Supporting Information for

Low-temperature solid-state growth of three-dimensional bicontinuous nanoporous graphene with tunable porosity for lithium-sulfur batteries

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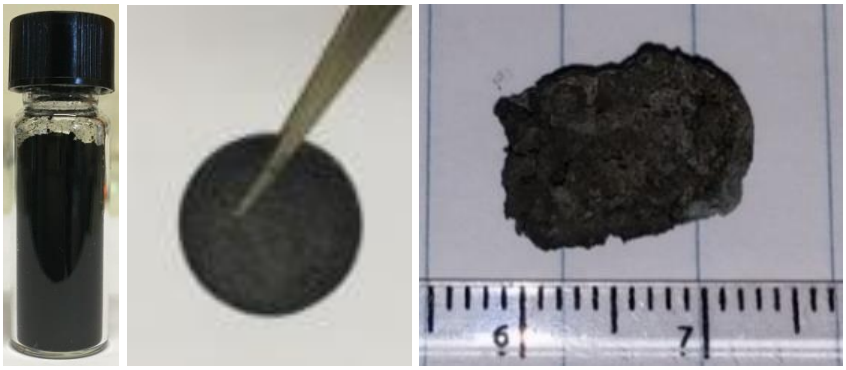

Fig. S1 Nanoporous graphene (NPG) in the forms of powder (left), chip (middle) and bulk (right).

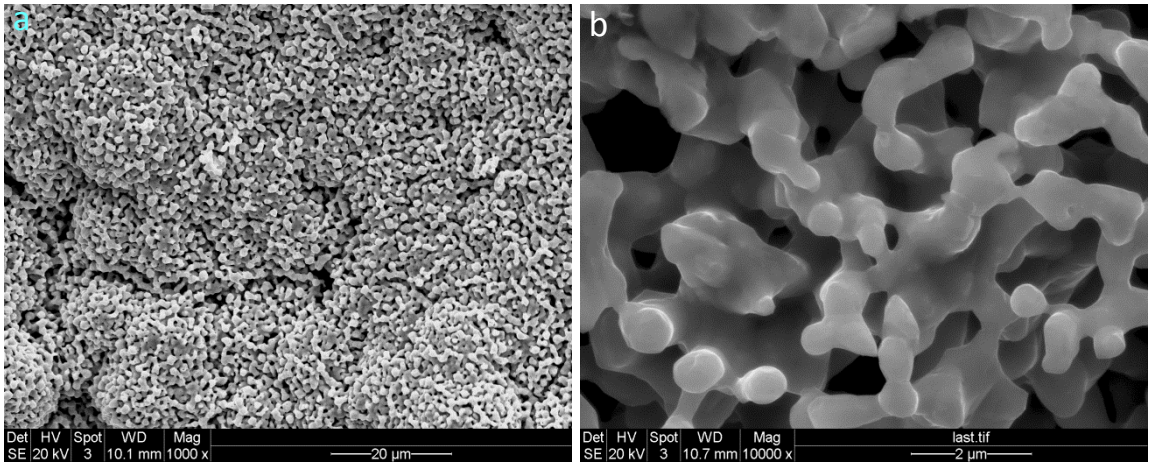

Fig. S2 SEM micrographs showing NPNi-1000 synthesized at 600 °C: (a) overview and (b) close view.

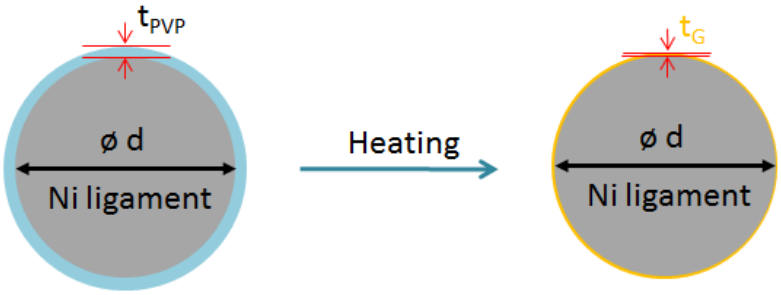

Fig. S3 Schematic of PVP coating on Ni ligament and converted to graphene (G) thin film.

