Supplemental Information

Sodiated Na_xSnSb nanoparticles embedded in N-doped graphene sponges direct uniform Na nucleation and smooth plating for high efficiency Na metal batteries

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The SnSb/NG sponges were synthesized by a two-step process[1]. Typically, 4 mg heavily oxidized graphene oxide, 1.2 mg $SnCl_2 \cdot 2H_2O$, 1.5 mg $SbCl_5$ and 5 g urea were mixed in 20 mL water and treated at 180 ^oC in a hydrothermal autoclave for 10 hours. The graphene oxide sheets were reduced and doped with nitrogen by reacting with urea during this process and self-assembled into macroporous frameworks. This process produces $SnSbO_x$ nanocrystals that strongly attach to graphene sheets via defect-driven heterogeneous nucleation (Figure S1) [2]. The as-produced sponges were freeze-dried in order to preserve their macroporosity and then reduced under 4% H_2/Ar at 500 ^oC for 4 hours in the second step to fabricate the SnSb/NG sponge.

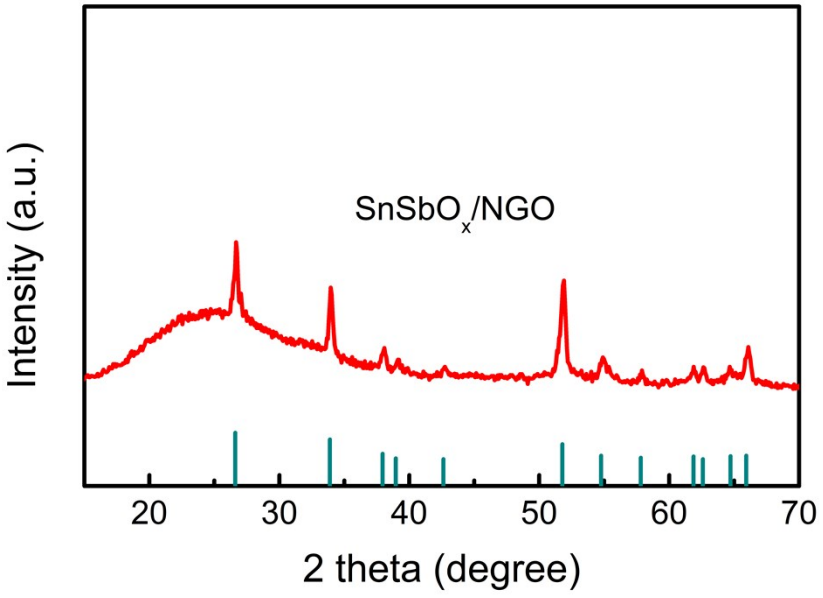

Figure S1 XRD pattern of the as-prepared 3D $SnSbO_x$/NGO composite.

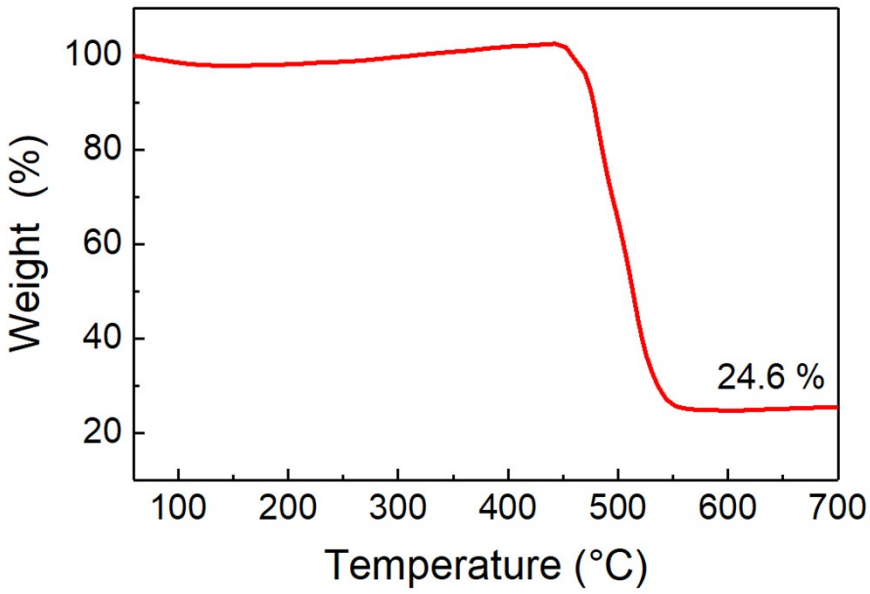

Figure S2 Based on TGA result profile of the SnSb/NG composite carried out from 50 to 700 °C in air condition, the SnSb NPs content in the composite was calculated to be approximately 20 wt%.[3]

