

## Article

# Interfacial Modulation of Graphene by Polythiophene with Controlled Molecular Weight to Enhance Thermal Conductivity

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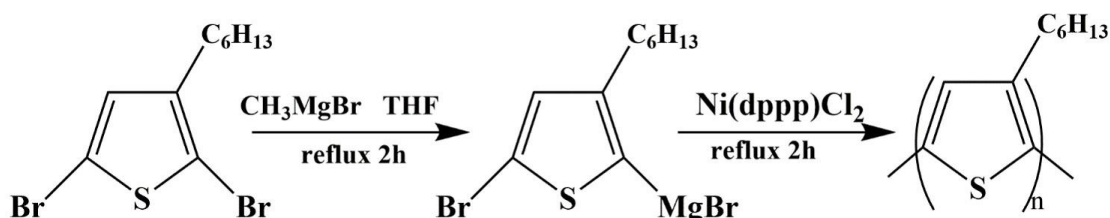


Figure S1. Synthetic routes of P3HT by GRIM method.

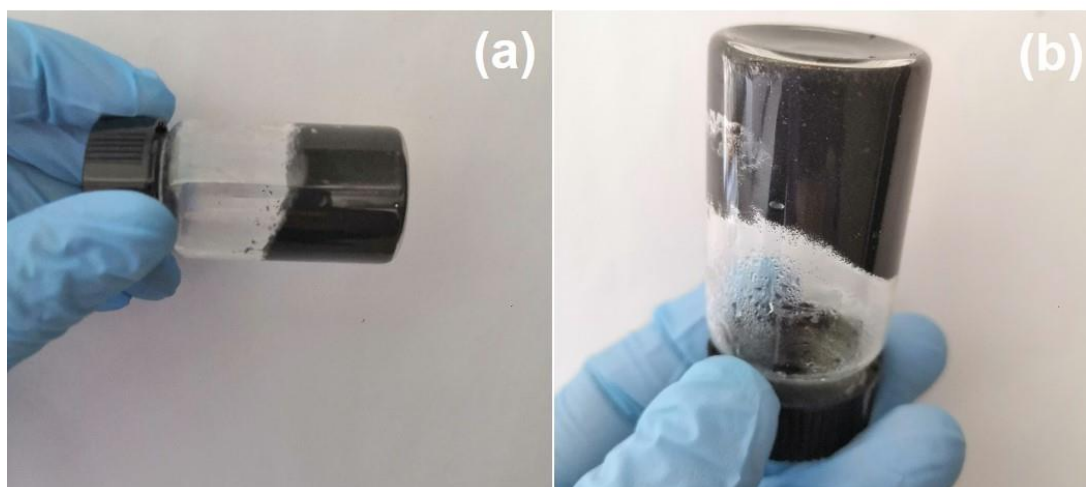


Figure S2. Optical images of 25 wt% GNS@P3HT/PVDF in (a) and (b).

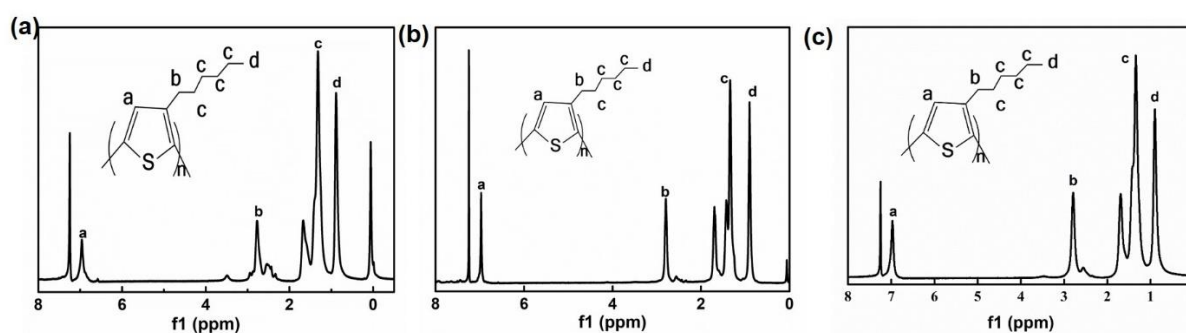


Figure S3.  $^1\text{H}$  NMR spectra of (a) P3HT(2000), (b) P3HT(10,000) and (c) P3HT(14,000).

