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Supporting Information: Flexible and High Thermal Conductivity Composites based on Graphite Nanoplates Paper Impregnated with Polydimethylsiloxane

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In a preliminary phase of this work, impregnation tests were carried out with different PDMS concentration. FESEM images of cross sections and surface morphologies of the 9% 1x (a), 17% 1x (b) and 29% 1x (c) samples are reported in (Figure S1). As expected, upon the evaporation of hexane, a highly porous structure is retained, as clearly evidenced by the presence of many air pockets sizing between a few μm to a few tens of μm.

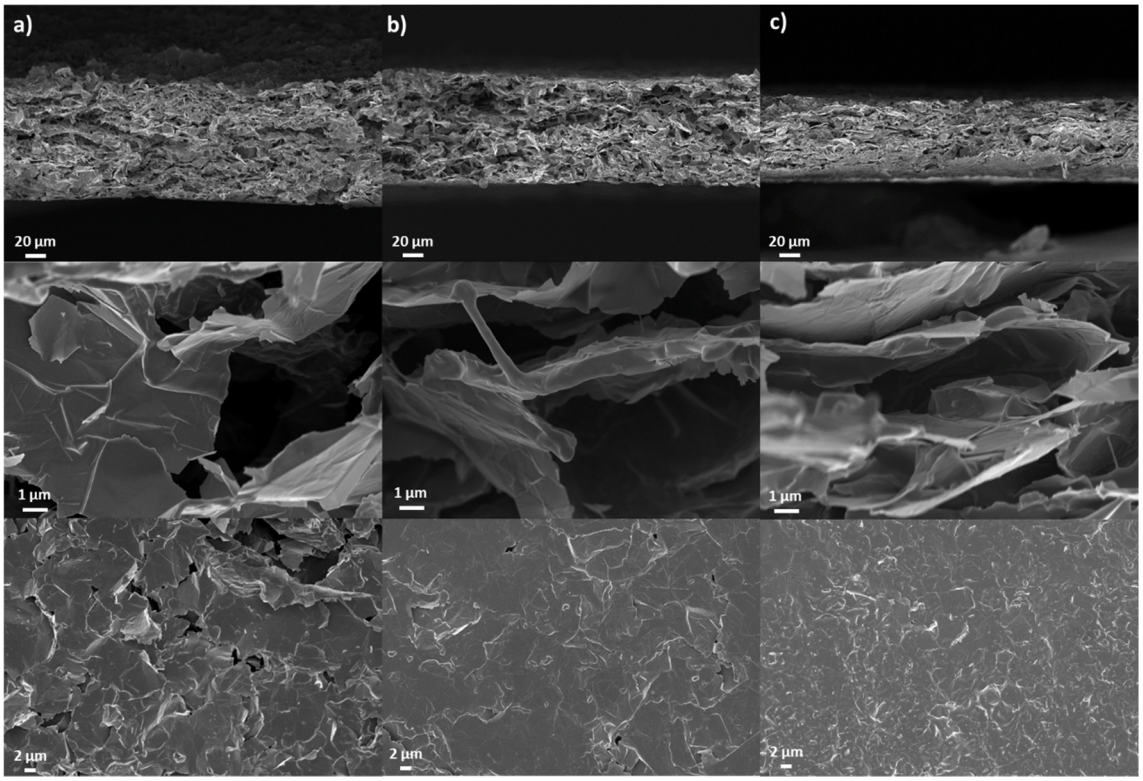

Figure S1. FESEM micrographs of GNP nanopaper impregnated with 9% 1x (**a**) 17% 1x (**b**) and 29% 1x (**c**) solution.

Aiming at the nanopaper porosity reduction, multiple impregnations of the various solutions at different concentrations are investigated (coded as 2x and 3x). Density (ρ) of the nanopapers was found to increase with the number of impregnations and the porosity is progressively reduced (Table S1). By this approach, low porosity values are obtained, by filling the voids with the polymer, and therefore lowering the relative percentage of GNP in the composite. Therefore, progressive densification of the nanopaper leads to a lower GNP fraction. For instance, a density of approx. 1 mg/mm^3 was obtained for 29% 3x nanopaper, corresponding to 10 wt.% GNP content. Furthermore, it is worth noting that

similar wt.% contents of GNP and vol.% porosity can be achieved with different impregnation conditions (e.g., 17% 2x vs. 29% 1x).

