## **Supporting Information**

## Highly Efficient Electroplating of (220)-oriented Nano-twinned Copper

## in the Methanesulfonic Copper Baths

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## **Supplementary Figures**

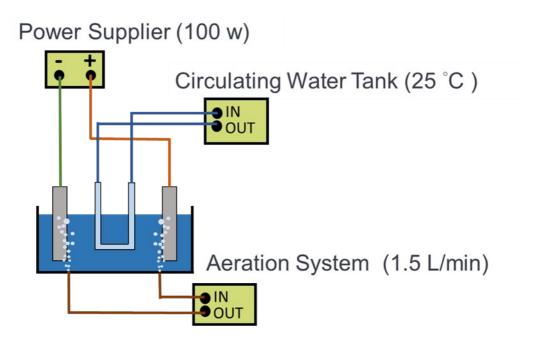


Figure S1. Schematic diagram of methanesulfonic copper baths for electroplating of (220)-oriented nano-twinned copper foil

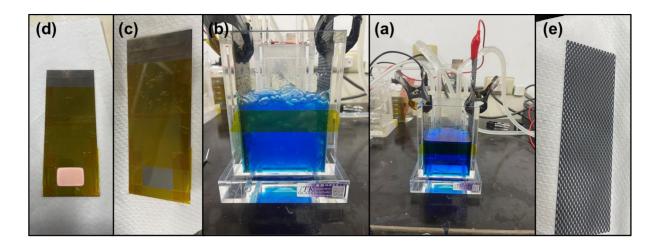


Figure S2. (a) Methanesulfonic copper baths before electroplating (b) Methanesulfonic copper baths during electroplating (c) The titanium plate with well-polished (d) The titanium plate after electroplating (e) The dimensionally stable anode (DSA<sup>®</sup>)

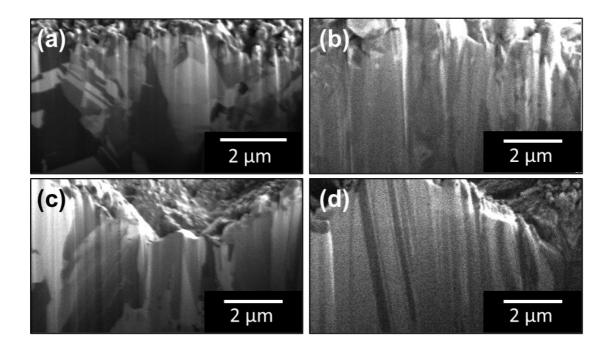


Figure S3. The ion-channeling images of the cross-section microstructure after the FIB milling for the Cu foils plated from methanesulfonic copper baths consisted of 0.86 M copper methanesulfonate (Cu(CH<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>) and 1.04 M methane-sulfonic acid (CH<sub>3</sub>SO<sub>3</sub>H) (a) without Cl<sup>-</sup> (b) 5 ppm, (c) 40 ppm, and (d) 120 ppm Cl<sup>-</sup>

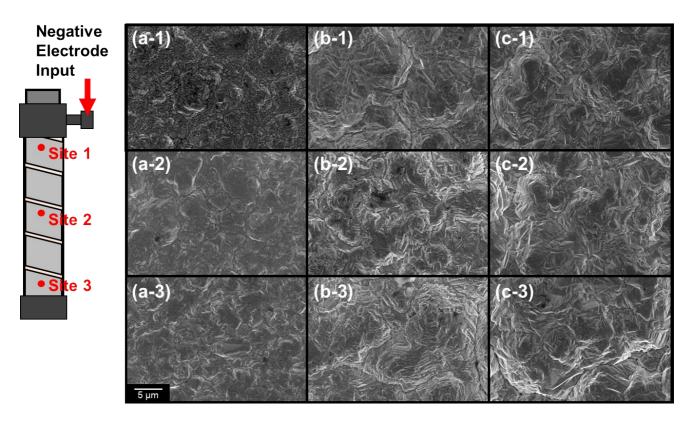


Figure S4. The SEM images of the Cu foils plated from methanesulfonic copper baths consisted of 0.86 M copper methanesulfonate (Cu(CH<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>) and 1.04 M methane-sulfonic acid (CH<sub>3</sub>SO<sub>3</sub>H) (a) without Cl<sup>-</sup> (b) 5 ppm, (c) 40 ppm in in-situ residual stress testing