Supporting Information for

Spider-Web Inspired Graphene Skeleton-Based High Thermal Conductivity Phase Change Nanocomposites for Battery Thermal Management

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Supplementary Figures

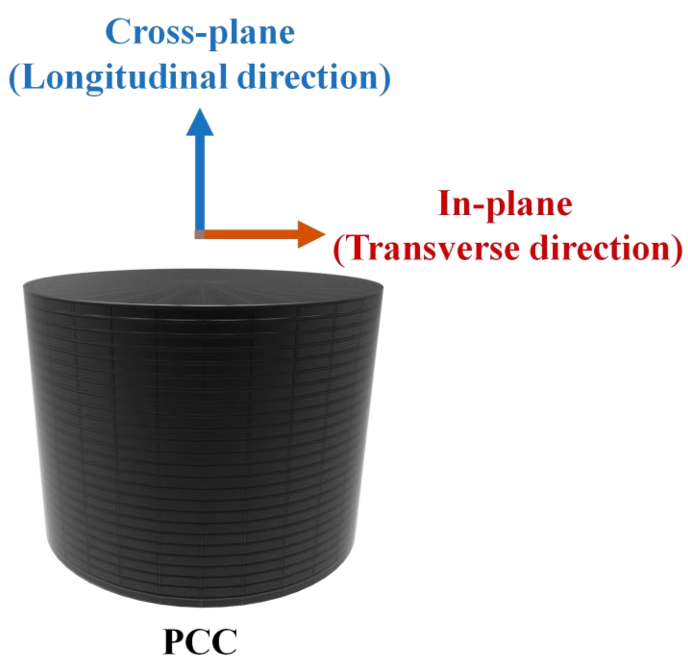

Fig. S1 Cross-plane and in-plane thermal conductivities of PCCs. Spider web-like structure and concentric ring structure are parallel to the in-plane direction.

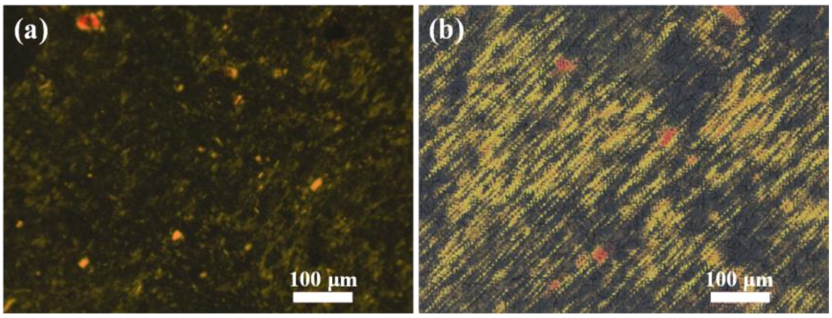

Fig. S2 POM images of 0.5 wt% **a** pure GO suspension and **b** GO suspension with 0.135 M KOH

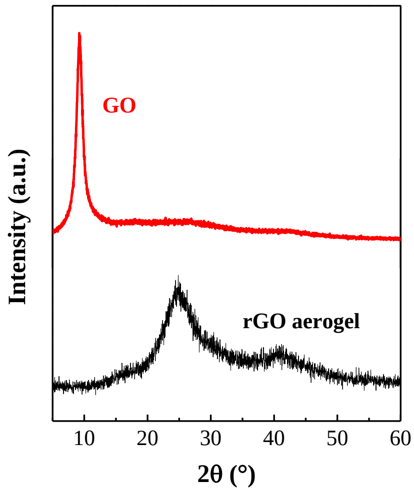

Fig. S3 X-Ray Diffraction (XRD) analysis of GO and freeze-dried rGO hydrogel (aerogel)

As shown in Fig. S3, obvious differences can be observed from XRD results between GO and freeze-dried hydrogel (aerogel). After hydrothermal reaction of GO, the XRD peak of GO at ~10° disappeared, which demonstrated that GO was partially reduced during hydrothermal reaction [S1].

