Supplementary Material

Graphene-Silver Hybrid Nanoparticles Embedded Phase Change Materials for Enhanced Thermal Management of Lithium-Ion Batteries

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1) Determination of thermophysical properties of PCM and NP-impregnated PCMs at a loading concentration of 0.1 wt%.

Average Density: The weighted average densities of the base PCM, PCM-GNPs, and PCM-GNP-Ag NPs were calculated by utilizing the individual densities provided by the manufacturers for paraffin wax, stearic acid, silver, and GNPs. The weighted average densities for each of these compositions were calculated using the effective density model introduced by Yu et al. [1].

Dynamic viscosity: The dynamic viscosity measurements of base PCM and those loaded with GNPs and GNP-Ag hybrid NPs were conducted using a Rheometer (Brookefield LVDV-III Ultra Cone Plate Rheometer, USA) to controlled temperature conditions within the range of 60°C to 75°C, covering the phase transition temperature range of the PCM. The average viscosity values recorded between 60 to 70 ºC are mentioned in Table 2 (of the main manuscript). The instrument employs the principle of rotational viscometry, where the torque required to rotate a cone spindle immersed in the sample is measured. It calculates the dynamic viscosity based on the applied torque and the rotational speed of the spindle. The accuracy of the viscosity values obtained using the Rheometer with Cone Spindle CPE-40 and a sample volume of 0.5 mL is within ± 1% of the Full Scale Range (FSR) recorded viscosity value, where FSR varies with cone spindle and rotational speed selection. The values reported are the average of multiple measurements taken at each temperature point, with a shear rate of 7.5 s^{-1}.

Analyzing the dynamic viscosity values across different temperatures reveals the temperature-dependent behavior of the PCM samples. Comparing the dynamic viscosity of the PCM with those loaded with GNPs and GNP-Ag hybrid NPs highlights the influence of NP addition on the increase in the viscosity as shown in Fig.S1, indicating potential alterations in the fluid dynamics and rheological properties of the PCM mixtures.

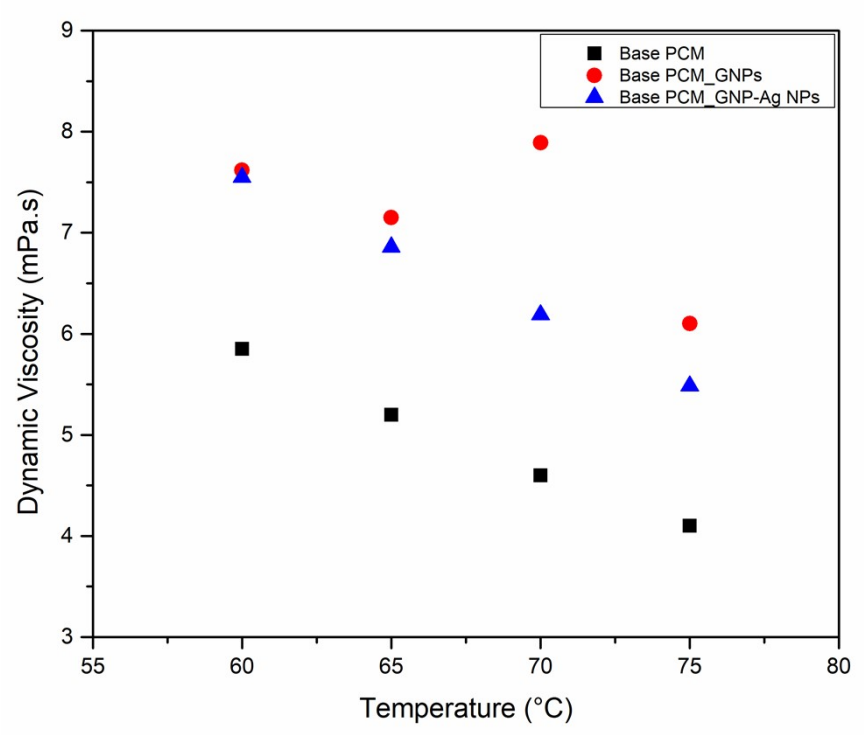

Fig. S1. Measured dynamic viscosity values of base PCM, and PCMs loaded with 0.1 % GNPs and GNP-Ag hybrid NPs at different temperatures.

