSKA ZMOGLJIVOST ELEKTRODE BaTiO<sub>3</sub>/PbO<sub>2</sub>, IZDELANE Z ELEKTRODEPOZICIJO PLAST NA PLAST

Govindan Muthuraman, Kannan Karunakaran, Il Shik Moon

Department of Chemical Engineering, Sunchon National University, #255 Jungangno, Suncheon 540-742, Jeollanam-do, Republic of Korea  
ismoon@sunchon.ac.kr

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Herein, the electrodeposition of BaTiO<sub>3</sub> and PbO<sub>2</sub> on Ti using the layer-by-layer method under different current densities (*CDs*) and times, was investigated. The weight difference in the deposited BaTiO<sub>3</sub> explains the BaTiO<sub>3</sub> weight decrease by one order with the increasing *CD* from 0.025 A cm<sup>-2</sup> to 0.125 A cm<sup>-2</sup> and also follows the same trend during the PbO<sub>2</sub> deposition. The PbO<sub>2</sub> deposition at different *CDs* demonstrates that the deposited PbO<sub>2</sub> weight increases by one order with the increasing *CD*. Also, cyclic voltammetry results explain the low and moderate deposition *CDs* and the time suitably shows the PbO<sub>2</sub> redox behavior. According to SEM and XRD, a *CD* of 0.05 A cm<sup>-2</sup> affects the formation of crystalline BaTiO<sub>3</sub> and PbO<sub>2</sub> more than higher or lower *CDs*. Finally, the BaTiO<sub>3</sub> and PbO<sub>2</sub> layer-by-layer electrode electrodeposited at a moderate *CD* showed a better stability than the electrode including only PbO<sub>2</sub>. The use of BaTiO<sub>3</sub> is promising for the stability of the PbO<sub>2</sub> electrode preparation.

Keywords: BaTiO<sub>3</sub>, PbO<sub>2</sub>, electrodeposition, layer by layer, electrode stability

Preiskovana je bila elektrodepozicija BaTiO<sub>3</sub> in PbO<sub>2</sub> na Ti, z uporabo metode plast na plast, pri različnih časih in gostotah toka (*CD*). Razlike v teži BaTiO<sub>3</sub> razložijo naraščanje teže BaTiO<sub>3</sub> za red velikosti zaradi naraščanja *CD*, od 0,025 A cm<sup>-2</sup> do 0,125 A cm<sup>-2</sup>. Podoben trend je bil opažen tudi pri nanašanju PbO<sub>2</sub>. Pri nanašanju PbO<sub>2</sub>, različni *CD* kažejo naraščanje teže nanosenega PbO<sub>2</sub> za red velikosti z naraščanjem *CD*. Tudi ciklična voltometrija razloži majhen in srednji *CD* in čas ustrezno kaže redoks vedenje PbO<sub>2</sub>. SEM in XRD z 0,05 A cm<sup>-2</sup> vodita nastanek kristaliničnega BaTiO<sub>3</sub> in PbO<sub>2</sub> bolj kot višji in nižji *CD*. Končno se kaže boljša stabilnost elektrode elektronanešenega BaTiO<sub>3</sub> in PbO<sub>2</sub> plast na plast pri zmernih *CD*, kot pa pri PbO<sub>2</sub> elektrodi. Uporaba BaTiO<sub>3</sub> je obetajoča za stabilnost priprave PbO<sub>2</sub> elektrode.

Ključne besede: BaTiO<sub>3</sub>, PbO<sub>2</sub>, elektro nanašanje, plast na plast, stabilnost elektrode