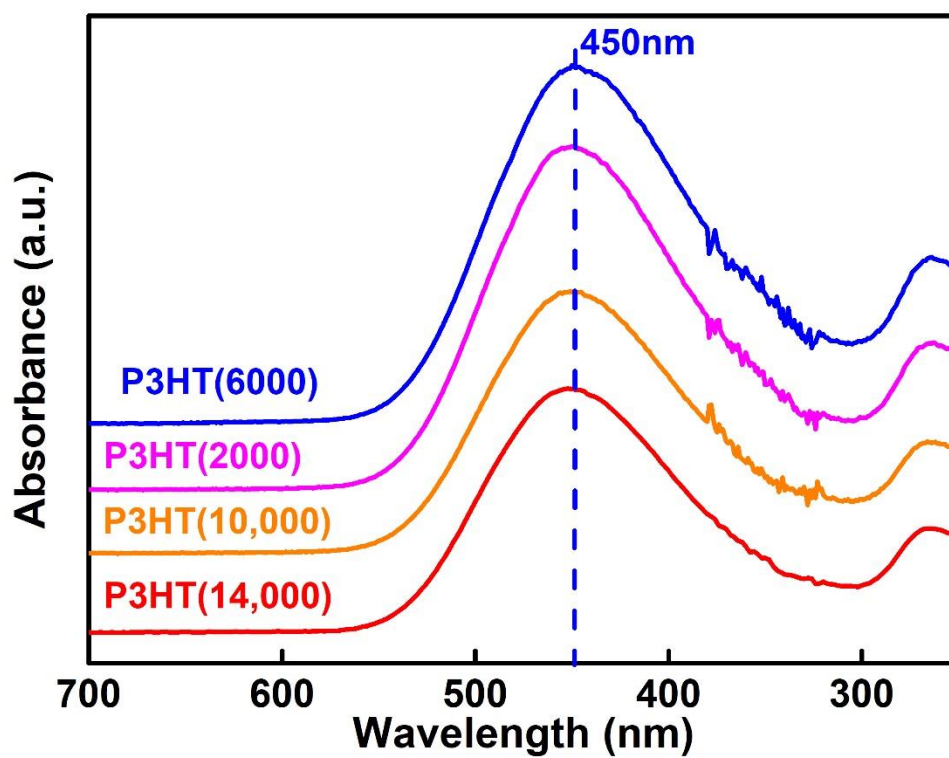


Figure S4. UV-vis spectra of P3HT at different molecular weights.