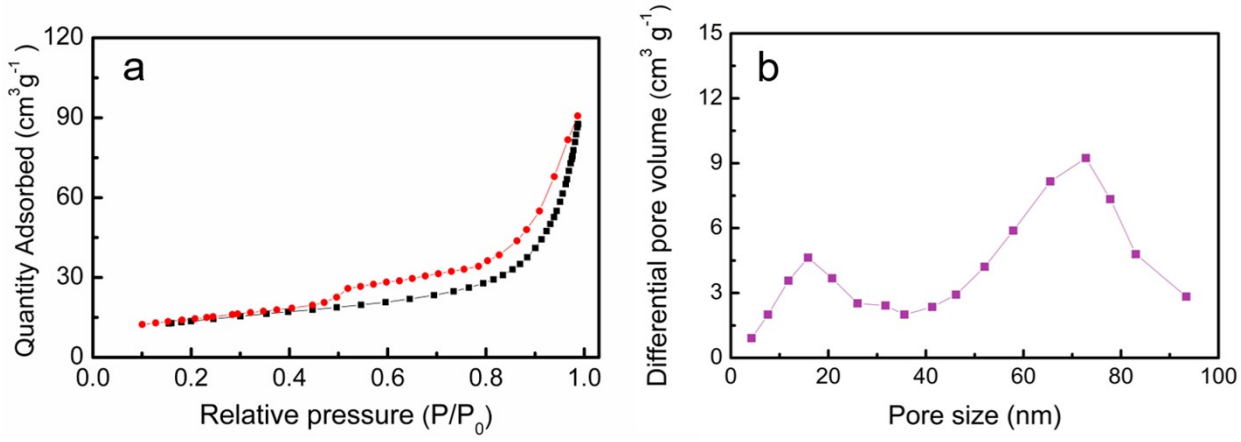

Figure S3 The nitrogen absorption-desorption isotherms and pore size distribution of the as-prepared 3D sponge.

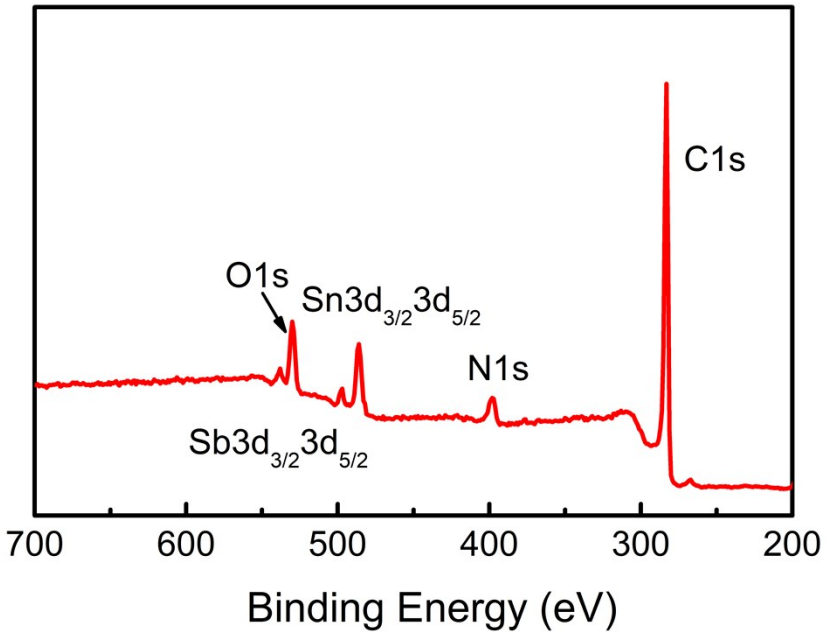

Figure S4 XPS spectrum of the as-prepared 3D $SnSbO_x$/NGO composite.