Across all PCM variants, a consistent decrease in dynamic viscosity with increasing temperature was observed. Interestingly, while both GNPs and GNP-Ag hybrid NPs led to increased viscosity compared to the base PCM, the magnitude of this effect varied between the two nanoparticle types. Specifically, at 70°C, PCM-GNPs exhibited a higher dynamic viscosity than PCM-GNP-Ag NPs due to the potential influence of aggregate GNP morphology and composition on viscosity behavior.

Thermal Conductivity: The TPS 500 S instrument employs the transient planar source method for thermal conductivity measurements, generating a brief heat pulse within the Kapton sensor via Joule heating. The measurement process utilized parameters such as an initial temperature of 33°C, an output power of 25 mW, a measurement time of 5 s, a radius of 3.189 mm, and a thermal coefficient of resistance (TCR) of 0.004930 K^{-1}. These parameters were crucial for accurately determining the thermal conductivity of the sample. As the pulse

traverses through the sensor and into the sample material, it induces temperature changes, measured by temperature sensors embedded in the Kapton sensor. The thermal conductivity of the sample under study was determined with an accuracy better than 5%. This measured thermal conductivity value is a critical indicator of the material's heat transfer characteristics.

The base PCM's thermal conductivity is recorded to be 0.270 $W.m^{-1}K^{-1}$. Incorporating 0.1 wt% GNPs increases this to 0.301 $W.m^{-1}K^{-1}$, while adding 0.1 wt% GNP-Ag hybrid NPs further enhances it to 0.325 $W.m^{-1}K^{-1}$. These reference values represent distinct compositions and illustrate the effect of additives on improving thermal conductivity. GNPs, for instance, are known for their high thermal conductivity; when added to the PCM, they enhance their thermal conductivity. Similarly, Ag NPs possess excellent thermal conductivity properties, and when combined with GNPs, they further boost the material's ability to conduct heat. These additives create pathways for more efficient heat transfer within the material, thus resulting in higher overall thermal conductivity values.

Specific heat capacity and latent heat of solidification: Specific heat capacities (liquid and solid) and latent heat of solidification of all the samples were calculated using the T-history method as described by Radhakrishnan and Sobhan [2]. The calculated base PCM-specific heat capacity values are 2.16 $J.g^{-1}K^{-1}$ and 5.01 $J.g^{-1}K^{-1}$ for liquid and solid, respectively. By incorporating 0.1 wt% GNP these values increased to 2.64 $J.g^{-1}K^{-1}$ and 5.14 $J.g^{-1}K^{-1}$, respectively. Further for 0.1 wt% GNP-Ag hybrid NPs, the values increased to 2.43 $J.g^{-1}K^{-1}$ and 5.56 $J.g^{-1}K^{-1}$ for the corresponding liquid and solid phases. The improvements observed are attributed to the increased surface area offered by the NPs within the PCM matrix. This enhanced surface area fostered efficient heat transfer processes due to stronger PCM molecules and NPs interactions. Consequently, thermal conductivity is improved, leading to higher specific heat capacities in both liquid and solid phases. Additionally, the calculated latent heat of solidification for the base PCM was determined to be 147 $J.g^{-1}$. However, with the inclusion

of 0.1 wt% GNP and GNP-Ag hybrid NPs, this value decreased to 140 $J.g^{-1}$ and 144 $J.g^{-1}$, respectively. This reduction is beneficial for enhancing the thermal performance of PCMs.

References

1 W. Yu, D. M. France, S. Choi and J. L. Routbort, *OSTI OAI (U.S. Department of Energy Office of Scientific and Technical Information)*, DOI:https://doi.org/10.2172/919327.

2 N. Radhakrishnan and C. B. Sobhan, *Heat and Mass Transfer*, 2022, **58**, 1811–1828.