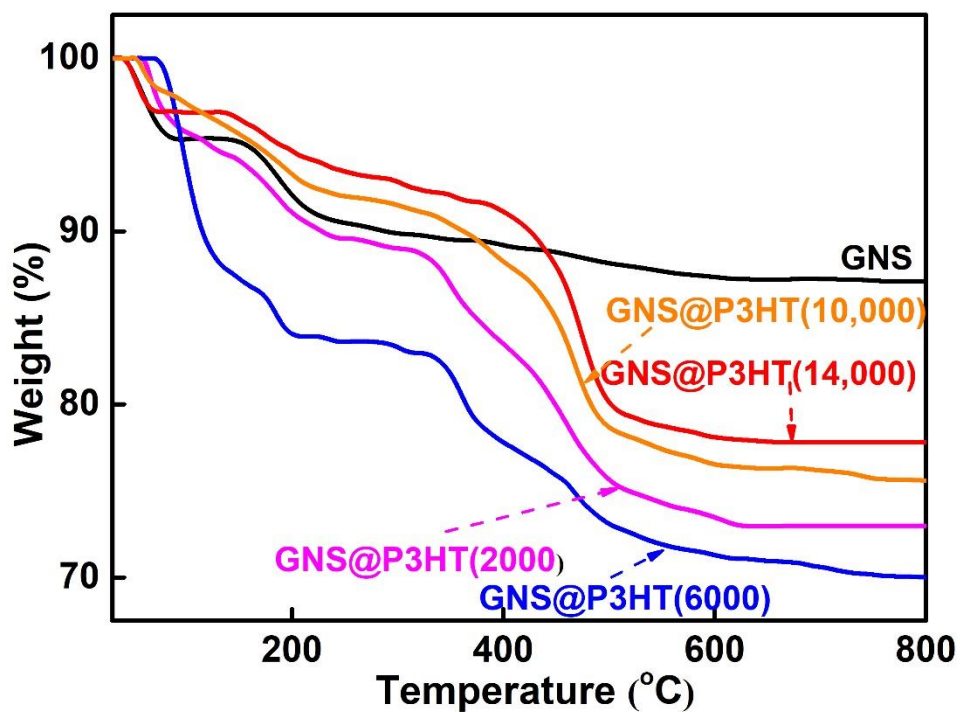
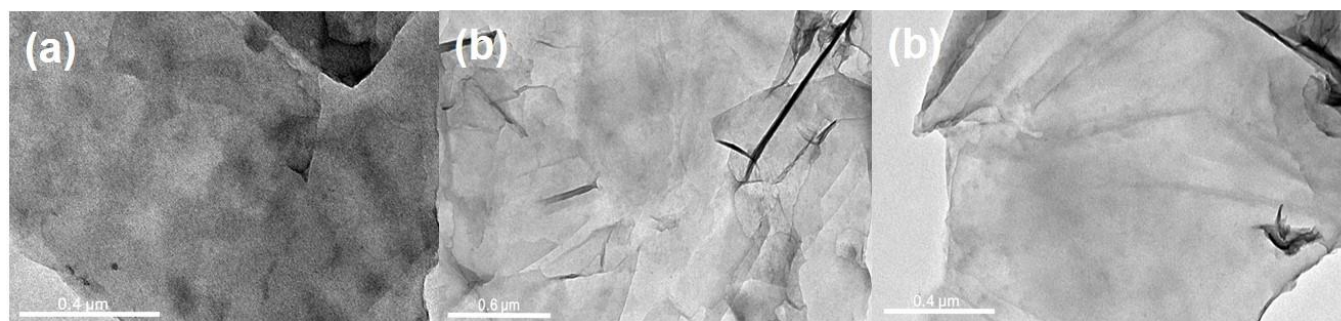
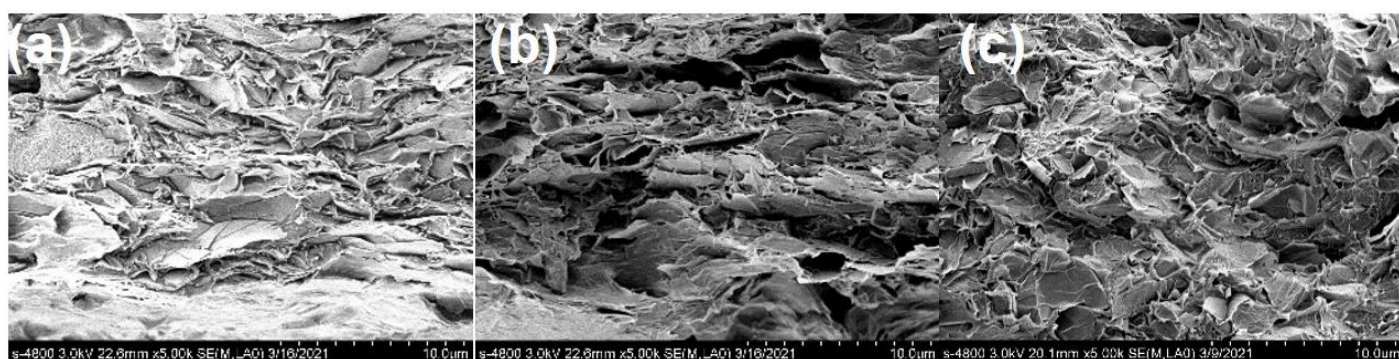


Figure S5. TGA curves of GNS@P3HT with different molecular weights.



**Figure S6.** TEM images of (a) GNS@P3HT(2000), (b) GNS@P3HT(10,000) and (c) GNS@P3HT(14,000).



**Figure S7.** SEM images of (a) 20 wt% GNS@P3HT(2000)/PVDF, (b) 20 wt% GNS@P3HT(10,000)/PVDF and (c) 20 wt% GNS@P3HT(14,000)/PVDF.

