Supporting Information

High Density 3D-Boron Nitride & 3D-Graphene for High Performance Nano-Thermal Interface Material

Manuela Loeblein,[†,‡,$] Siu Hon Tsang,[||] Matthieu Pawlik,[‡] Eric Jian Rong Phua,[||,#] Han Yong,[$] Xiao Wu Zhang,[$] Chee Lip Gan[||,#] and Edwin Hang Tong Teo[†,#,]*

[†]School of Electrical and Electronic Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore

[‡]CNRS-International NTU Thales Research Alliance (CINTRA) UMI 3288, Research Techno Plaza, 50 Nanyang Drive, Singapore, Singapore 637553

[||] Temasek Laboratories@NTU, 50 Nanyang Avenue, Singapore 639798, Singapore

[#] School of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore

[$]Institute of Microelectronics, Agency for Science, Technology and Research (A*Star), 11 Science Park Road, Singapore 117685, Singapore

[*]Corresponding Author Email: HTTEO@ntu.edu.sg

KEYWORDS: three-dimensional graphene, three-dimensional h-BN, nano-thermal interface material, hot-spot removal, thermal management

Applicability of the laser flash technique for the case of porous 3D-foams:

Due to the porous structure of the 3D-foams, prior to laser flash measurements it must be assessed whether the structure can still comply to the requirement of "thickness of sample >> optical penetration depth of laser".

In order to verify that the optical penetration depth is much smaller than the thickness of the sample, direct measurements of the transmission of the sample were conducted. Since the LINSEIS XFA 500 laser flash uses a Xenon lamp as shot source, which covers almost the complete wavelength range from visible to invisible, in the following, the complete range from 200 nm to ca. 1000 nm is considered.

Transmission measurement of an uncompressed 3D-foam of same thickness and ppi as used for the laser flash measurement was performed via UV-vis spectroscopy (Shiimadzu UV-2450 Spectrophotometer). The measurement was repeated on several different spots, all obtained curves displayed the same behavior. A representative curve is shown in **Figure S1**.

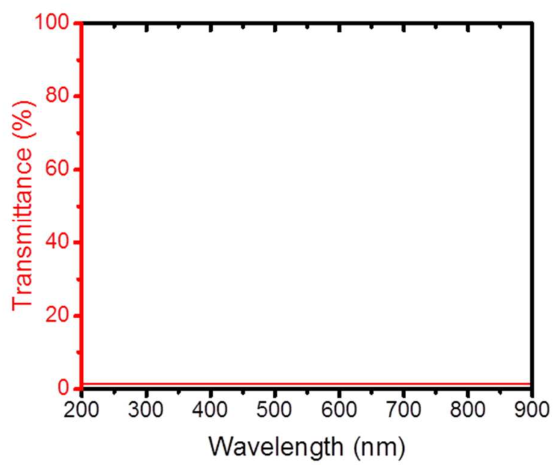


Figure S1. UV-vis spectroscopy results for transmittance of 3D-foam

It can be seen that the 3D-foam has a negligible transmittance of 1.3%, thus there is no through-hole light transmission. This makes it absolutely clear that no light passes through the samples, i.e. the penetration depth remains much lower than the sample thickness.

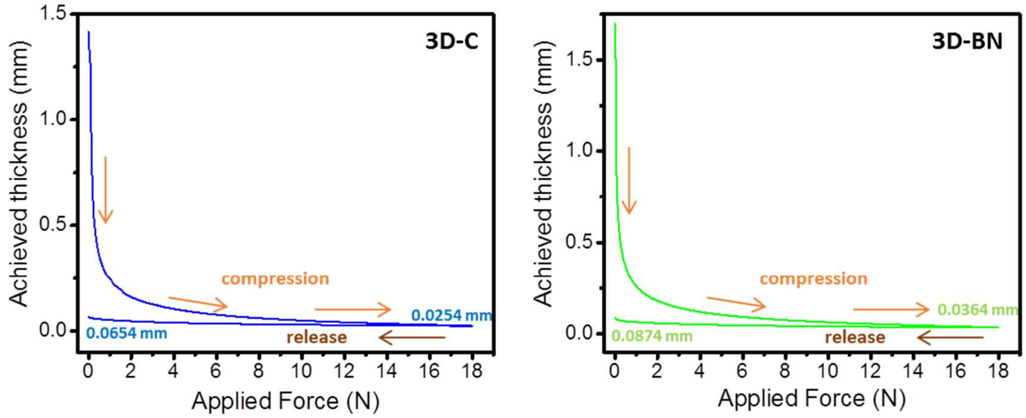


Figure S2. Study of the relationship applied force – achieved compression. **(a)** 3D-C; **(b)** 3D-BN.

