

A Novel and Simple Aluminum /Sol-gel-derived Amorphous Aluminium Oxide Multilayer Film with High Energy Density

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1. The preparation of AmAO thin film

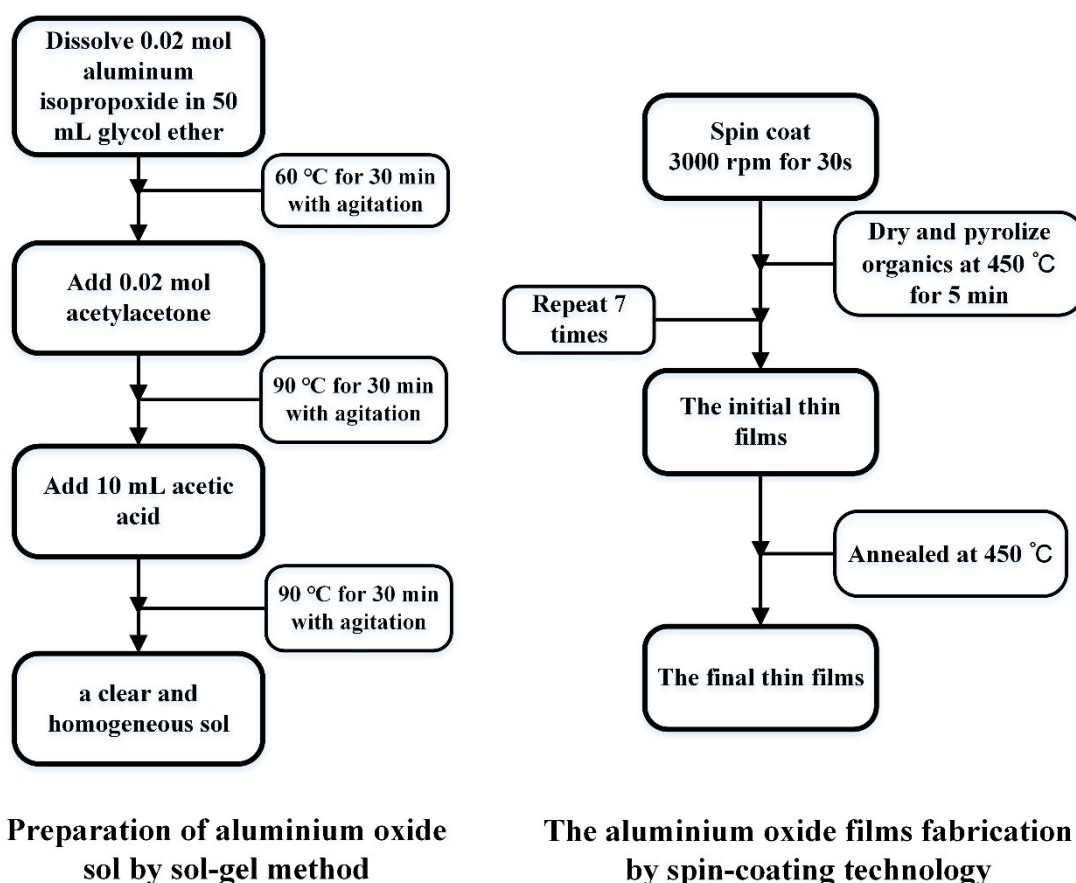


Fig. S1 The preparation of AmAO thin film

2. The interfaces of Au/AmAO and Al/AmAO

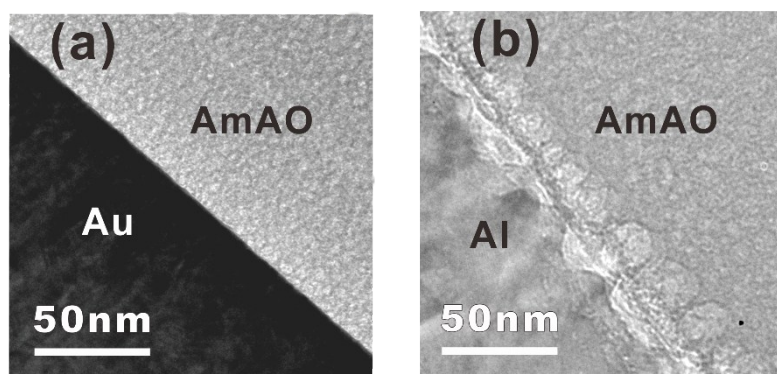


Fig. S2 The comparative interfaces of (a) Au/AmAO and (b) Al/AmAO

3. Electric equivalent circuit

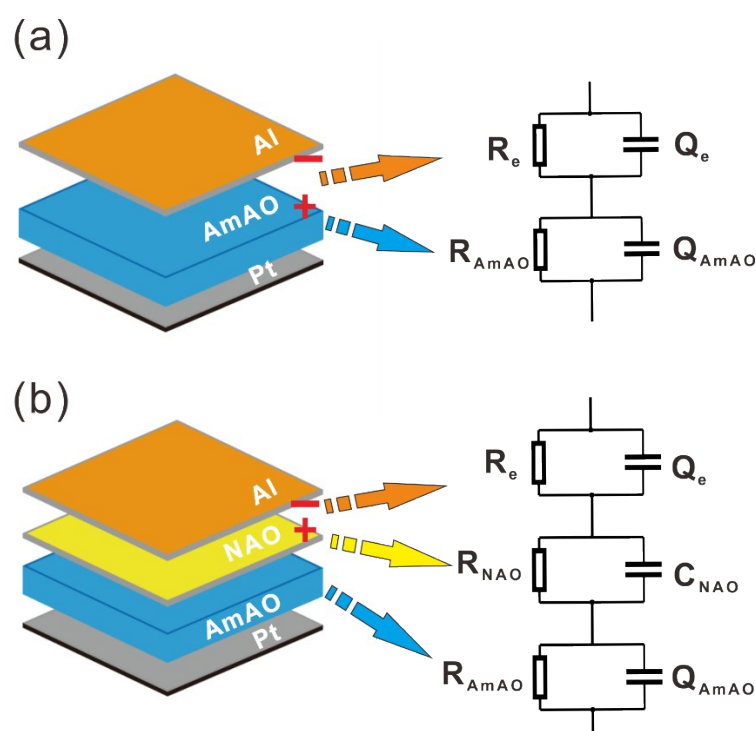


Fig. S3 Schematic representations and the corresponding electric equivalent circuits of capacitor (a) before and (b) after applying voltage

Various equivalent circuits have been proposed in the aqueous electrolysis system. One of the most widely used models is $R_e(R_pC_p)(R_bC_b)$ ¹. Here, R_e is the electrolytic resistance; R_p and C_p describe the resistance and capacitance of outer porous layer; R_b and C_b are the corresponding resistance and capacitance of the barrier layer. Differing

from the aqueous electrolysis system, the electrolyte and aluminum/electrolyte interfacial characteristics have changed owing to solid electrolyte (AmAO layer) replacing aqueous electrolyte. Hence, the amendment of equivalent circuit is inevitable. The circuit consists of two or three (RC) elements, which are corresponded to electric double layer, AmAO and/or NAO layer. However, on account of a large number of defect charges existing in double electric layer and AmAO layer, the constant phase angle element (Q) is also considered to instead of a pure capacitance (C) in the two elements. Namely, the derivative equivalent circuits are $(Q_e R_e)(Q_{AmAO} R_{AmAO})$ before applying voltage and $(Q_e R_e)(C_{NAO} R_{NAO})(Q_{AmAO} R_{AmAO})$ after applying voltage, as shown in Fig. S3. R_e is the double electric layer resistance and Q_e describes the properties of double electric layers. Q_{AmAO} represents the property of AmAO layer and R_{AmAO} is the corresponding resistance of AmAO. R_{NAO} and C_{NAO} refer to the resistance and capacitance properties of NAO layer, respectively.