## 1 INTRODUCTION

PbO<sub>2</sub> clearly emerges as an attractive material used as an anode for a direct oxidation of organic compounds due to its high oxygen-evolution potential, low price, relative stability under the high positive potentials required, stability at high temperatures and ease of preparation.<sup>1-3</sup> Its high overpotential for O<sub>2</sub> evolution allows the application of potentials to about 2.0 V versus a saturated calomel electrode (SCE) in an acidic medium without vigorous O<sub>2</sub> evolution.<sup>4</sup> The PbO<sub>2</sub> electrodes have some disadvantages, i.e., they corrode at high rates under reducing conditions and in some acids, and they have poor mechanical properties. Its composites with various oxides (e.g., Al<sub>2</sub>O<sub>3</sub>, RuO<sub>2</sub>, and TiO<sub>2</sub>) are known<sup>4-6</sup> for a high catalytic activity and stability. Among many ways of preparation, such as the sol-gel technology, the plasma-chemical method, etc., the electrochemical synthesis is the most promising method, easy to implement, allowing the technological parameters to be varied smoothly for a better control of the

composition and properties of the resulting composites.<sup>4-6</sup>

The  $\alpha$ - and  $\beta$ -types of PbO<sub>2</sub>, applied layer by layer on metal anodes have been widely used in electrolysis.<sup>7,8</sup> Generally, titanium is not a viable substrate for practical electrodes in electrodepositing non-ferrous metals. Aluminum is relatively cheap and has a good conductivity. The electrode material obtained by electrodepositing lead dioxide on an Al substrate has huge market prospects. A stress-free intermediate  $\alpha$ -PbO<sub>2</sub> coating is produced with electrodeposition from an alkaline lead bath<sup>9</sup> and it plays the role of the binder on the top  $\alpha$ -PbO<sub>2</sub> coating, improving the service life of the electrode. A non-conducting ceramic material has also been used as the substrate to achieve a high stability of PbO<sub>2</sub> with the fluorine resin as the co-dopant on the upper layer.<sup>10</sup>

In the present investigation, perovskite-type BaTiO<sub>3</sub> is applied to Ti as the lower layer using the hydrothermal electrodeposition method. As the top/upper layer, PbO<sub>2</sub> is to be applied. The effects of the thickness of both layers are controlled with the current density and the deposition

time to achieve the electrode stability and activity. Thus, the main work of this paper deals with the layer-by-layer deposition of BaTiO<sub>3</sub> and PbO<sub>2</sub> and its influence on the PbO<sub>2</sub> electrode stability and electrochemical application as a sandwich-type electrode.

## 2 EXPERIMENTAL DETAILS

### 2.1 Electrodeposition

Electrolysis was performed using a DC power supply, BS 32C (0–100 V, 0–50 A) from the Korea Switching Company, Korea, using the constant-current mode (the galvanostatic mode). Before the electrolysis start, the anode was initially immersed in the electrolyte for 1 min to stabilize its surface state. The deposition was performed in two steps: in the first step, BaTiO<sub>3</sub> was formed as the lower coating on a pretreated Ti electrode at 65 °C using a current-density range of 0.025–0.125 A cm<sup>-2</sup> for 15–60 min. The BaTiO<sub>3</sub> deposited electrode was washed after its deposition in hot ammonia water adjusted to pH 11 to minimize the BaCO<sub>3</sub> formation. It was then rinsed in purified Millipore water and cleaned in ethanol with an ultrasonic cleaner for 1 min. Then, β-PbO<sub>2</sub> was deposited as the upper layer using a current-density (*CD*) range of 0.025–0.125 A cm<sup>-2</sup> for 15–60 min in a 0.1 M HNO<sub>3</sub> medium at 65 °C.

### 2.2 Analysis

Cyclic-voltammetry (CV) measurements were performed using a VersaSTAT3 from Princeton Applied Research, USA. The electrochemical cell was a three-electrode cell with a working electrode, a platinum-plate counter electrode and an Ag/AgCl reference electrode. The working electrode was prepared with the electrodeposition method. Scanning electron microscopy of the prepared PbO<sub>2</sub> electrodes was carried out with Zeiss EVO MA10 to investigate the surface morphology of the films. The XRD patterns of the as-prepared PbO<sub>2</sub> samples were obtained from an X'PERT-PRO X-ray diffractometer with Cu-Kα radiation ( $\lambda = 0.1540598$  nm). The electrolysis was done using a DC power supply from KSC, Korea, with an applied *CD* of 0.3 A cm<sup>-2</sup> in 1 M H<sub>2</sub>SO<sub>4</sub>.

