

## Supplementary information

### Structuring 3D-Printed Polypropylene Composites with Vertically Aligned Mesophase Pitch-based Carbon Fibers for Enhanced Through-Plane Thermal Conductivity and Mechanical Properties

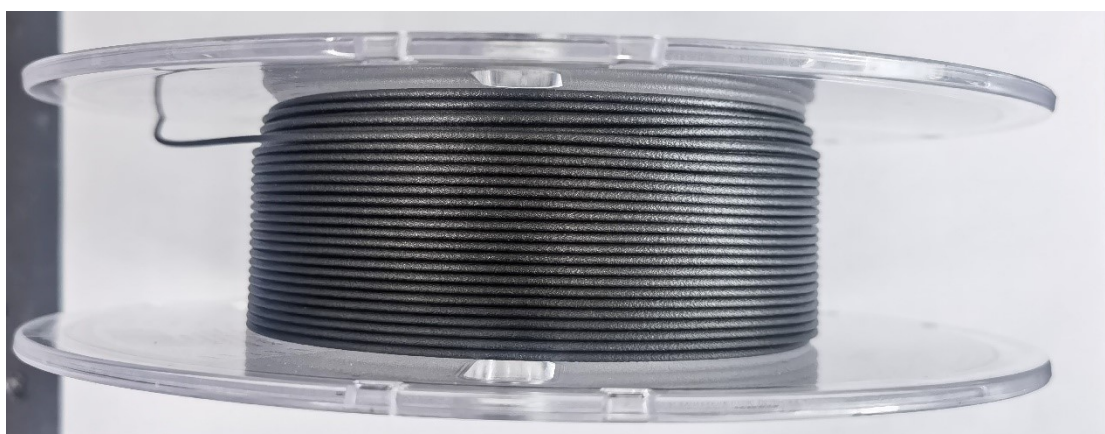
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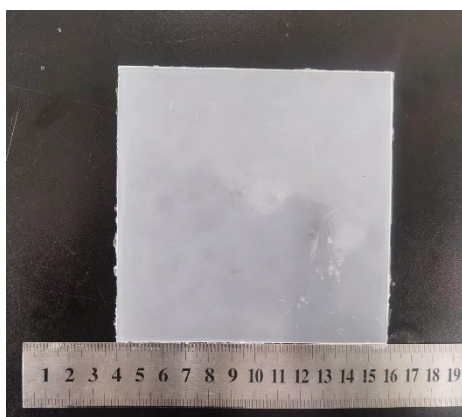
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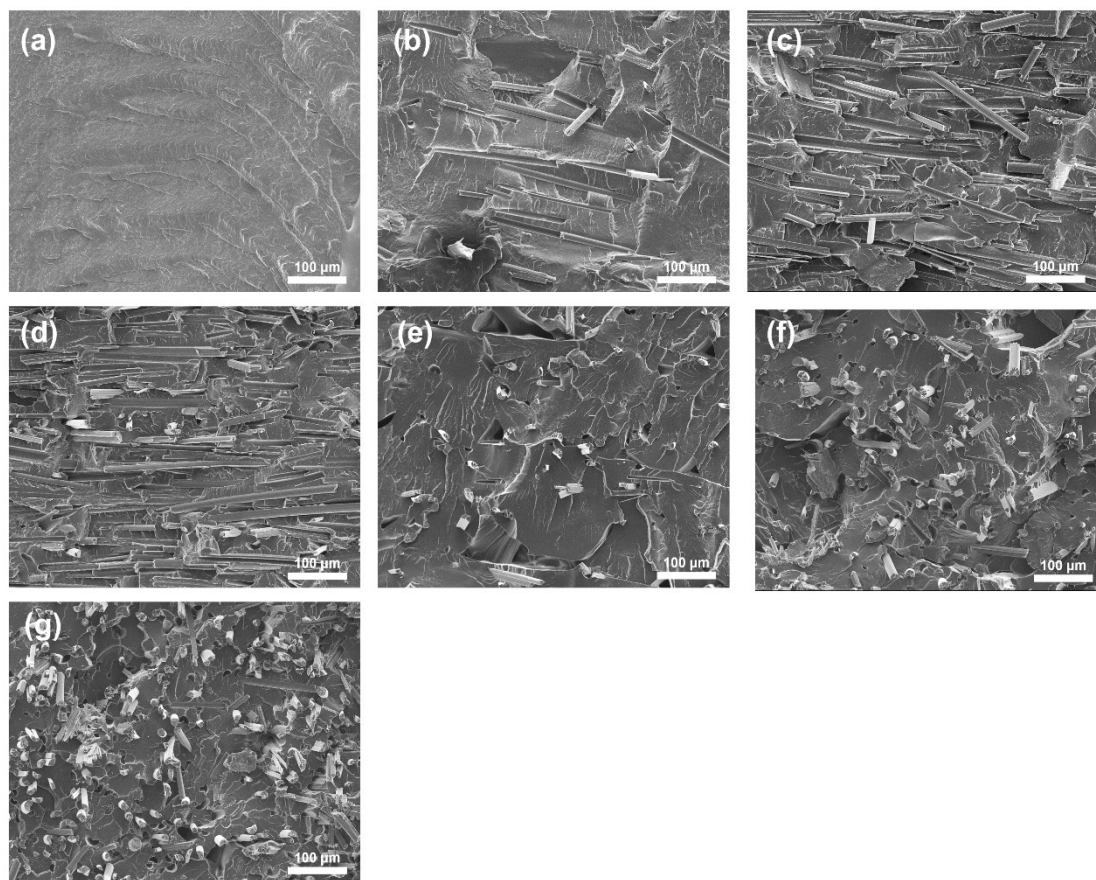
**Figure S1.** The CF/PP filaments with a uniform diameter of 1.75mm.