Calculation of PVP coating converted to graphene film on Ni:

Below conditions are required: suppose all the Ni ligaments were equivalent to a long Ni wire of diameter *d* and length *l*. Thin PVP coating and graphene film can be considered as very thin tube compared with Ni ligaments. Thus, the thickness of PVP coating t_{PVP} and the thickness of graphene film t_G can be ignored when calculate the volume of PVP and graphene film because $d >> t_{PVP} > t_G$. The solubility of carbon follows Eq. 1:

$$\ln S = 2.480\text{-}4880/T \quad (1)$$

where S is the solubility in grams of carbon per 100 grams of nickel, *T* is the temperature (K).

The volume of Ni ligaments (V_{Ni}) can be presented in the following equation:

$$V_{Ni} = l \times \pi \times d^2 \times \frac{1}{4} \quad (2)$$

Then, the mass of Ni ligaments (M_{Ni}) can be estimated with Eq. 3,

$$M_{Ni} = \rho_{Ni} \times l \times \pi \times d^2 \times \frac{1}{4} \quad (3)$$

Similarly, we have the volume of PVP coating (V_{PVP}) in the following Eq. 4

$$V_{PVP} = l \times \pi \times d \times t_{PVP} \quad (4)$$

and the mass of PVP (M_{PVP}) in Eq. 5:

$$M_{PVP} = \rho_{PVP} \times l \times \pi \times d \times t_{PVP} \quad (5)$$

The volume of graphene film (V_G) follows Eq. 6:

$$V_G = l \times \pi \times d \times t_G \quad (6)$$

The mass of graphene film (M_G) follows Eq. 7:

$$M_G = \rho_{graphite} \times l \times \pi \times d \times t_G \quad (7)$$

The mass of carbon after heat treatment at 800 °C is 4.1 wt.% of PVP by heating PVP under H_2/Ar using same condition with heat treatment of NPNi/PVP. So, the mass of carbon (M_C) generated from PVP at 800 °C is

$$M_C = 0.041 \times M_{PVP} \quad (8)$$

The solubility of C in Ni is ~0.126 g at 800 °C calculated based on Eq.1. So, when the carbon in Ni at 800 °C is saturated, the amount M_{C0} of carbon dissolved in Ni ligaments of M_{Ni} equals to 0.00126*M_{Ni}.

When all the solid carbon from PVP decomposition converted to graphene,

$$M_C = M_G \leq M_{c0} = 0.00126\, M_{Ni} \quad (9)$$

When M_G=0.00126 M_{Ni},

$$\rho_{graphite} \times l \times \pi \times d \times t_G = 0.00126 \times \rho_{Ni} \times l \times \pi \times d^2 \times \frac{1}{4} \quad (10)$$

So,

$$\rho_{graphite} \times t_G = 0.00126 \times \rho_{Ni} \times d \times \frac{1}{4} \quad (11)$$

When $\rho_{graphite} \approx 2.26$ g cm^{-3} and $\rho_{Ni} = 8.908$ g cm^{-3}, we have

$$t_G \approx 1.24 \times 10^{-3} d \quad (12)$$

When the maximum ligament size is 1500 nm, t_G is ~1.9 nm.

Also, we have $M_G = 0.041\, M_{PVP}$,

So,

$$\rho_{graphite} \times l \times \pi \times d \times t_G = 0.041 \times \rho_{PVP} \times l \times \pi \times d \times t_{PVP} \quad (13)$$

When $\rho_{pvp} = 1.2$ g cm^{-3}, we have the relationship between PVP and graphene,