## 3 RESULTS AND DISCUSSIONS

### 3.1 Selection of deposition conditions

As the *CD* and time are the key factors to control the deposition, the initial work was done to identify the suitable deposition time and *CD* for the first and second layers of BaTiO<sub>3</sub> and PbO<sub>2</sub>, respectively. First, the BaTiO<sub>3</sub> layer was deposited using four different *CD*s of (0.025, 0.05, 0.1, 0.125) A cm<sup>-2</sup> with four different durations such as (15, 30, 45, 60) min, by keeping the PbO<sub>2</sub> (the second layer) deposition parameters (*CD* = 0.05 A cm<sup>-2</sup>, 30 min duration) constant. As mentioned in

the experimental section, the BaTiO<sub>3</sub> and PbO<sub>2</sub> depositions were done in different solutions and the obtained results are tabulated in **Table 1**. It is seen from the 1<sup>st</sup> row and 4<sup>th</sup> column of **Table 1** that the weight of the deposited BaTiO<sub>3</sub> shows no consistency with different deposition times (15–60 min) within a single *CD* (0.025 A cm<sup>-2</sup>); a similar inconsistency is also shown for the PbO<sub>2</sub> deposition, the 5<sup>th</sup> column. However, with the increasing *CD* during the BaTiO<sub>3</sub> deposition from 0.025 to 0.125 A cm<sup>-2</sup>, shown in the 1<sup>st</sup> to the 4<sup>th</sup> rows, the deposited BaTiO<sub>3</sub> weight is reduced by one order from 0.01 to 0.001 (the 5<sup>th</sup> column).

**Table 1:** Deposited-electrode weight difference in each step of: 1) BaTiO<sub>3</sub> and 2) PbO<sub>2</sub> at different current densities and times

**Tabela 1:** Razlika v masi nanešene elektrode za vsako stopnjo: 1) BaTiO<sub>3</sub> in 2) PbO<sub>2</sub> pri različnih gostotah tokov in časih

BaTiO <sub>3</sub> coating		PbO <sub>2</sub> coating		Electrode weight difference (g)	
<i>CD</i> <sup>@</sup> (A cm <sup>-2</sup> )	Time (min)	<i>CD</i> <sup>@</sup> (A cm <sup>-2</sup> )	Time (min)	BaTiO <sub>3</sub>	PbO <sub>2</sub>
0.025	15	0.05	30	0.0222	0.2837
	30		0.0181	0.2929	
	45		-0.0034	0.3043	
	60		0.024	0.3041	
0.05	15	0.05	30	-0.0027	0.0649
	30		0.0052	0.2681	
	45		0.0055	0.2969	
	60		-0.0087	0.2997	
0.1	15	0.05	30	0.0094	0.0908
	30		0.0054	0.0217	
	45		-0.0089	0.0338	
	60		-0.0076	0.0800	
0.125	15	0.05	30	0.0032	0.0702
	30		-0.0372	*	
	45		0.0629	*	
	60		0.0397	0.0456	
0.025	30	0.025	15	0.0159	-0.0318
			30	0.0091	-0.0133
			45	0.0084	0.1065
			60	0.0167	0.4092
0.025	30	0.05	15	-0.0594	0.2779
			30	0.0105	0.2188
			45	-0.0180	0.0140
			60	-0.0065	0.5551
0.025	30	0.1	15	0.1020	0.1611
			30	-0.0423	0.3547
			45	-0.0137	0.4100
			60	-0.0362	0.2384
0.025	30	0.125	15	*	*
			30	*	*
			45	*	*
			60	*	*