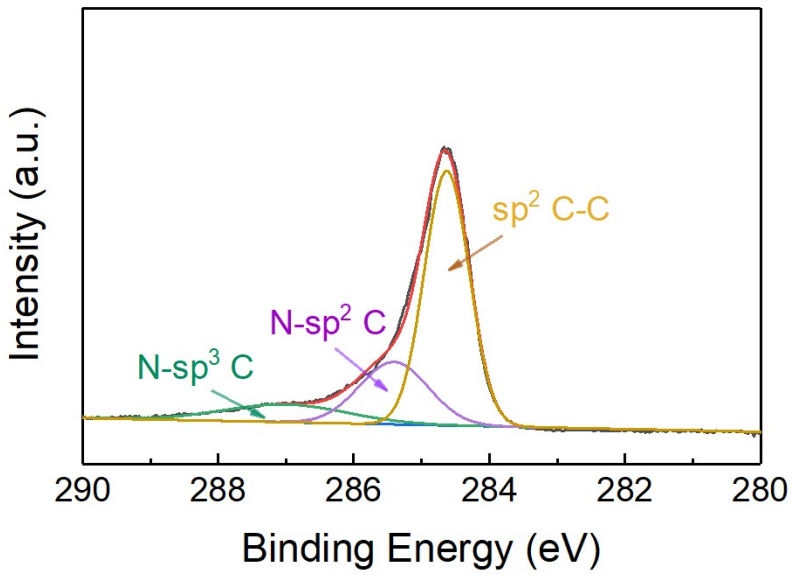

Figure S5 C1s XPS spectrum of the as-prepared composite.

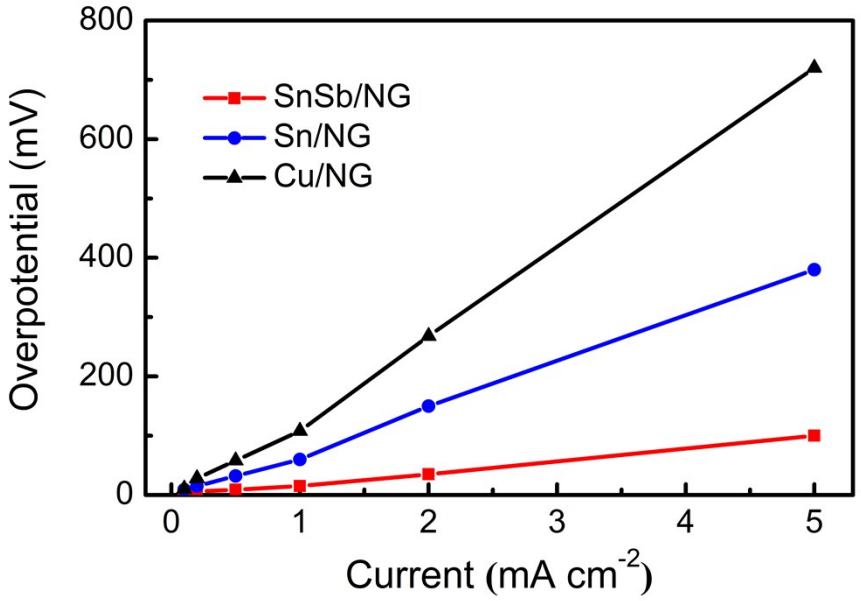

Figure S6 Comparison of the overpotential for Na nucleation on different substrate at the current from 0.01 to 5.0 mA cm^{-2}.

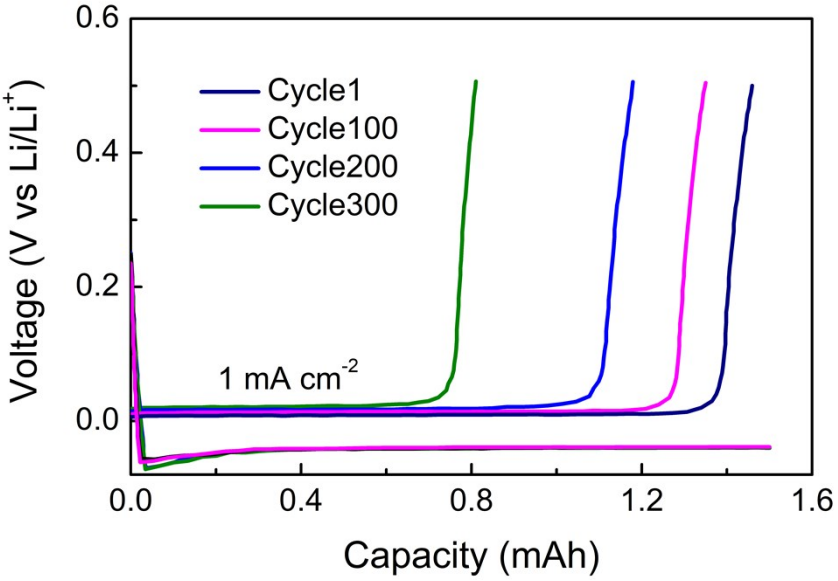

Figure S7 Voltage profiles during 300 cycles of repeated Na plating/stripping in Sn/NG, 1.5 mAh Na was plated for each cycle.