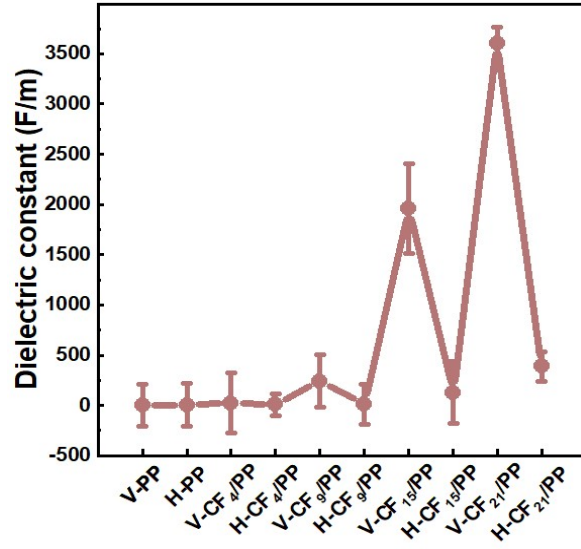
**Figure S2.** A  $120 \times 120 \times 1.5$  mm PP substrate.



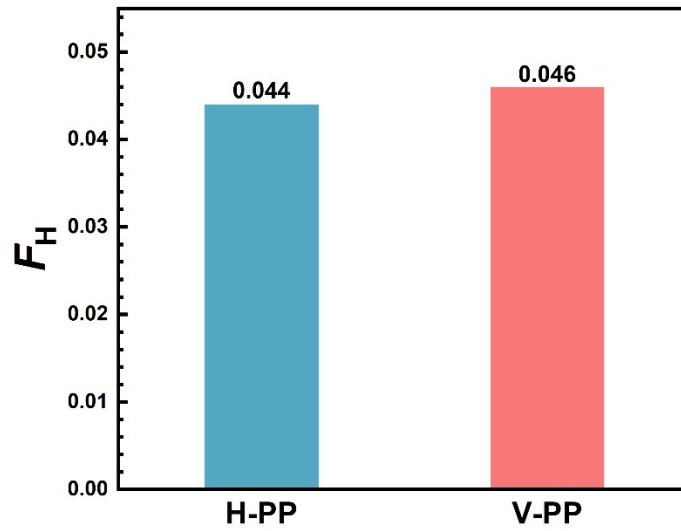
**Figure S3.** DR-III thermal conductivity instrument with heat-flow method.



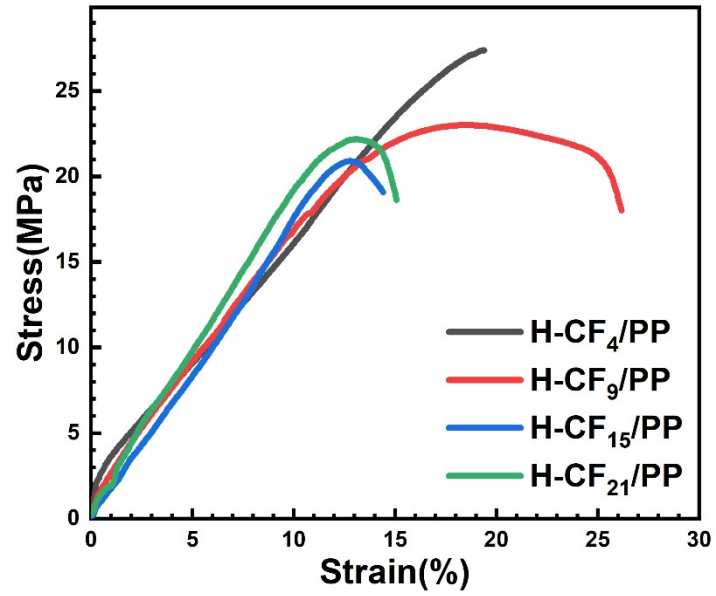
**Figure S4.** (a) Cross-sectional morphology of PP. (b-d) Cross-sectional morphology of H-CF/PP with CF contents of 4, 9, and 15 vol%, respectively. (e-g) Cross-sectional morphology of V-CF/PP with CF contents of 4, 9, and 15 vol%, respectively.