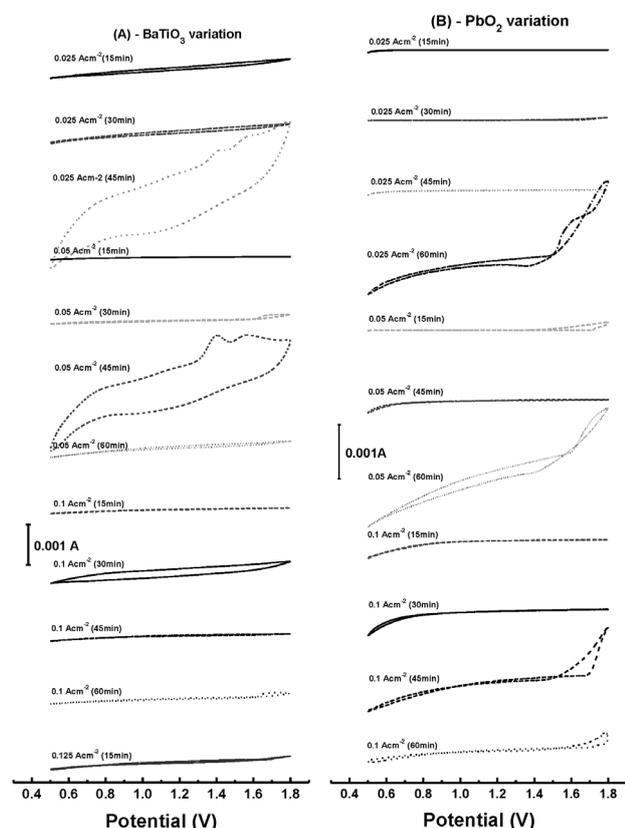
\*Dissolution of electrode, <sup>@</sup>*CD* = current density, lower than the original weight

In a similar way, the deposited PbO<sub>2</sub> weight also decreases by one order with the increasing *CD* (the 6<sup>th</sup> column – from 0.1 to 0.01). At the same time, the results are different if the PbO<sub>2</sub> deposition *CD* is varied at a

fixed  $CD$  and time of the BaTiO<sub>3</sub> deposition (5<sup>th</sup> to 8<sup>th</sup> rows of **Table 1**) where the weight of BaTiO<sub>3</sub> is maintained constant but the deposited PbO<sub>2</sub> weight is increased by one order with the increasing  $CD$  (the 6<sup>th</sup> column of the 5<sup>th</sup> to 8<sup>th</sup> rows in **Table 1**). It is maintained on the basis of the results that the formation of BaTiO<sub>3</sub> on the Ti electrode influenced further deposition of PbO<sub>2</sub>, which means that the conductivity was lower when BaTiO<sub>3</sub> completely covered the electrode due to the dielectric properties of BaTiO<sub>3</sub>. A similar trend applied to the PbO<sub>2</sub> deposition carried out at a high  $CD$  and a fixed, low  $CD$  of the BaTiO<sub>3</sub> deposition where a complete dissolution of the deposited film was observed.

Further, through a CV analysis, the electron-transfer behavior of the deposited electrode can be inferred on the basis of PbO<sub>2</sub> redox properties. **Figure 1a** shows the PbO<sub>2</sub> redox response to the effects of various  $CD$ s and

times of the BaTiO<sub>3</sub> deposition, where no redox peaks for PbO<sub>2</sub> are observed except for two  $CD$  variations in the BaTiO<sub>3</sub> deposition: 0.025 (45 min) A cm<sup>-2</sup> and 0.05 (45 min) A cm<sup>-2</sup>. In all the remaining conditions, only a charge transfer like the CV response is observed. In the case of the variation in the PbO<sub>2</sub> deposition, 60 min and 0.025 A cm<sup>-2</sup> or 0.05 A cm<sup>-2</sup>,  $CD$  only shows redox peaks that resemble PbO<sub>2</sub>,<sup>11</sup> as shown in **Figure 1b**. Under another two conditions, a  $CD$  of 0.1 A cm<sup>-2</sup> over 45 min and 60 min deposition times, the PbO<sub>2</sub> deposition indicates a low oxidation current. All the remaining conditions show a charging current like the CV response without any redox peaks as not enough PbO<sub>2</sub> is exposed on the electrode surface. This is well correlated with the deposited weight of PbO<sub>2</sub> in the 1<sup>st</sup>, 2<sup>nd</sup> and 7<sup>th</sup> rows of the 6<sup>th</sup> column of **Table 1**, where only the deposited PbO<sub>2</sub> weight is higher than in the other conditions.