$$t_G \approx 0.0218\, t_{pvp} \quad (14)$$

Also, we have $t_G = 0.34\, n$, where *n* is the number of graphene layer. So

$$n \approx 0.064\, t_{pvp} \quad (15)$$

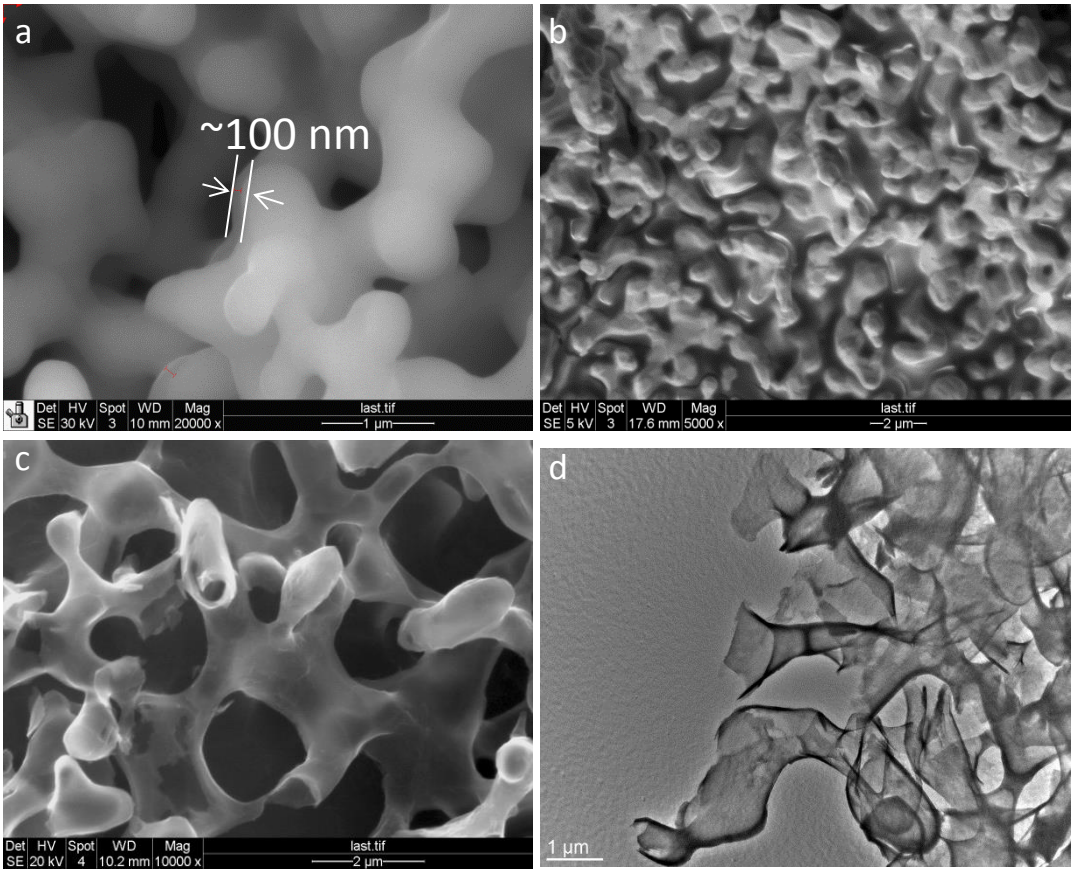

Fig. S4 (a) PVP coated nanoporous Ni by dipping the nanoporous Ni in 0.25 g ml^{-1} of PVP solution, (b) PVP coated nanoporous Ni by dipping the nanoporous Ni in 0.4 g ml^{-1} of PVP solution, (c and d) SEM and TEM of nanoporous graphene with thick walls obtained by using 0.4 g ml^{-1} of PVP solution.