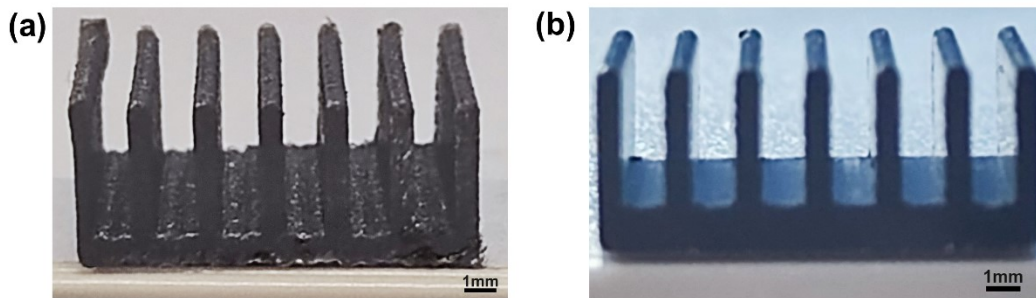
**Figure S5.** The dielectric constant of 3D printed CF/PP composites.



**Figure S6.** The  $F_H$  of CF/PP composites from 2D WAXD patterns.

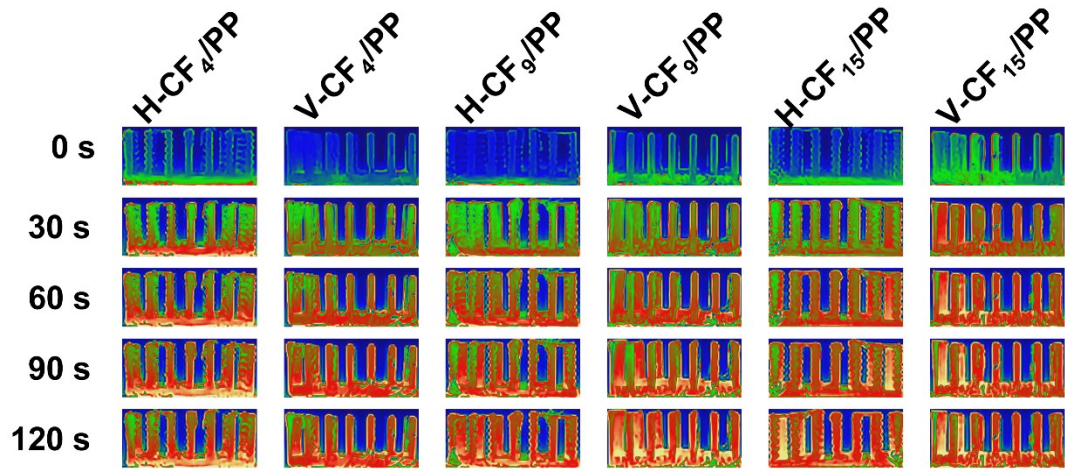


**Figure S7.** Stress-strain curves of H-CF/PP composites with varying CF loading.



**Figure S8.** Digital photos of different printing direction of CF/PP composites: (a) V-CF<sub>21</sub>/PP and (b) commercialized product.





**Figure S9.** Infrared thermography of 3D-CF/PP composite material with varying CF loading.

**Table S1** FDM parameters used for the 3D printing of CF/PP scaffold.

Parameters	Value
Print nozzle diameter (mm)	0.4
Layer height (mm)	0.4
Nozzle temperature (°C)	200
Printing bed temperature (°C)	70
Feed rate (mm/s)	30
Infill pattern	Linear infill pattern
Print infill (%)	100

**Table S2** Values of melting temperatures, melting enthalpies and degree of crystallinity temperatures for CF/PP composites obtained by DSC analysis.

Sample	Melting temperature (°C)	Enthalpy (J/g)	Degree of crystallinity (%)
PP	149.09	61.62	29.77
CF <sub>4</sub> /PP	148.74	52.25	25.24
CF <sub>9</sub> /PP	147.16	46.04	22.24
CF <sub>15</sub> /PP	147.43	44.46	21.48
CF <sub>21</sub> /PP	151.19	39.96	19.30

**Table S3** Values of initial decomposition temperature, peak temperature and residual CF rate for CF/PP composites obtained by thermogravimetric analysis.

Sample	Initial decomposition temperature (°C)	Peak temperature (°C)	Residual CF rate (%)
PP	392.02	432.50	1.36
CF <sub>4</sub> /PP	421.94	450.00	7.78
CF <sub>9</sub> /PP	431.45	455.83	26.10
CF <sub>15</sub> /PP	437.16	461.67	31.20
CF <sub>21</sub> /PP	437.58	461.68	39.80