**Table S1.** Density of the GNS/PVDF and GNS@P3HT/PVDF membrane with different filler mass fractions.

| Mass fraction             | Mass (g) | Height (mm) | Density (g cm <sup>-3</sup> ) |
|---------------------------|----------|-------------|-------------------------------|
| PVDF                      | 0.022    | 0.083       | 1.64                          |
|                           | 0.024    | 0.086       | 1.75                          |
|                           | 0.021    | 0.085       | 1.52                          |
| 1wt%GNS/PVDF              | 0.020    | 0.078       | 1.61                          |
|                           | 0.021    | 0.071       | 1.84                          |
|                           | 0.023    | 0.079       | 1.77                          |
| 1wt%GNS@P3HT(2000)/PVDF   | 0.027    | 0.094       | 1.78                          |
|                           | 0.026    | 0.091       | 1.77                          |
|                           | 0.028    | 0.098       | 1.76                          |
| 1wt%GNS@P3HT(6000)/PVDF   | 0.023    | 0.084       | 1.72                          |
|                           | 0.025    | 0.089       | 1.74                          |
|                           | 0.024    | 0.086       | 1.75                          |
| 1wt%GNS@P3HT(10,000)/PVDF | 0.024    | 0.085       | 1.73                          |
|                           | 0.023    | 0.083       | 1.76                          |
|                           | 0.025    | 0.089       | 1.73                          |
| 1wt%GNS@P3HT(14,000)/PVDF | 0.023    | 0.081       | 1.74                          |
|                           | 0.024    | 0.084       | 1.75                          |
|                           | 0.025    | 0.087       | 1.75                          |
| 5wt%GNS/PVDF              | 0.026    | 0.089       | 1.78                          |
|                           | 0.027    | 0.095       | 1.77                          |
|                           | 0.026    | 0.092       | 1.74                          |
| 5wt%GNS@P3HT(2000)/PVDF   | 0.021    | 0.071       | 1.80                          |
|                           | 0.023    | 0.079       | 1.77                          |

