Supporting information

Morphology-driven multifunctionality: Tailoring ZnO for enhanced absorption dominant EMI shielding and energy harvesting in PVDF/MWCNT nanocomposites

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S1. Dispersion analysis test for nanofillers (MWCNT and ZnO)

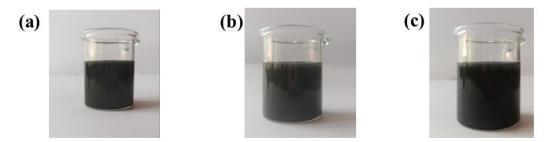


Fig. S1 Dispersion of MWCNTs in DMAc with PVP (a) immediately after probe sonication (b) 24 hours after probe sonication (c) 3 days after probe sonication.

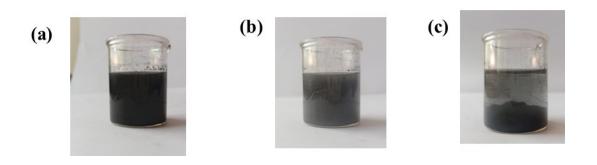


Fig. S2 Dispersion of MWCNTs in DMAc without PVP (a) immediately after probe sonication (b) 24 hours after probe sonication (c) 3 days after probe sonication.

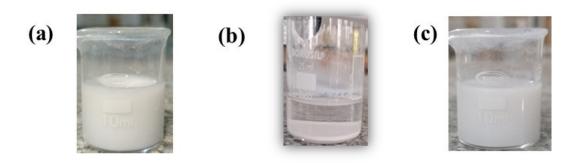


Fig. S3 Dispersion of ZnO nanoparticles in DMAc solvent (a) immediately after probe sonication without PVP (b) after 30 min probe sonication without PVP (c) after 24 hrs probe sonication with PVP.

To improve the dispersion of multi-walled carbon nanotubes (MWCNTs) and ZnO nanostructures within the PVDF matrix when solution blending is employed for the preparation of hybrid nanocomposites, first, the dispersion stability of MWCNT and ZnO nanoparticles is tested in the chosen solvent namely DMAc. Several strategies have been explored and reported in the literature for improving the dispersion of carbon-based fillers in polymer matrices, including polymer wrapping, surface functionalization, and the use of surfactants. ¹ Among these, surfactant-assisted dispersion offers a non-destructive and efficient approach, enhancing interfacial adhesion between filler particles and the polymer matrix. In contrast, surface functionalization, though effective, can sometimes alter or degrade the inherent properties of the carbon nanostructures, making it a less favourable approach for applications where the structural integrity of the filler is crucial.² The use of PVP as a surfactant in this study effectively addresses dispersion challenges, ensuring homogeneous filler distribution in the solvent and, consequently, within the polymer matrix.

In a typical experiment, 0.05 g of MWCNTs was probe-sonicated separately for 30 minutes in 8 mL of N,N-dimethylacetamide (DMAc) without any surfactant. It was observed that the nanotubes rapidly settled out of the solution, indicating poor dispersion in the chosen solvent. In contrast, when 1 wt.% of PVP was introduced into the system, the dispersion remained stable for an extended period, showing no visible agglomeration even after three days. PVP is known for its strong affinity towards carbon nanostructures, as reported in the literature, and acts as an effective dispersing agent. ³ The improved dispersion of MWCNTs in DMAc with 1 wt.% PVP is clearly illustrated in Fig. S1a–c, where uniform distribution is observed even after three days. Conversely, Fig. S2a–c demonstrates the rapid sedimentation of MWCNTs in the absence of PVP, where noticeable settling occurs within 24 hours.

In a similar fashion ZnO nanoparticles are dispersed in DMAc solvent after one hour probe sonication with and without 1 wt.% PVP. It can be seen from Fig. S3b that neat ZnO nanoparticles settle fast within half an hour, whereas with 1 wt.% PVP loading, the dispersion is stable after a day also as shown in Fig. S3c. Thus, addition of PVP improves the dispersion of both MWCNT and also ZnO. Hence, in this work, PVP has been used to effectively disperse the nanoparticles.

S2. Schematic diagram of PVDF-MWCNT-ZnO hybrid nanocomposite-based devices for piezoelectric and triboelectric measurements

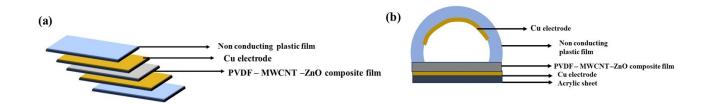


Fig. S4 Schematic diagram of device fabrication with PVDF–MWCNT-ZnO hybrid nanocomposite for (a) piezoelectric (b) triboelectric measurements.

The device fabrication for piezoelectric measurement with PVDF-7 wt.% MWCNT-ZnO hybrid nanocomposite is shown in Fig. S4a. The hybrid nanocomposite is sandwiched between two copper electrodes. On top of the copper electrodes acrylic sheet of same area is kept. For triboelectric measurement, the device comprises of arched acrylic sheet with copper electrode kept inside which will be sticked on the side of the active material. The active material is kept on the copper electrode and acrylic sheet as depicted in Fig. S4b. Finger tapping will be imparted on top of the curved acrylic sheets so that the contact between the copper electrode and the active material. Upon releasing the force, the configuration generates a potential difference which is measured.

References

- Y. Zhu, H. Wang, H. Li and J. Zhu, *Journal of Wuhan University of Technology-Mater. Sci. Ed.*, 2016, **31**, 1219–1225.
- D. Sebastián, I. Suelves, R. Moliner and M. J. Lázaro, *Carbon N Y*, 2010, **48**, 4421–4431.
- Y. Huang, Y. Zheng, W. Song, Y. Ma, J. Wu and L. Fan, *Compos Part A Appl Sci Manuf*, 2011,
 42, 1398–1405.

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- **Fig. S3** Dispersion of ZnO nanoparticles in DMAc solvent (a) immediately after probe sonication without PVP (b) after 30 min probe sonication without PVP (c) after 24 hrs probe sonication with PVP.
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