SUPPORTING INFORMATION

Enhanced Thermal Conductivity for Poly(vinylidene fluoride) Composites with Nano-carbon Fillers

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SUPPLEMENT

THE ^{13}C-NMR and ^{1}H-NMR of SF was obtained from 400 MHz AVANCE III (Bruker, Switzerland), which was often used to denote the number of carbon atom and hydrogen atom and their position in compounds, as shown in **Figure S1. Figure S1** (a) shows a single weak peak at 80 ppm, indicating that only one kind of carbon exists in the SF molecular. In addition, **Figure S1** (b) demonstrates that there are two kind of protons.

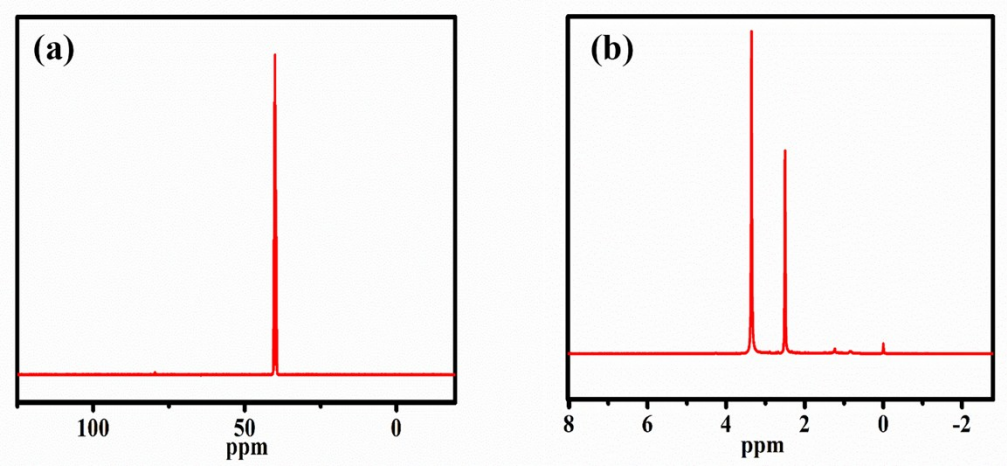

Figure S1. (a) ^{13}C and (b) ^{1}H NMR of SF.

The optical micrograph (OM) image of GS was captured by optical microscope (OM, Leica DM2500M, Germany) and shown in **Figure S2**. From **Figure S2**, we can see the typical morphology of GS deposited on Si substrate and find that GS tend to aggregate and the later size of them is ranging from a few to approximately 20 μm, which is agreement with the observation in SEM.

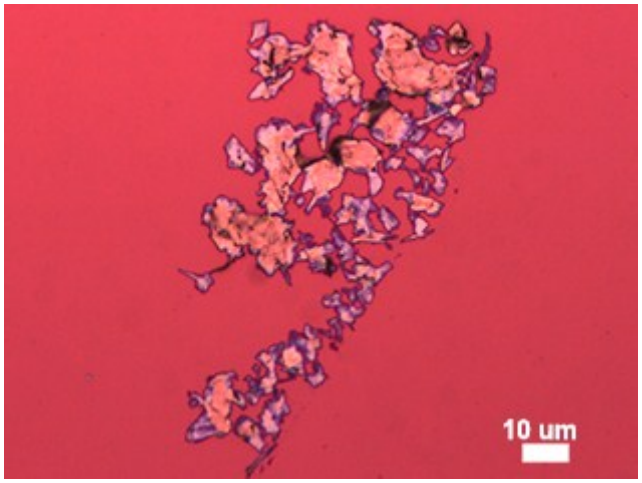

Figure S2. Optical micrograph (OM) image of GS.

The heating DSC thermograms of neat PVDF and PVDF composites was obtained by Pyris Diamond

DSC (Perkin-Elmer, American) and shown in **Figure S3. Figure S3** demonstrates that the melting temperature of neat PVDF, PVDF/SF, PVDF/CNT, and PVDF/GS, which corresponds to 162.81 °C, 163.17 °C, 167.99 °C and 166.49 °C, respectively.