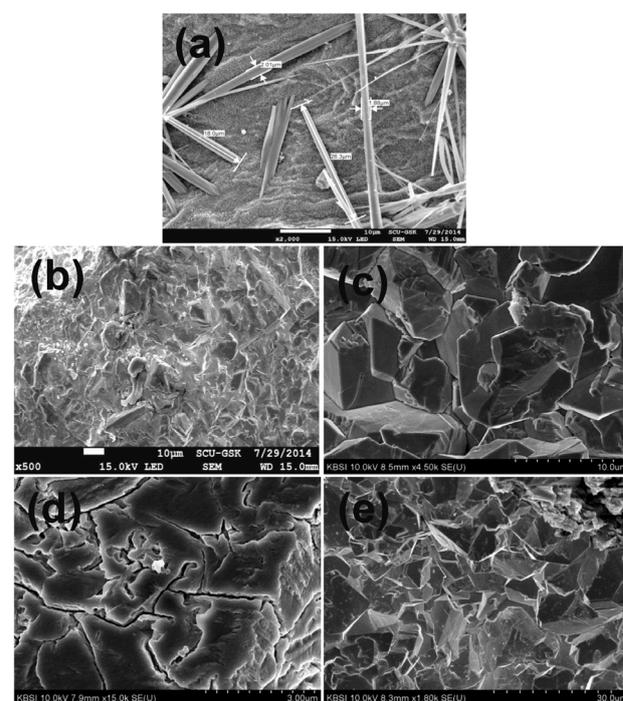


**Figure 1:** CV results for electrodeposited BaTiO<sub>3</sub>/PbO<sub>2</sub> using different current densities and times in a 0.1 M phosphate buffer solution at a scan rate of 20 mV s<sup>-1</sup>: a) variation in BaTiO<sub>3</sub> deposition current density and time with fixed current density and time (0.05 A cm<sup>-2</sup>, 30 min) of PbO<sub>2</sub> deposition, b) variation in PbO<sub>2</sub> deposition current density and time with fixed current density and time (0.025 A cm<sup>-2</sup>, 30 min) of BaTiO<sub>3</sub> deposition

**Slika 1:** Rezultati ciklične voltametrije elektro nanešenega BaTiO<sub>3</sub>/PbO<sub>2</sub>, pri uporabi različnih gostot tokov in časov, v 0,1 M fosfatni puferski raztopini pri hitrosti skeniranja 20 mV s<sup>-1</sup>: a) spreminjanje gostote toka in časa nanašanja BaTiO<sub>3</sub> od stalne gostote toka in časa (0,05 A cm<sup>-2</sup>, 30 min) pri nanašanju PbO<sub>2</sub>, b) spreminjanje gostote toka in časa nanašanja PbO<sub>2</sub> pri stalni gostoti toka in časa (0,025 A cm<sup>-2</sup>, 30 min) nanašanja BaTiO<sub>3</sub>

### 3.2 Morphological characterization

SEM images of the BaTiO<sub>3</sub> and PbO<sub>2</sub> deposited at different conditions are depicted in **Figure 2**. The first layer of BaTiO<sub>3</sub> shows no distinctive difference in the SEM image and it looks almost like a needle structure in micrometer size, as shown in **Figure 2a**. Both layers deposited at 0.025 A cm<sup>-2</sup> show a densely deposited PbO<sub>2</sub> layer (**Figure 2b**). There is a defect in the PbO<sub>2</sub>