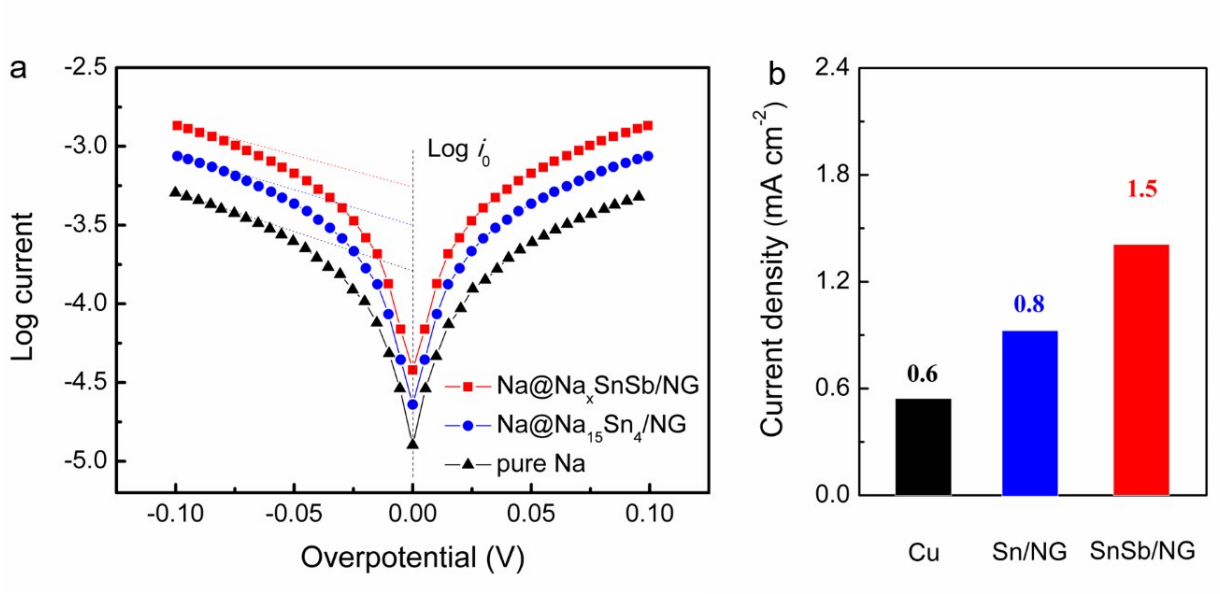

Figure S8 Comparison of Tafel plots of different anodes and the derived exchange current density.

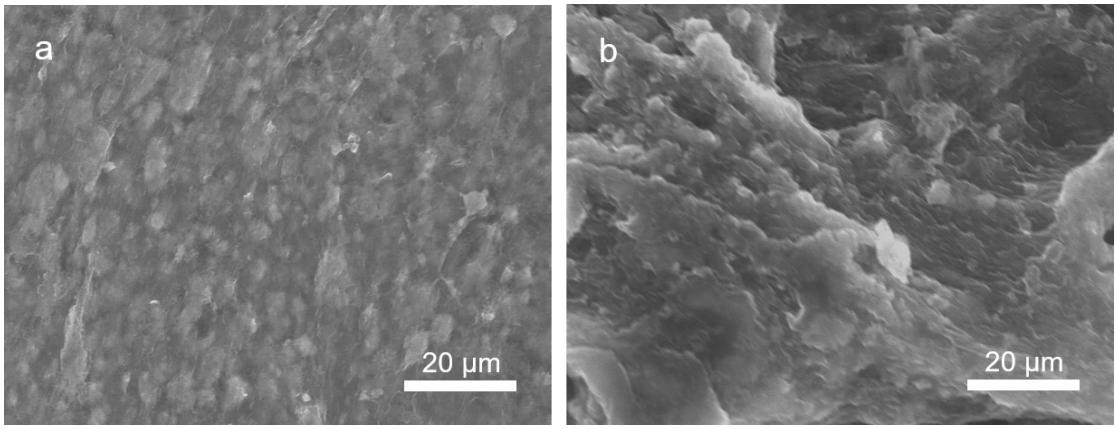

Figure S9 Comparison of SEM images of Na@Na_xSnSb/NG and pure Na electrode after 100 cycles

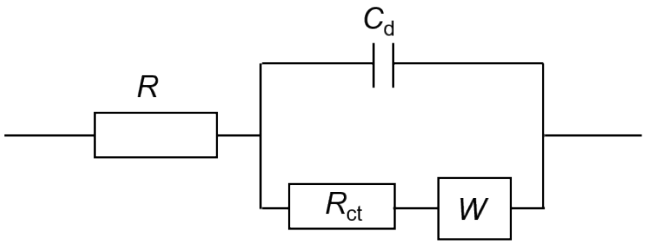

Figure S10: Equivalent circuit for modeling the electrochemical impedance spectrum. R: electrolyte resistance; C_d: contant phase elements; R_{ct}: charge transfer resistance at Li surface; W: Warburg impedance.

