Supplementary Information for

Simultaneous Multi-Surface Anodizations and Stair-like Reverse Biases Detachment of Anodic Aluminum Oxides in Sulfuric and Oxalic Acid Electrolyte

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**(1) Surface morphologies of aluminum specimen during pretreatment procedure**

Highly purified (99.99%) aluminum (Al) plates were cut into rectangular parallelepipeds with width, length, and thickness of 20.0 mm, 50.0 mm, and 1.0 mm, respectively, through Guillotine cut with sheared edges. As shown in scanning electron microscope (SEM) image (Fig. S1a), left and bottom surfaces in a pristine Al substrate (*i.e.*, before electropolishing) have much rougher morphologies than front surfaceS[1](#_ENREF_1" \o "Hong, 2015 #107). After electropolishing procedure, the roughness of each surface clearly reduced as shown in Fig. S1b. However, morphologies of side and bottom surfaces in an electropolished Al substrate were still poorer than front surfaceS[1](#_ENREF_1" \o "Hong, 2015 #107). Fig. S1c shows a corner-view SEM image of the Al substrate covered with the anodic aluminum oxides (AAOs), which were fabricated through main simultaneous multi-surfaces anodization (SMSA). The qualities of the AAOs, such as initial nanopore’s diameter (*D*P0) and shapes, directly reflected the morphologies of electropolished Al surfaces. In our previous report, correlation between AAO fabricating conditions including surface morphologies and their characteristics were systematically investigatedS[2](#_ENREF_2" \o "Jeong, 2017 #169).

**(2) Detached AAOs from both sides and bottom surfaces through SRBs**

Figs. S4a and S4b show that each AAO was well detached from corresponding surface with equal dimensions in sulfuricS[1](#_ENREF_1" \o "Hong, 2015 #107) and oxalicS[2](#_ENREF_2" \o "Jeong, 2017 #169) acid electrolytes, respectively. As increasing the applied sequences, AAOs from two sides and bottom surfaces became narrower, which reflected the reduction of the total anodizing area (Fig. S4b).

**(3) Characteristics of AAOs from sides and bottom surfaces**

Fig. S5 exhibits the SEM images of the AAOs detached from left (a, c, and e) and right (b, d, and f) surfaces of the same Al substrate at every odd sequences shown in Fig. S4b. Nanopores were more randomly distributed with more complicated shapes and sizes, comparing with those of Front- and Back-AAOs (Fig. 3). These results were attributed that the two sides and bottom surfaces of the Al substrate had much poorer morphologies than front and back surface, even after electropolishing procedure as shown in Fig. S1.

Numerical values of the *D*P0 and interpore distance (*D*int) were extracted from the SEM images using an image processing software (ImageJ, National Institute of Health)S[2](#_ENREF_2). Qualitative behaviors of *D*P0 and *D*int shown in the SEM images (Figs. 3 and S5) were quantitatively agreed as compared in Fig. S6 and Table S1.

If morphologies of sides and bottom surfaces of a pristine Al substrate would be enhancing as good as those of front and back surfaces, the AAOs with comparable *D*P0 and *D*int can be fabricated independent of surface locations based on the fact that electric fields in the SMSA were applied to the normal direction of the Al substrateS[1-3](#_ENREF_1" \o "Hong, 2015 #107).

**References**

S1. Hong, Y. K., Kim, B. H., Kim, D. I., Park, D. H. Joo, J. High-yield and environment-minded fabrication of nanoporous anodic aluminum oxide templates. *RSC Adv.* **5** (34), 26872-26877, doi:10.1039/c5ra00198f (2015).

S2. Jeong, S. H. *et al.* Massive, eco-friendly, and facile fabrication of multi-functional anodic aluminum oxides: application to nanoporous templates and sensing platforms. *RSC Adv.* **7** (8), 4518-4530, doi:10.1039/C6RA25201J (2017).

S3. Lee, W. Park, S.-J. Porous Anodic Aluminum Oxide: Anodization and Templated Synthesis of Functional Nanostructures. *Chem. Rev.* **114** (15), 7487-7556 (2014).

**Table S1** Characteristic dimensions of the as-detached AAOs shown in Figs. 3 and S5 with respect to the locations of corresponding surfaces.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Applied Sequence**  **(*n*)** | ***D*P0 (nm)** | | ***D*int (nm)** | |
| **Front** | **Back** | **Front** | **Back** |
| 3 | 44.2 ± 2.71 | 44.6 ± 3.61 | 97.7 ± 1.99 | 97.8 ± 2.85 |
| 4 | 44.9 ± 1.72 | 44.9 ± 2.37 | 98.1 ± 2.64 | 97.8 ± 2.36 |
| 5 | 45.2 ± 1.89 | 44.7 ± 1.88 | 97.8 ± 2.53 | 97.7 ± 1.70 |
| **Applied Sequence**  **(*n*)** | ***D*P0 (nm)** | | ***D*int (nm)** | |
| **Left** | **Right** | **Left** | **Right** |
| 1 | 44.2 ± 4.32 | 43.1 ± 1.55 | 97.6 ± 2.15 | 97.8 ± 0.92 |
| 3 | 34.9 ± 3.97 | 37.7 ± 6.14 | 98.6 ± 1.63 | 98.3 ± 1.19 |
| 5 | 42.4 ± 1.03 | 34.7 ± 2.30 | 96.9 ± 1.22 | 97.8 ± 1.03 |



**Fig. S1** Corner-view of SEM images of (a) pristine and (b) electropolished Al substrate. (c) A corner-view of SEM image taken right after 1st Main-SMSA. Plural AAOs were formed on corresponding surfaces of the Al substrate. These figures have been modified from [S1].



**Fig. S2** Schematic arrangement of the Pt wire and Al substrate in top-view.



**Fig. S3** Photographs of the (a) pristine and (b) electropolished Al substrate.



**Fig. S4** (a) Photograph of the remaining Al substrate and 5 AAOs detached from the corresponding surfaces through 1st sequence in sulfuric acid electrolyte under fixed temperature of 0 °C. This figure has been reprinted from [S1]. (b) Photograph of the remaining Al substrate and all the AAOs produced from the every immersed surfaces from 1st to 5th sequences in oxalic acid electrolyte under fixed temperature of 15 °C. Each abbreviation indicated the corresponding surface (L: Left, R: Right, F: Front, Ba: Back, and Bo: Bottom). This figure has been reprinted from [S2].



**Fig. S5** SEM images of the open-pore side of the AAO detached from the both sides surfaces of the Al substrate; (a) 1st-Left AAO, (b) 1st-Right AAO, (c) 3rd-Left AAO, (d) 3rd-Right AAO, (e) 5th-Left AAO, and (f) 5th-Right AAO. The insets show the SEM images of the barrier sides of the corresponding AAOs. All the scale bars are 400 nm. This figure has been reprinted from [S2].



**Fig. S6** Comparison of the *D*P0 and *D*int in the as-detached AAOs from various surfaces in oxalic acid electrolyte at fixed electrolyte temperature of 15 oC.