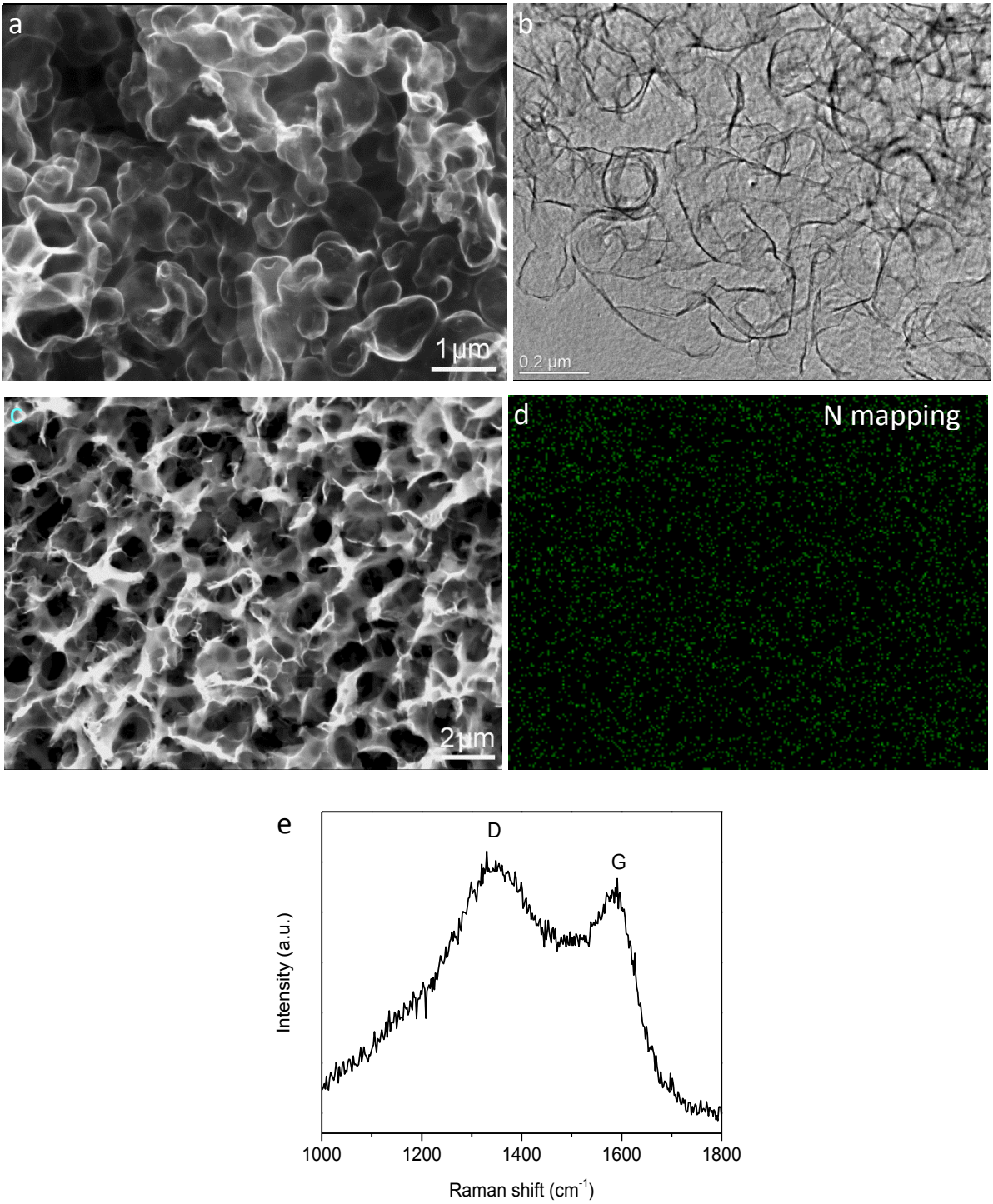

Fig. S5 (a and b) SEM image and TEM image of the NPG prepared by using sucrose as the precursor; (c and d) SEM and N mapping of NPG prepared by using PVP and dicyandiamide as carbon sources. (e) Raman spectra of N-doped NPG.

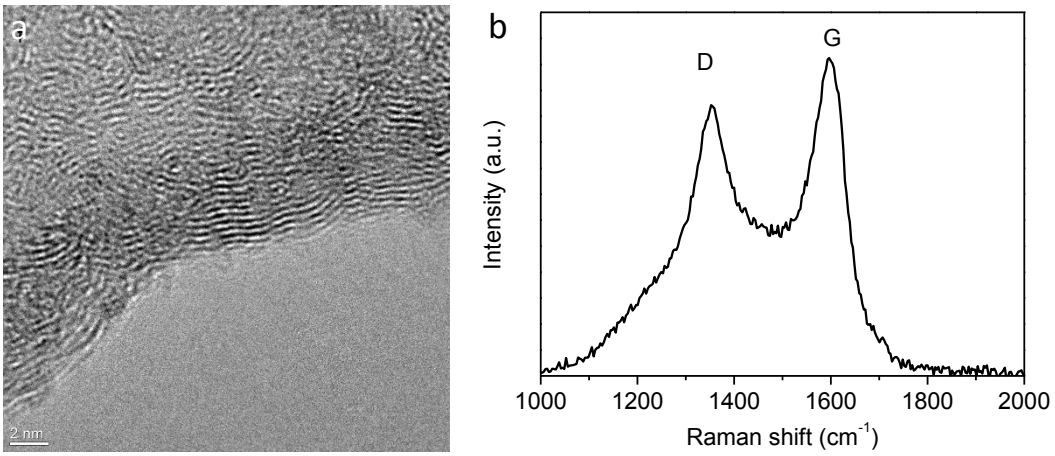

Fig. S6 (a) HR-TEM and (b) Raman spectrum of nanoporous graphene obtained at 700 °C, showing the disordered layer structure and low graphitization.