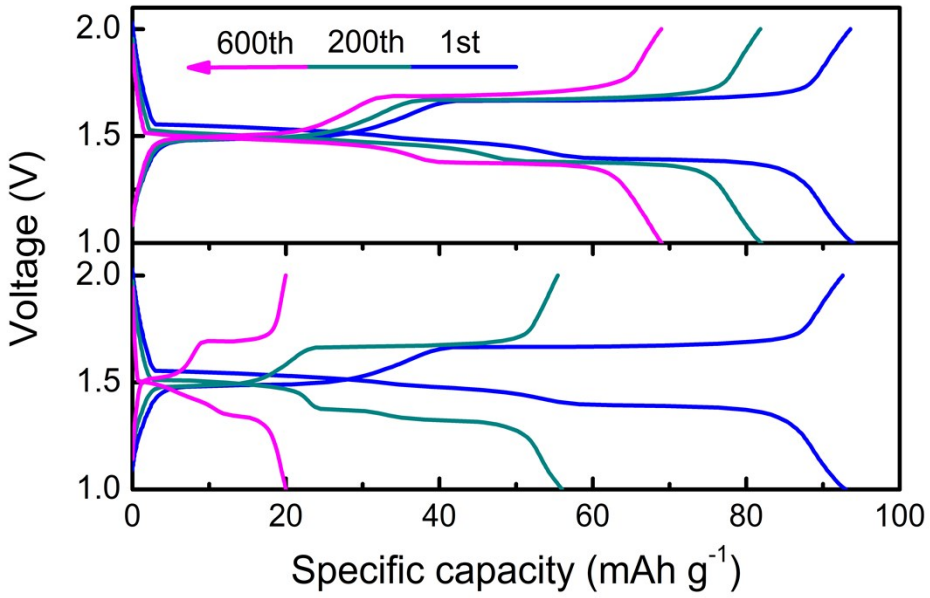

Figure S11 Voltage profiles of full cells tested at 1 C with Na@Na_xSnSb/NG and Na@$Na_{15}Sn_4$/NG anode.

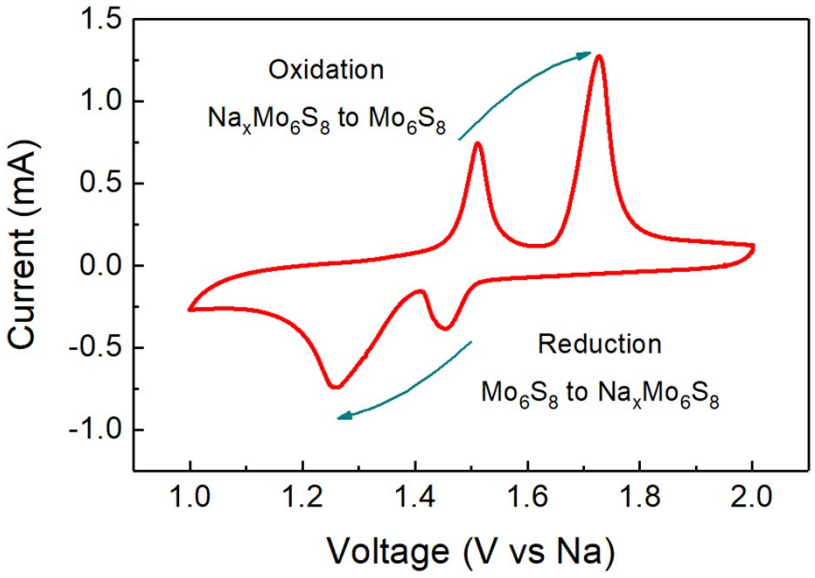

Figure S12 CV curve of the Na‖Mo_6S_8 full cell at a scan rate of 0.1 mV s^{-1}.

1. Y. Cheng, Y. Shao, L.R. Parent, M.L. Sushko, G. Li, P.V. Sushko, N.D. Browning and C. Wang, J. Liu, *Adv. Mater.,* 2015, **27**, 6598−6605.
2. X. Li, W. Qi, D. Mei, M.L. Sushko, I. Aksay and J. Liu, *Adv. Mater.* 2012, **24**, 5136−5141.
3. J. Qin, T. Wang, D. Liu, E. Liu, N. Zhao, C. Shi, F. He, L. Ma and C. He, *Adv. Mater.,* 2018, **30**, 1704670.