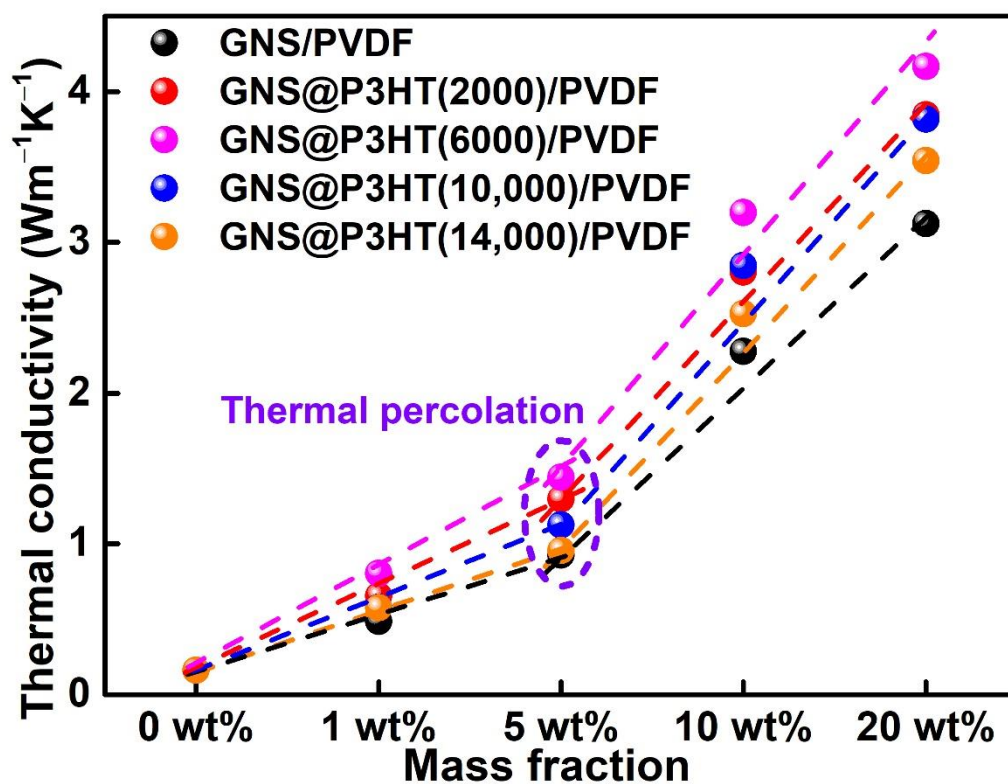
|                            |       |       |      |
|----------------------------|-------|-------|------|
|                            | 0.022 | 0.076 | 1.79 |
| 5wt%GNS@P3HT(6000)/PVDF    | 0.021 | 0.073 | 1.75 |
|                            | 0.021 | 0.075 | 1.73 |
|                            | 0.020 | 0.071 | 1.75 |
|                            | 0.024 | 0.085 | 1.75 |
| 5wt%GNS@P3HT(10,000)/PVDF  | 0.024 | 0.087 | 1.73 |
|                            | 0.023 | 0.082 | 1.77 |
|                            | 0.025 | 0.087 | 1.75 |
| 5wt%GNS@P3HT(14,000)/PVDF  | 0.024 | 0.085 | 1.75 |
|                            | 0.023 | 0.082 | 1.77 |
|                            | 0.018 | 0.063 | 1.81 |
| 10wt%GNS/PVDF              | 0.019 | 0.067 | 1.80 |
|                            | 0.020 | 0.068 | 1.81 |
|                            | 0.024 | 0.081 | 1.79 |
| 10wt%GNS@P3HT(2000)/PVDF   | 0.025 | 0.087 | 1.77 |
|                            | 0.024 | 0.085 | 1.76 |
|                            | 0.022 | 0.077 | 1.76 |
| 10wt%GNS@P3HT(6000)/PVDF   | 0.022 | 0.079 | 1.73 |
|                            | 0.023 | 0.081 | 1.76 |
|                            | 0.023 | 0.081 | 1.79 |
| 10wt%GNS@P3HT(10,000)/PVDF | 0.024 | 0.085 | 1.78 |
|                            | 0.025 | 0.089 | 1.77 |
|                            | 0.025 | 0.089 | 1.75 |
| 10wt%GNS@P3HT(14,000)/PVDF | 0.027 | 0.098 | 1.74 |
|                            | 0.026 | 0.092 | 1.73 |
|                            | 0.025 | 0.086 | 1.81 |
| 20wt%GNS/PVDF              | 0.022 | 0.078 | 1.73 |
|                            | 0.024 | 0.081 | 1.83 |
|                            | 0.022 | 0.073 | 1.85 |
| 20wt%GNS@P3HT(2000)/PVDF   | 0.022 | 0.075 | 1.83 |
|                            | 0.021 | 0.071 | 1.84 |
|                            | 0.025 | 0.085 | 1.80 |
| 20wt%GNS@P3HT(6000)/PVDF   | 0.024 | 0.081 | 1.81 |
|                            | 0.026 | 0.088 | 1.82 |
|                            | 0.021 | 0.071 | 1.85 |
| 20wt%GNS@P3HT(10,000)/PVDF | 0.023 | 0.079 | 1.83 |
|                            | 0.021 | 0.073 | 1.81 |
|                            | 0.023 | 0.081 | 1.79 |
| 20wt%GNS@P3HT(14,000)/PVDF | 0.026 | 0.089 | 1.77 |
|                            | 0.025 | 0.085 | 1.80 |