4. TG-DSC of AmAO thin film

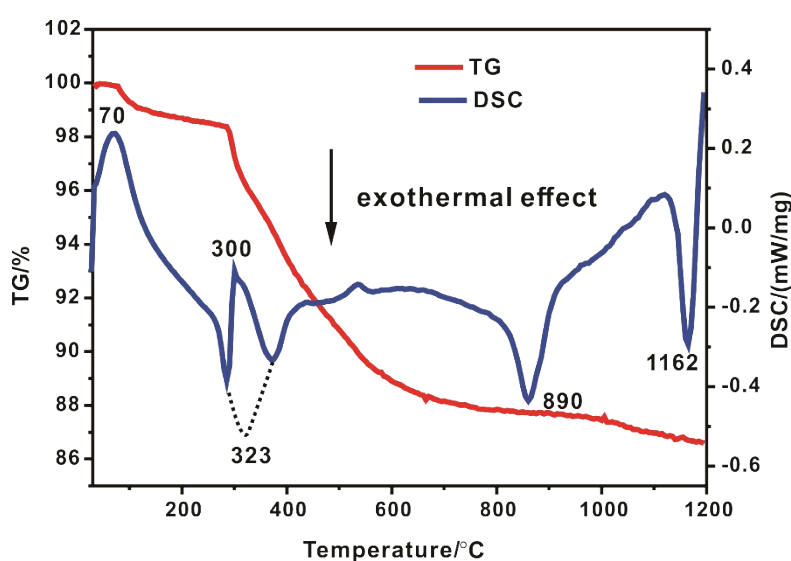
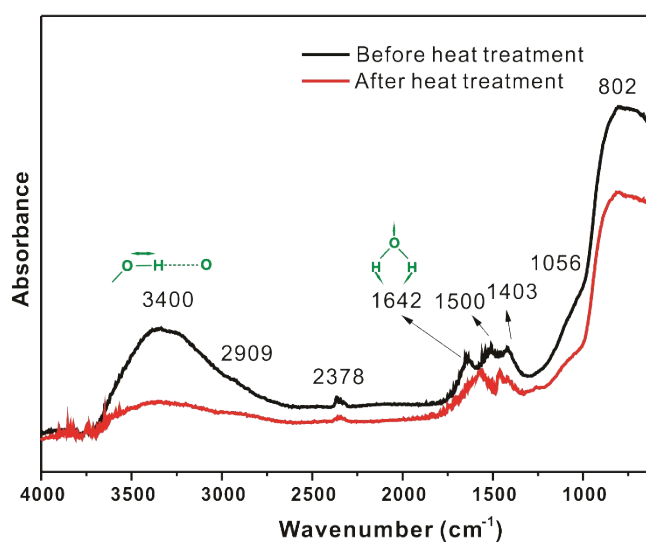


Fig. S4 TG-DSC curves of AmAO thin film

Tab. S1 Temperatures and their assignments for DSC of AmAO layer

Temperatures (°C)	Assignments
70	The desorption of physical absorption water
300	The desorption of chemical absorption water
323	Transformation from boehmite to γ -alumina
890	Transformation from γ -aluminium oxide to θ + α -aluminium oxide
1162	Transformation from θ + α -aluminium oxide to α -aluminium oxide

5. The FT-IR spectra of AmAO layer

**Fig. S5** The comparative FT-IR spectra of specimens before and after heat treatment at 100 °C for 1800 s**Tab. S2** Frequencies and their assignments for FT-IR of aluminum oxide thin film

Wavenumber (cm ⁻¹)	Assignments
802	ν_s (AlO ₆) ³⁻⁴
1056	ν (Al-OH) ⁵
1403	γ (C-C) ⁶

1500	γ (C=O) ⁶
1642	γ (H-O-H) ⁷
2378	Physisorped CO ₂ molecules ⁶
2700-3700	ν (O-H) ⁷

Obviously, the intensity of the typical peaks of hydroxyl groups (located at 1642 and 3400 cm⁻¹) decreased indicating the large number of water molecules were removed.

6. References

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