Table S1. Density, porosity and thermal diffusivity values calculated for impregnated nanopapers.

Sample Code	ρ (g/cm³)	Wt.% GNP	Vol.% Porosity	Thermal Diffusivity α (mm²/s)	
				Cross-Plane	In-Plane
9% 1x	0.27 ± 0.05	47.8 ± 1.0	83 ± 4	1.44 ± 0.05	51.3 ± 2.1
9% 3x	0.79 ± 0.05	25.9 ± 0.1	39 ± 4	1.24 ± 0.05	24.2 ± 0.4
17% 1x	0.48 ± 0.05	28.7 ± 0.1	64 ± 4	0.93 ± 0.05	30.8 ± 0.4
17% 2x	0.60 ± 0.05	24.7 ± 0.1	54 ± 4	1.26 ± 0.03	14.6 ± 0.8
29% 1x	0.67 ± 0.05	22.5 ± 0.1	47 ± 4	1.33 ± 0.01	12.7 ± 0.2
29% 2x	0.85 ± 0.05	16.4 ± 0.1	28 ± 4	0.75 ± 0.01	12.2 ± 0.4
29% 3x	1.03 ± 0.05	10.3 ± 0.1	7 ± 4	1.00 ± 0.01	8.6 ± 0.4

Thermal diffusivity of the different impregnated nanopapers were evaluated by light flash analyses, in both in-plane and cross-plane modes (Table S1), for correlation with nanopaper density and GNP content (Figure S2).

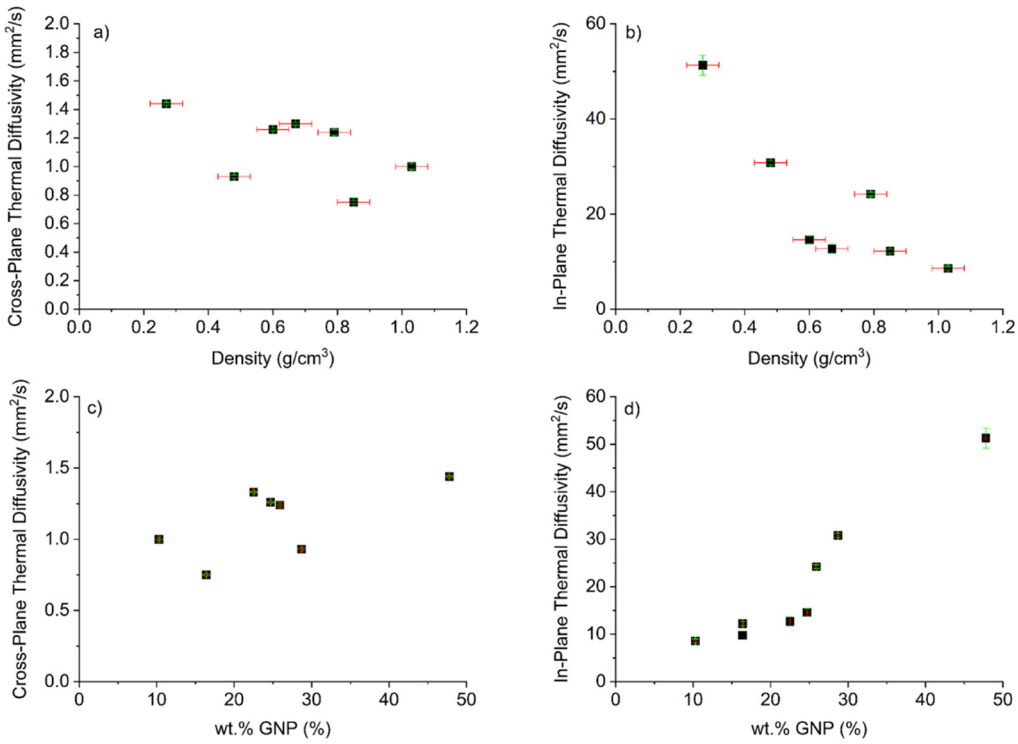

Figure S2. Thermal diffusivity of PDMS impregnated nanopapers, as a function of nanopaper density (**a**,**b**) and as a function of GNP content (**c**,**d**).