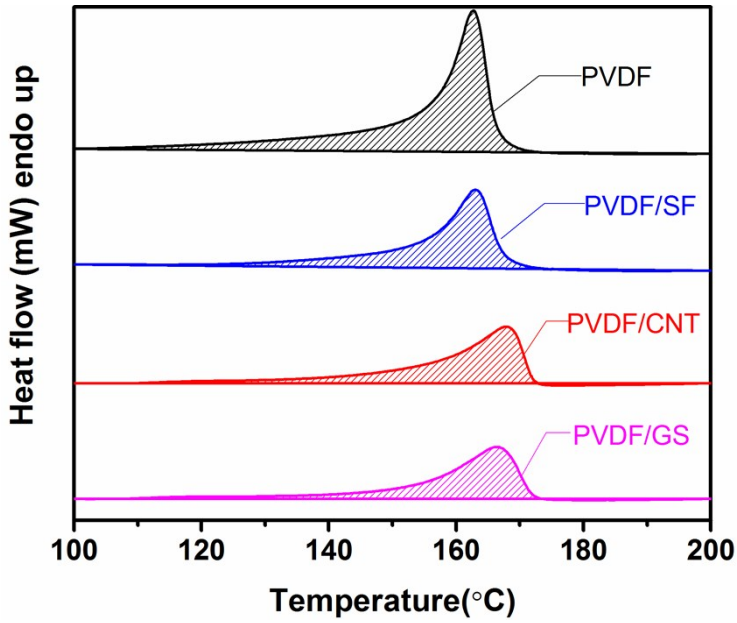

Figure S3. DSC thermograms of neat PVDF and PVDF composites.

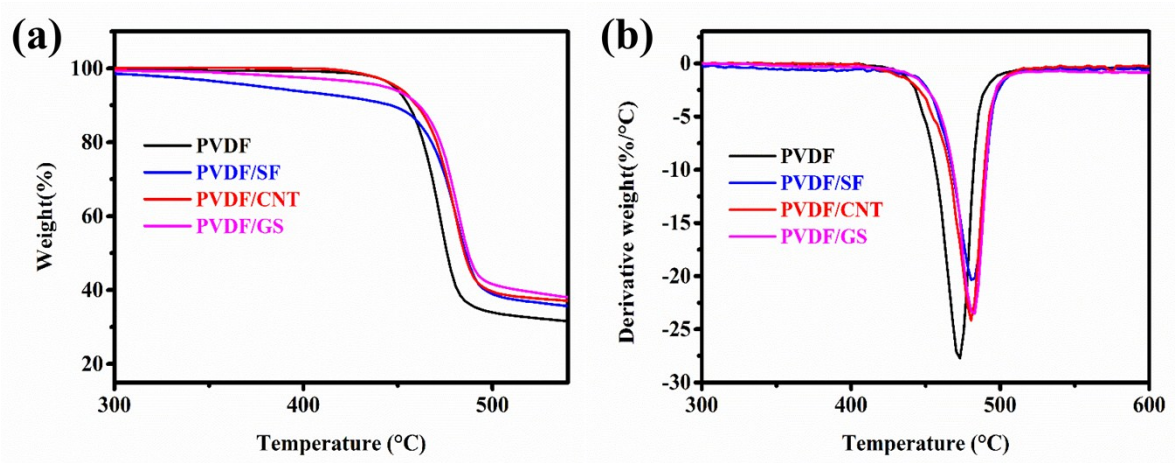

Figure S4. (a) TGA and (b) DTG curve of the neat PVDF and PVDF composites.