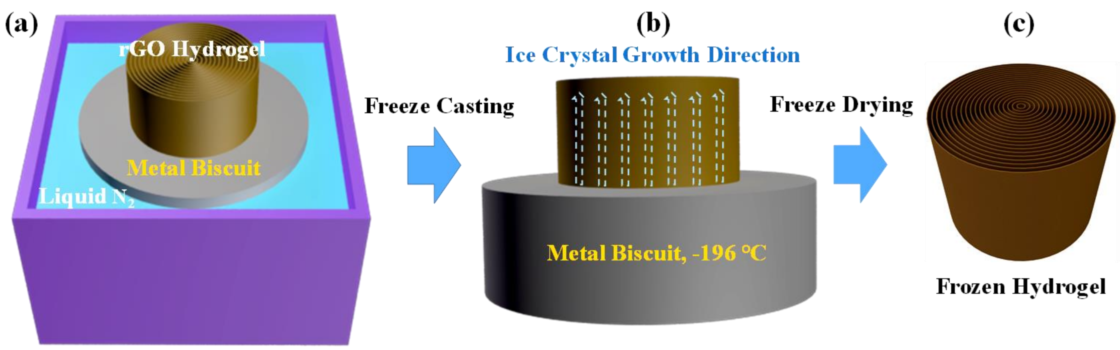

Fig. S4 Schematic diagram of unidirectional freeze casting for GO hydrogel. Schematic diagram of **a** unidirectional freeze casting device, **b** GO hydrogel was placed on a precooled metal block, and **c** frozen GO hydrogel

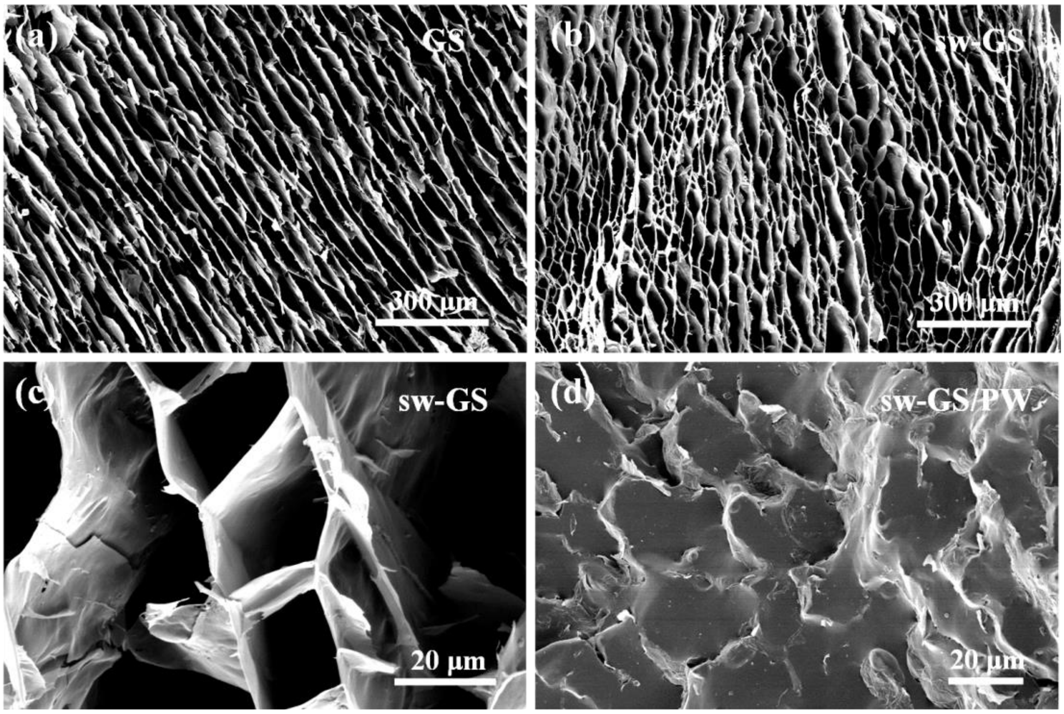

Fig. S5 SEM images of **a** GS, **b**, **c** sw-GS and **d** sw-GS/PW

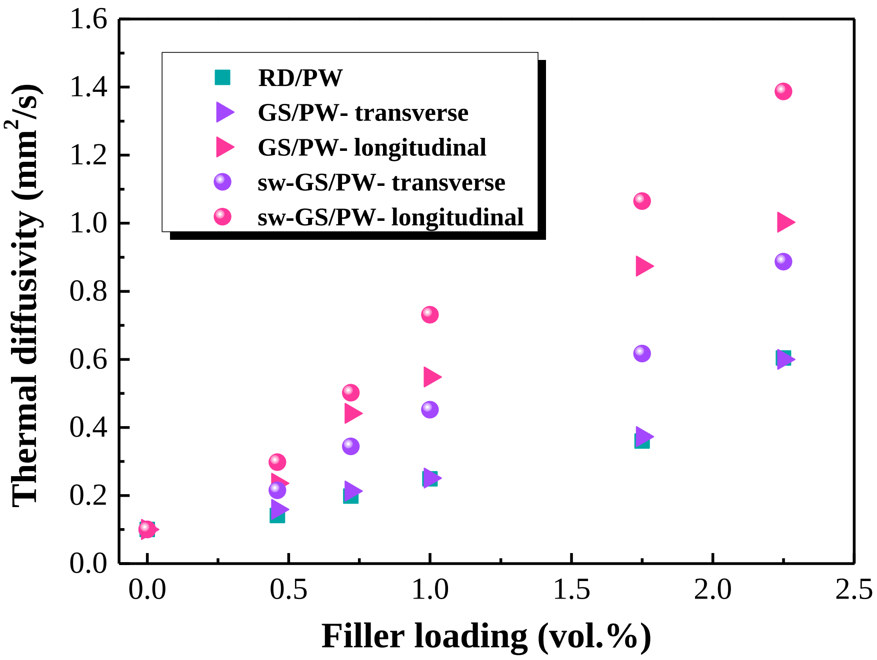

Fig. S6 Thermal diffusivity (at 25 °C) of different PCCs at different filler loading

