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Stabilizing Na metal anode with NaF interface on spent cathode carbon from aluminum electrolysis

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1. Experimental section

Preparation of purified spent cathode carbon (PSCC): The spent cathode carbon (SCC) was purified by the alkali. In a typical process, 18.0 g of sodium hydroxide was dissolved in 100.0 mL of distilled water and 10.0 g of SCC was added into the above solution. Then 0.5 mL of absolute ethyl alcohol was injected into the mixture and sonicated for 90 min. After rinsed with deionized water for five times and dried at 60 C for 24 h, the purified spent cathode carbon was obtained.

Electrochemical Measurement: SCC, conductive carbon (Super P) and polyvinylidene difluoride binder (PVDF) with a mass ratio of 8:1:1 were mixed in N-methyl-2-pyrrolidinone (NMP) solution for 20 min, after that the slurry is coated onto a Cu foil with doctor blade and subsequently dried in a vacuum oven at 80°C for 12 h. PSCC electrode was made by the same method above just replace SCC with PSCC. The loading mass of active materials was about $1.0 \sim 1.2$ mg cm⁻². Na foil and glass fiber filters (Whatman) were used as counter electrode and separator, respectively. And the electrolyte used in symmetric and half cell was 1.0 M NaCF₃SO₃ in bis(2-methoxyethyl) ether (DME). The amount of electrolyte used for test is about $120 \,\mu$ L for each cell. LAND electrochemical testing system were used to study the electrochemical performance. Electrochemical impendence spectroscopy (EIS) measurements were conducted on an electrochemical workstation (Gamry instruments), the frequency range operated 0.1Hz to 1.0 MHz and a voltage amplitude of 5 mV.

Characterization: The morphologies and element distributions of materials were probed by scanning electron microscope (SEM, MIRA3 TESCAN). Micro-structure of materials was observed by Transmission electron microscopy (TEM) and highresolution transmission electron microscopy (HRTEM) images were taken on a JEM-2100 (JEOL, Japan). X-ray diffraction tests were measured on RigakuMiniFlex 600a with Cu K_{α} radiation (λ =0.154056 nm). And the experiments were carried out with a scan rate of 5° min⁻¹ from 10° to or 80°. The X-ray photoelectron spectroscopy (XPS) spectra was obtained by Thermo ESCALAB 250 spectrometer.



Figure. S1 XPS spectra of PSCC



Figure. S2 (a) and (b) HRTEM images of SCC, insert is the SAED images of NaF in SCC (c) SEM

image of PSCC, (d) the corresponding element mapping images of PSCC.



Figure. S3 N_2 adsorption–desorption isotherms of (a) SCC and (c) PSCC; and the corresponding pore-size distribution of (b) SCC and (d) PSCC.



Figure. S4 The magnified figure of Fig. 2a.



Figure. S5 The selected voltage profiles in the plating/stripping process.



Figure. S6 CE values of the plating/stripping cycles of different electrodes at 5mA cm⁻² for 5mAh cm⁻².



Figure. S7 Voltage-time profiles of SCC and PSCC symmetric cells at 3 mA cm⁻² for 3 mA h cm⁻².



Fig. S8 EIS Nyquist plots of the symmetric cells.



Figure. S9 Tafel plot in exchange current density test.



Figure. S10 SEM images of SCC when plating for different capacity of Na at 0.1 mA cm⁻² (a) 1 mAh cm⁻², (b) 3 mAh cm⁻², SEM images of PSCC when plating for different capacity of Na at 0.1 mA cm⁻² (c) 1 mAh cm⁻², (c) 3 mAh cm⁻². The scale bar is 200 μ m.