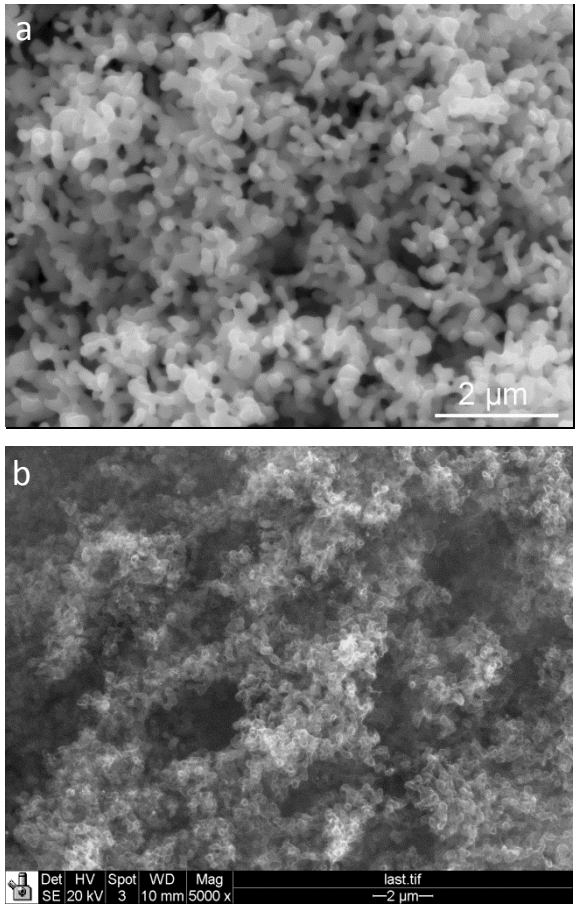

Fig. S7 (a) SEM of nanoporous Ni with average ligament size ~160 nm, (b) low magnification SEM image of NPG-160.

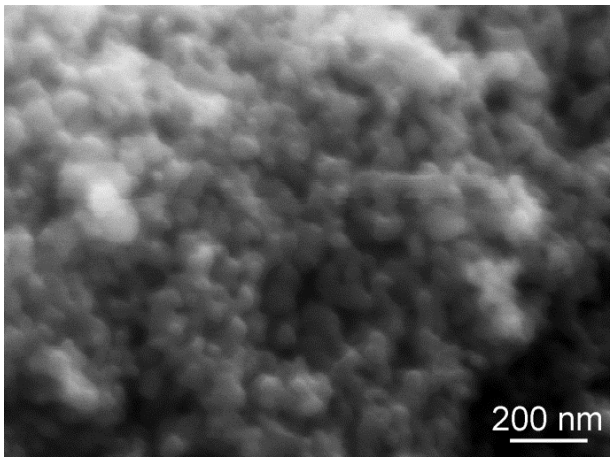

Fig. S8 SEM micrograph of nanoporous Ni with average ligament size 10-50 nm.

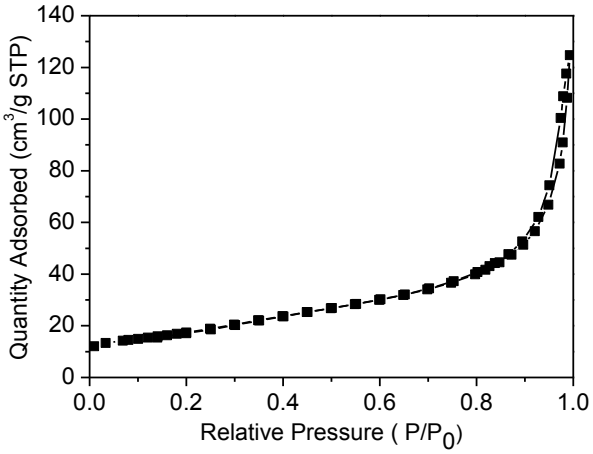

Fig. S9 Nitrogen adsorption- desorption isotherm of carbon black