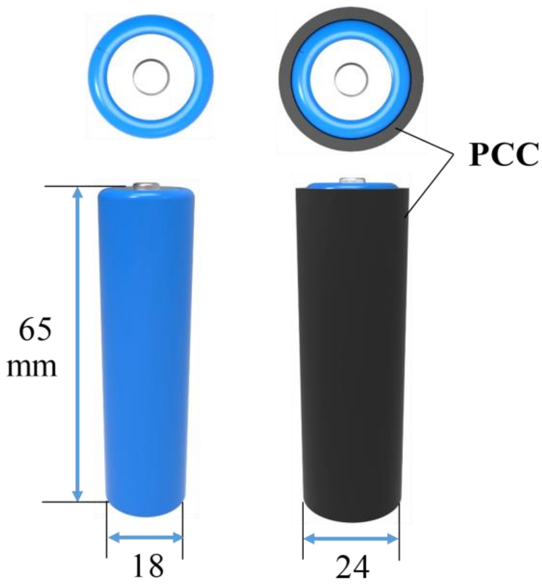

Fig. S7 Schematic diagram of commercial lithium-ion batteries used in thermal management application. Left: battery without wrap; Right: battery with wrap

Finite Element Model

The heat transfer processes of the three PCCs (RD/PW, GS/PW and sw-GS/PW composites) with different filler structures were modeled in Fenics (FEM) [S2]. A simplified model was employed to analyze the temperature and heat flux magnitude distribution of the three PCCs during heating. The heat sources and heat sinks were applied on the left and right surfaces of PCCs, respectively. The periodic boundary conditions were applied on the other surfaces. According to the previous test results and reported results, the κ of graphene oxide sheets and paraffin wax were 350 W/m·K and 0.19 W/m·K, respectively. The filler (skeleton) content of three PCCs was ~10 vol%. In addition, as shown in Fig. S8, the transverse thermal conductivity values of the PCCs based on the model clearly followed the tendency of sw-GS/PW > GS/PW > RD/PW, which is different from experimental results (sw-GS/PW > RD/PW > GS/PW). This could result from the filler (sw-GS powder) retaining some of the interconnected structure of sw-GS in the RD/PW composites. However, in the FEM simulation model, randomly dispersed GO sheets were simulated as the fillers within the RD/PW composites.

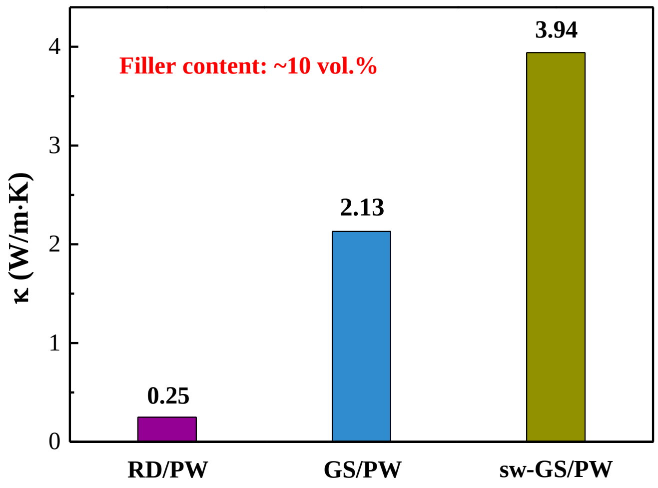

Fig. S8 Simulated thermal conductivities of the PCCs based on the simulation model. The filler (skeleton) content is ~10 vol%.

Supplementary References

[S1] Y. Wang, S. Chou, H. Liu, S. Dou, Reduced graphene oxide with superior cycling stability and rate capability for sodium storage. Carbon **57**, 202-208 (2013). https://doi.org/10.1016/j.carbon.2013.01.064

[S2] M. S. Alnæs, J. Blechta, J. Hake, A. Johansson, B. Kehlet et al., The fenics project version 1.5. Archive of Numerical Software **3**, 9-23 (2015). https://doi.org/10.11588/ans.2015.100.20553