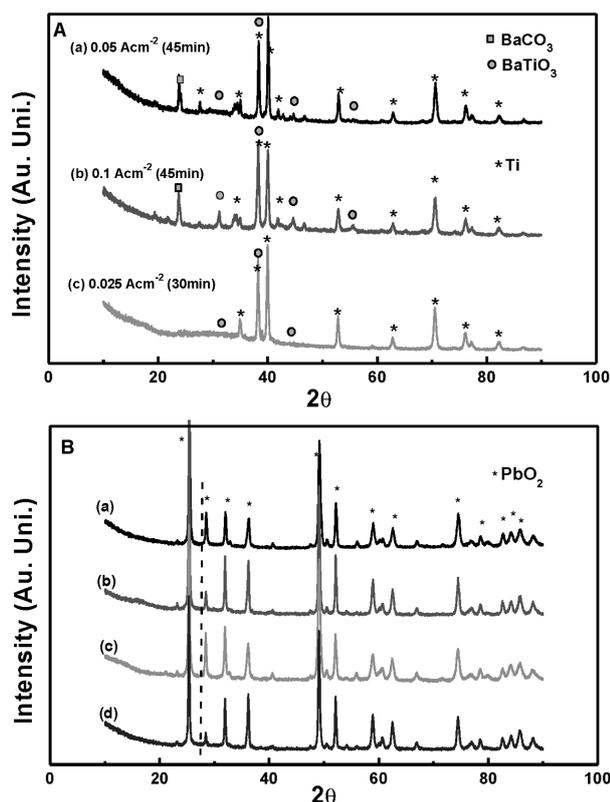


**Figure 2:** SEM images of BaTiO<sub>3</sub> and PbO<sub>2</sub> electrodes, deposited at different  $CD$ s and times: a) BaTiO<sub>3</sub><sup>0.1(45 min)</sup>, b) BaTiO<sub>3</sub><sup>0.025(30 min)/PbO<sub>2</sub><sup>0.025(60 min)</sup>, c) BaTiO<sub>3</sub><sup>0.025(30 min)/PbO<sub>2</sub><sup>0.1(60 min)</sup>, d) BaTiO<sub>3</sub><sup>0.05(45 min)/PbO<sub>2</sub><sup>0.05(30 min)</sup>, e) BaTiO<sub>3</sub><sup>0.1(45 min)/PbO<sub>2</sub><sup>0.05(30 min)</sup></sup></sup></sup></sup>

**Slika 2:** SEM-posnetki BaTiO<sub>3</sub> in PbO<sub>2</sub> elektrod, nanešenih pri različnih  $CD$  in časih: a) BaTiO<sub>3</sub><sup>0.1(45 min)</sup>, b) BaTiO<sub>3</sub><sup>0.025(30 min)/PbO<sub>2</sub><sup>0.025(60 min)</sup>, c) BaTiO<sub>3</sub><sup>0.025(30 min)/PbO<sub>2</sub><sup>0.1(60 min)</sup>, d) BaTiO<sub>3</sub><sup>0.05(45 min)/PbO<sub>2</sub><sup>0.05(30 min)</sup>, e) BaTiO<sub>3</sub><sup>0.1(45 min)/PbO<sub>2</sub><sup>0.05(30 min)</sup></sup></sup></sup></sup>

coating if the PbO<sub>2</sub> deposition *CD* increased to 0.1 A cm<sup>-2</sup> (**Figure 2c**). The film cracking is more dominant if both layers were deposited at a *CD* of 0.05 A cm<sup>-2</sup> (**Figure 2d**). The film cracking is more enhanced if the BaTiO<sub>3</sub> layer was deposited at 0.1 A cm<sup>-2</sup> (**Figure 2e**). When both layers were deposited at a low *CD* of 0.025 A cm<sup>-2</sup>, there is a smooth layer with smaller particles.