TGA and DTG curves of neat PVDF and PVDF composites were characterized by a TGA 209 F3 (NETZSCH, Germany) and shown in **Figure S4.** From **Figure S4 (a)**, one can see samples exhibit similar degradation profile, suggesting degradation mechanism of PVDF does not significantly change in the case of nano-carbon fillers. What's more, the addition of fillers result in shifting to higher temperatures for TGA curves, which indicates that nano-carbon fillers bring a good improvement on thermal stability of PVDF matrix. **Figure S4 (b)** show the DTG curves of neat

PVDF and PVDF composites. The peak temperatures of DTG curves reveal the temperature where the maximum weight loss rate attained. Obviously, PVDF composites with nano-carbon fillers possess a slight higher temperature as compared to that of neat PVDF.

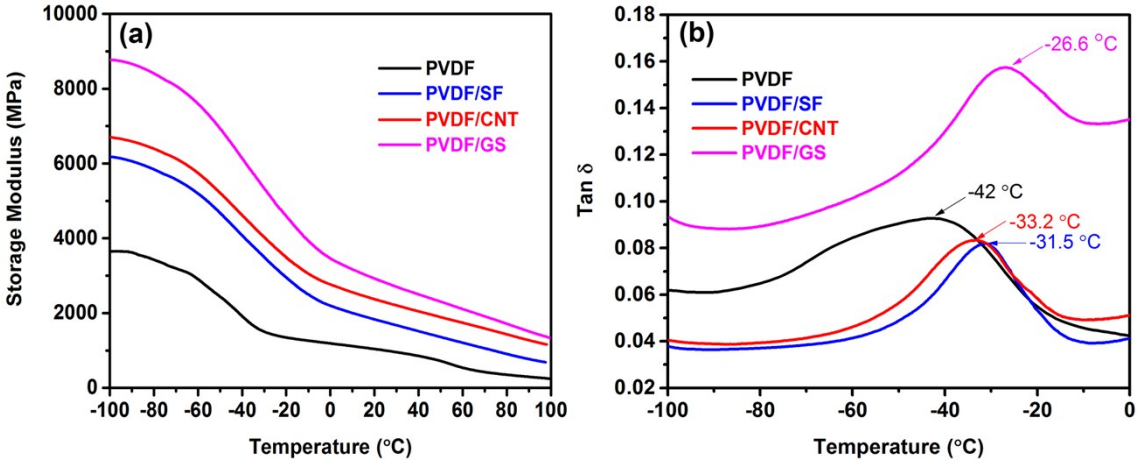

Figure S5. (a) Storage modulus and (b) loss factors of neat PVDF and PVDF composites as a function of test temperature.

Figure S5 exhibits the DMA analysis results of neat PVDF and PVDF composites as the function of the temperature. **Figure S5 (a)** obviously reveal that addition of nano-carbon materials significantly improve storage modulus of neat PVDF. The storage modulus of neat PVDF was proved to be 3.5 GPa at -80 °C and PVDF composites containing SF, CNT, and GS were 5.6, 6.2, 8.1 GPa, respectively, which correspond to 60%, 77.1%, 131% enhancement in comparison to neat PVDF. The reason why PVDF composites have high storage modulus may be because of strong interaction between nano-carbon materials and PVDF matrix, Moreover, it should be noted that storage modulus of both neat PVDF and PVDF composites exhibit high temperature dependences and gradually decrease with temperature increasing. **Figure S5 (b)** shows loss factors of neat PVDF and PVDF composites. The peak of loss factors determine glass transition temperature (Tg) of neat PVDF and PVDF composites. Neat PVDF represents a Tg at -42 °C. When SF, CNT, and GS are added into PVDF matrix, Tg increases to -31.5, -33.2, and -26.6 °C, respectively. The increase in Tg is believed to result from the restriction in molecular motion and higher degree of crosslinking demonstrating obvious changes in polymer chain dynamics[1, 2]. For PVDF composites, the addition of nano-carbon materials interfacial interactions between fillers and PVDF matrix and hinder molecular mobility, which finally leads to the increasement in Tg.

References

1. M. Fang, K. Wang, H. Lu, Y. Yang and S. Nutt, *Journal of Materials Chemistry*, 2009, **19**, 7098.
2. S. Ganguli, A. K. Roy and D. P. Anderson, *Carbon*, 2008, **46**, 806-817.