**Table S2.** Physical properties of the GNS/PVDF and GNS@P3HT/PVDF membrane with different filler mass fractions.

| Mass fraction | Density<br>(g cm <sup>-3</sup> ) | Specific heat<br>capacity<br>(J g <sup>-1</sup> K <sup>-1</sup> ) | Thermal<br>diffusivity<br>(mm <sup>2</sup> s <sup>-1</sup> ) | Thermal<br>conductivity<br>(W m <sup>-1</sup> K <sup>-1</sup> ) |
|---------------|----------------------------------|---|--|---|
| PVDF          | 1.645                            | 1.153   | 0.087  | 0.165   |
|               | 1.753                            | 1.144   | 0.081  | 0.162   |
|               | 1.514                            | 1.137   | 0.093  | 0.160   |
| 1wt%GNS/PVDF  | 1.609                            | 1.100   | 0.261  | 0.462   |

|                                |       |       |        |       |
|--------------------------------|-------|-------|--------|-------|
|                                | 1.844 | 1.062 | 0.256  | 0.501 |
|                                | 1.766 | 1.122 | 0.245  | 0.485 |
| 1wt%GNS@P3HT(2000)/PVDF        | 1.779 | 1.094 | 0.344  | 0.670 |
|                                | 1.766 | 1.125 | 0.330  | 0.656 |
|                                | 1.760 | 1.073 | 0.342  | 0.646 |
| 1wt%GNS@P3HT(6000)/PVDF        | 1.723 | 1.103 | 0.418  | 0.794 |
|                                | 1.737 | 1.108 | 0.428  | 0.824 |
|                                | 1.752 | 1.107 | 0.414  | 0.803 |
| 1wt%GNS@P3HT(10,000)/PVDF      | 1.733 | 1.044 | 0.328  | 0.593 |
|                                | 1.755 | 1.059 | 0.311  | 0.578 |
|                                | 1.726 | 1.034 | 0.319  | 0.569 |
| 1wt%GNS@P3HT(14,000)/PVDF      | 1.739 | 1.087 | 0.310  | 0.586 |
|                                | 1.753 | 1.111 | 0.291  | 0.567 |
|                                | 1.746 | 1.097 | 0.301  | 0.576 |
| 5wt%GNS/PVDF                   | 1.779 | 1.113 | 0.467  | 0.925 |
|                                | 1.766 | 0.950 | 0.528  | 0.886 |
|                                | 1.739 | 0.940 | 0.590  | 0.964 |
| 5wt%GNS@P3HT(2000)/PVDF        | 1.803 | 1.148 | 0.639  | 1.323 |
|                                | 1.770 | 1.111 | 0.6591 | 1.296 |
|                                | 1.787 | 1.096 | 0.6552 | 1.283 |
| 5wt%GNS@P3HT(6000)/PVDF        | 1.753 | 1.106 | 0.743  | 1.441 |
|                                | 1.727 | 1.094 | 0.751  | 1.419 |
|                                | 1.745 | 1.091 | 0.773  | 1.471 |
| 5wt%GNS@P3HT(10,000)/PVDF      | 1.754 | 1.022 | 0.628  | 1.126 |
|                                | 1.731 | 1.043 | 0.650  | 1.174 |
|                                | 1.769 | 0.919 | 0.644  | 1.047 |
| 5wt%GNS@P3HT(14,000)/PVDF      | 1.754 | 1.054 | 0.517  | 0.955 |
|                                | 1.747 | 1.030 | 0.535  | 0.962 |
|                                | 1.769 | 1.022 | 0.521  | 0.942 |
| 10wt%GNS/PVDF                  | 1.814 | 1.181 | 1.078  | 2.310 |
|                                | 1.796 | 1.126 | 1.127  | 2.279 |
|                                | 1.805 | 1.141 | 1.081  | 2.227 |
| 10wt%GNS@P3HT(2000)/PVDF       | 1.794 | 1.077 | 1.451  | 2.804 |
|                                | 1.772 | 1.026 | 1.431  | 2.603 |
|                                | 1.758 | 1.055 | 1.512  | 2.805 |
| 10wt%GNS@P3HT(6000)/PVDF       | 1.756 | 1.148 | 1.586  | 3.198 |
|                                | 1.732 | 1.139 | 1.643  | 3.241 |
|                                | 1.764 | 1.158 | 1.527  | 3.119 |
| 10wt%GNS@P3HT(10,000)/PVD<br>F | 1.794 | 1.041 | 1.536  | 2.869 |
|                                | 1.780 | 1.048 | 1.526  | 2.847 |
|                                | 1.765 | 1.043 | 1.532  | 2.821 |
| 10wt%GNS@P3HT(14,000)/PVD<br>F | 1.752 | 0.981 | 1.471  | 2.528 |
|                                | 1.739 | 0.968 | 1.487  | 2.504 |
|                                | 1.725 | 0.993 | 1.493  | 2.558 |
| 20wt%GNS/PVDF                  | 1.810 | 1.119 | 1.544  | 3.125 |
|                                | 1.733 | 1.116 | 1.633  | 3.158 |
|                                | 1.832 | 1.127 | 1.503  | 3.104 |
| 20wt%GNS@P3HT(2000)/PVDF       | 1.847 | 1.170 | 1.796  | 3.881 |
|                                | 1.829 | 1.157 | 1.823  | 3.857 |
|                                | 1.838 | 1.148 | 1.804  | 3.805 |
| 20wt%GNS@P3HT(6000)/PVDF       | 1.804 | 1.057 | 2.186  | 4.169 |

|                           |       |       |       |       |
|---------------------------|-------|-------|-------|-------|
|                           | 1.811 | 1.051 | 2.118 | 4.030 |
|                           | 1.825 | 1.073 | 2.13  | 4.171 |
| 20wt%GNS@P3HT(10,000)/PVD | 1.851 | 1.057 | 2.019 | 3.952 |
| F                         | 1.826 | 1.042 | 2.011 | 3.826 |
|                           | 1.809 | 1.023 | 1.983 | 3.670 |
| 20wt%GNS@P3HT(14,000)/PVD | 1.794 | 0.992 | 2.053 | 3.655 |
| F                         | 1.772 | 1.004 | 1.956 | 3.482 |
|                           | 1.802 | 1.017 | 1.965 | 3.599 |



**Figure S8.** Thermal conductivity of the PVDF membranes with GNS and GNS@P3HT fillers with different filler loading.