**Figure 3a** shows the XRD patterns of the BaTiO<sub>3</sub> deposited electrodes using different *CD*s and times with a fixed PbO<sub>2</sub> deposition. With the applied *CD* of 0.05 A cm<sup>-2</sup>, the BaTiO<sub>3</sub> peak reflections are less intense at  $2\theta$  values of about 31.03, 38.58, 41.88, and 55.47 (VRC# 01-075-0213) along with the Ti reflections. Additionally, BaCO<sub>3</sub> also appeared on the surface, with a  $2\theta$  peak at about 23.85 (VRC# 00-044-1487) that might have occurred after the deposition of BaTiO<sub>3</sub> due to high pH.<sup>12</sup> The BaTiO<sub>3</sub> peak intensity increased when the deposition *CD* increased to 0.1 A cm<sup>-2</sup> as shown on curve b in **Figure 3a**. The lowest *CD* (0.025 A cm<sup>-2</sup>) caused a decrease in the crystallinity of BaTiO<sub>3</sub>, which turned to the amorphous phase; see a broad peak between  $2\theta$  of



**Figure 3:** a) XRD patterns of current-density and time variation (mentioned in the figure) of BaTiO<sub>3</sub> deposition, b) XRD patterns of BaTiO<sub>3</sub>/PbO<sub>2</sub> prepared at various current densities and times: a) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.025(60 min)</sup>, b) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.1(60 min)</sup>, c) BaTiO<sub>3</sub><sup>0.05(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>, d) BaTiO<sub>3</sub><sup>0.1(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>

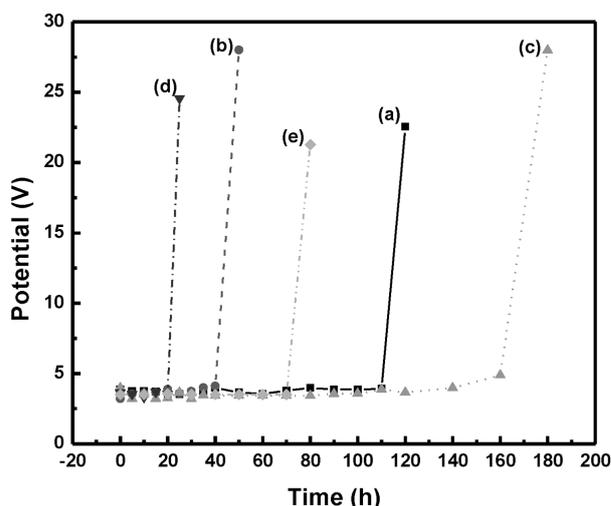
**Slika 3:** a) Rentgenogram spreminjanja gostote toka in časa pri nanašanju BaTiO<sub>3</sub>, b) rentgenogram BaTiO<sub>3</sub>/PbO<sub>2</sub> pripravljene pri različnih gostotah toka in časih: a) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.025(60 min)</sup>, b) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.1(60 min)</sup>, c) BaTiO<sub>3</sub><sup>0.05(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>, d) BaTiO<sub>3</sub><sup>0.1(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>

20–35 on curve c of **Figure 3**. In the case of the PbO<sub>2</sub> deposition, the deposited electrode using various *CD*s and times shows peaks for the PbO<sub>2</sub> formation (VRC# 01-076-0564) as shown in **Figure 3b**. The only difference found is a peak-intensity decrease at a  $2\theta$  value of about 28.49 when the PbO<sub>2</sub> is deposited at 0.1 A cm<sup>-2</sup> and 0.05 A cm<sup>-2</sup> (60 min and 30 min) on the top of the BaTiO<sub>3</sub> deposition using the 0.025 A cm<sup>-2</sup> and 0.1 A cm<sup>-2</sup> *CD*s in the 30 min and 45 min durations (**Figure 3b**, curves b and d).

It is evident from the results that the  $2\theta$  of 28.49 belongs to the  $\alpha(111)$  plane<sup>13</sup>, whose peak intensity is reduced, which means that the  $\beta$ -PbO<sub>2</sub> formation is predominant at this given condition. As seen on curve b in **Figure 3a**, the BaTiO<sub>3</sub> formation is more prominent at the 0.05 A cm<sup>-2</sup> *CD*, which means that the BaTiO<sub>3</sub> concentration increases the  $\beta$ -PbO<sub>2</sub> formation during the PbO<sub>2</sub> deposition. It is well known that the  $\beta$ -PbO<sub>2</sub> formation enhances the catalytic activity tremendously.<sup>14</sup>