PDMS/GNP nanopapers are expectedly showing lower diffusivities than pristine GNP nanopaper (cross plane 9.2 ± 0.1 mm²/s, in plane 101 ± 5 mm²/s). By analysing the diffusivity data as a function of density Figure S2 a,b, it is possible to verify a decreasing trend for both the cross-plane and in-plane diffusivity, with increasing density. This fact is explained by the increased quantity of PDMS between the GNP lamellae. Being PDMS a poorly conductive material, the increase in PDMS concentration is reflected in a decrease in the diffusivity in the composite, which is more evident in the in-plane orientation. Conversely, thermal diffusivity show an expected increase as a function of GNP content (Figure S2 c,d), which is particularly remarkable for in-plane measurements.

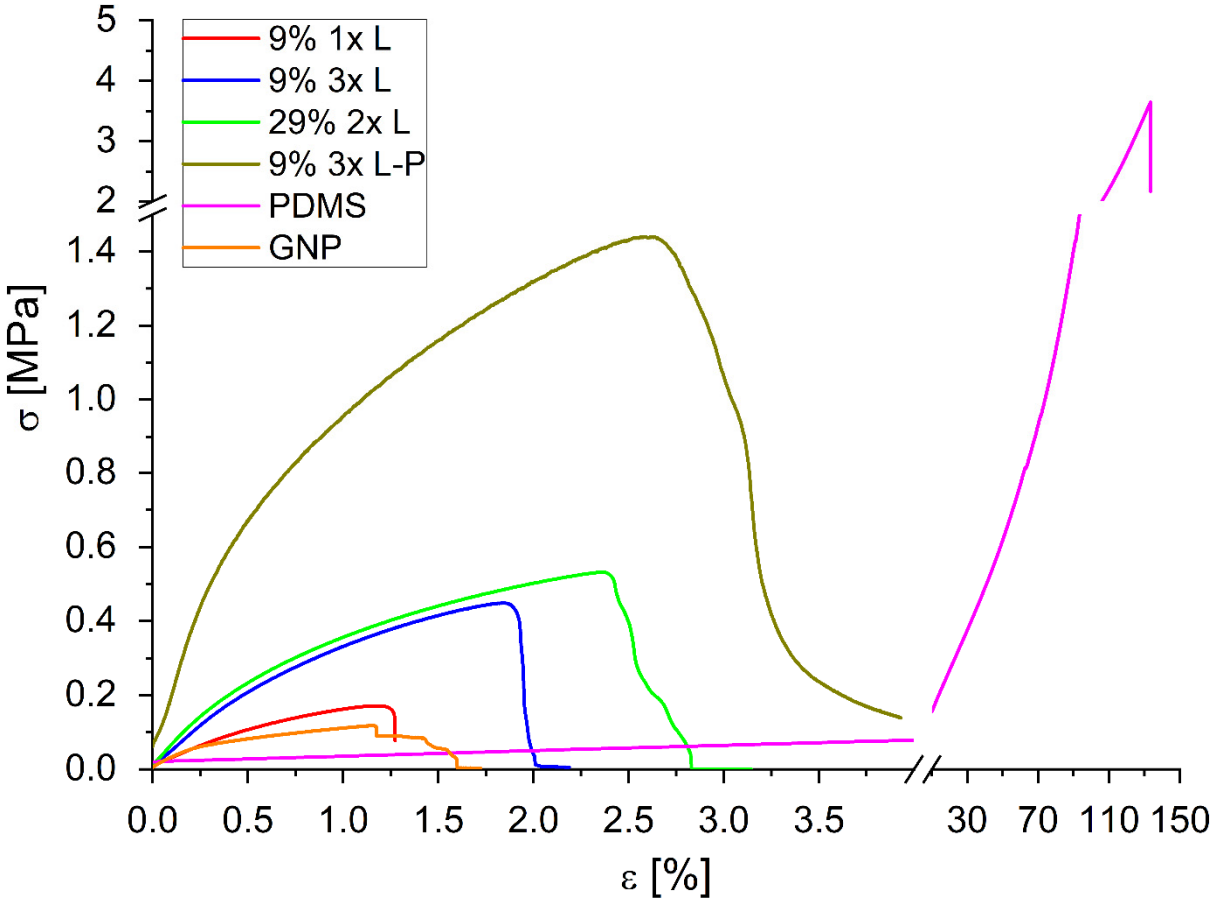

Figure S3. Representative stress-strain curves from tensile tests.