### 3.3 Stability analysis

In order to apply the prepared electrodes, the selectively prepared electrodes obtained their stability in 1 M H<sub>2</sub>SO<sub>4</sub> due to an enhanced *CD* of 0.3 A cm<sup>-2</sup>, as depicted in **Figure 4**. An electrode that was prepared at a *CD* of 0.05 A cm<sup>-2</sup> (45 min) for BaTiO<sub>3</sub> and at 0.05 A cm<sup>-2</sup> (30 min) for the PbO<sub>2</sub> deposition showed a 3.9 V cell voltage up to 110 h (**Figure 4**, curve a); and after the potential sharply increased to 22 V the prepared electrode was decomposed. In the case of the increase in the BaTiO<sub>3</sub> deposition at the *CD* of 0.1 A cm<sup>-2</sup> when the



**Figure 4:** Electrolysis of different electrodes in 1 M H<sub>2</sub>SO<sub>4</sub> at accelerated current density of 0.3 A cm<sup>-2</sup>: a) BaTiO<sub>3</sub><sup>0.05(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>, b) BaTiO<sub>3</sub><sup>0.1(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>, c) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.025(60 min)</sup>, d) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.1(60 min)</sup>, e) PbO<sub>2</sub><sup>0.025(60 min)</sup>

**Slika 4:** Elektroliza različnih elektrod v 1 M H<sub>2</sub>SO<sub>4</sub> pri pospešeni gostoti toka 0,3 A cm<sup>-2</sup>: a) BaTiO<sub>3</sub><sup>0.05(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>, b) BaTiO<sub>3</sub><sup>0.1(45 min)</sup>/PbO<sub>2</sub><sup>0.05(30 min)</sup>, c) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.025(60 min)</sup>, d) BaTiO<sub>3</sub><sup>0.025(30 min)</sup>/PbO<sub>2</sub><sup>0.1(60 min)</sup>, e) PbO<sub>2</sub><sup>0.025(60 min)</sup>

other conditions stayed the same, the decomposition of the electrode occurred at around 40 h, which means that the stability was reduced with the increasing *CD* (Figure 4, curve b).

In the case of the lowest *CD* (0.025 A cm<sup>-2</sup>) used for both the BaTiO<sub>3</sub> and PbO<sub>2</sub> depositions for 30 min and 60 min, respectively, the stability increased to 180 h (Figure 4, curve c). By keeping the BaTiO<sub>3</sub> *CD* of 0.025 A cm<sup>-2</sup> and changing the PbO<sub>2</sub> *CD* to 0.1 A cm<sup>-2</sup> over 60 min, the stability of the prepared electrode tremendously decreased to 20 h, as observed in Figure 4, curve d. Finally, only the PbO<sub>2</sub> electrode deposited at the *CD* of 0.025 A cm<sup>-2</sup> in 60 min, showing a stability of 48 h (Figure 4, curve e) explains a high influence of the BaTiO<sub>3</sub> layer on the stability of the PbO<sub>2</sub> electrode.

#### 4 CONCLUSIONS

We successfully investigated an electrodeposition of BaTiO<sub>3</sub> and PbO<sub>2</sub> on Ti using the layer-by-layer method under different conditions. The weight measurement confirms that the BaTiO<sub>3</sub> and PbO<sub>2</sub> formation is optimum at a moderate *CD* of 50 A cm<sup>-2</sup> and a deposition time of 30–45 min. In addition, CV results confirm the same finding through the redox behavior of PbO<sub>2</sub>. SEM and XRD results further prove that a moderate *CD* leads to crystalline BaTiO<sub>3</sub> and β-PbO<sub>2</sub> rather than α-PbO<sub>2</sub>. The layer-by-layer deposition of BaTiO<sub>3</sub> and PbO<sub>2</sub> makes PbO<sub>2</sub> more stable than it would be if there was only PbO<sub>2</sub>. A further application of the selectively prepared electrode is in progress.

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