The measurement was performed by placing uncompressed 1.67 mm thick samples centralized on the parallel plate compression clamps of a dynamic mechanical analyzer (DMA Q800) with a resolution of 1 nm. Used parameters were a force ramp from 0.001 N to 18 N at 0.1 N min^{-1} for compression, followed by a force ramp from 18 N to 0.001 N at 0.25 N min^{-1} for release.

Due to the system's limitations, the force could only be ramped up to 18 N, which did not achieve the full compression of the 3D-foams down to ~6.6 µm.

18 N yields a compression down to 25.4 µm for 3D-C and down to 36.4 µm for 3D-BN. From the graph it can be extrapolated that in order to reach 6.6 µm, a force of ~22.8 N is required for 3D-C and of ~24.6 N for 3D-BN. This could be achieved by compression with a weight of 2.3 – 2.5 kg without applying any additional external forces.

After release, a slight decompression in the range of 40 – 50 µm is visible, which for application as heat spreader would not be the case since the weight of the heat sink will not be removed.

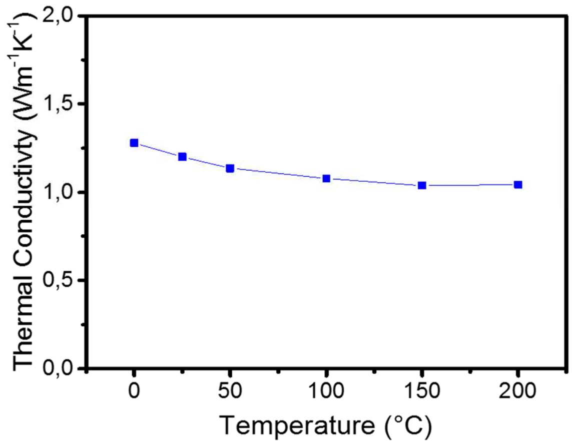
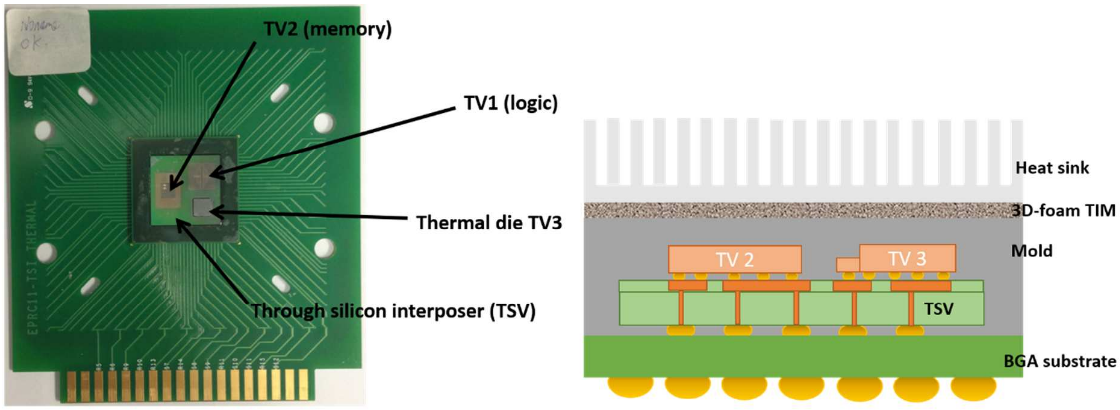


Figure S3. Thermal stability of 3D-C throughout a temperature range from 0°C to 200°C, measured through laser flash.



Photograph of the test chip without overmold

Schematic of test structure

Figure S4. Test device used to assess the material's performance under real conditions. **(a)** Photograph of the chip without any overmold; **(b)** schematic of the cross-sectional view of the test setup. TV1 stands for a dummy logic unit, TV2, dummy memory unit and TV3 for the thermal die array (source of the hot spot).

Table S1. Calculation of the thickness for the study of the evolution of the interface thermal resistance of 3D-C

Initial thickness of the 3D-foam t_{init} (mm)	Minimum thickness t_{min} (µm) (cf thermal boundary section for the calculation method)	t_e for an example roughness of 4 µm ($t_e = t_{min} + t_r$)
0.5	2	6
1	4	8
2	8	12
5	21	25
10	43	48

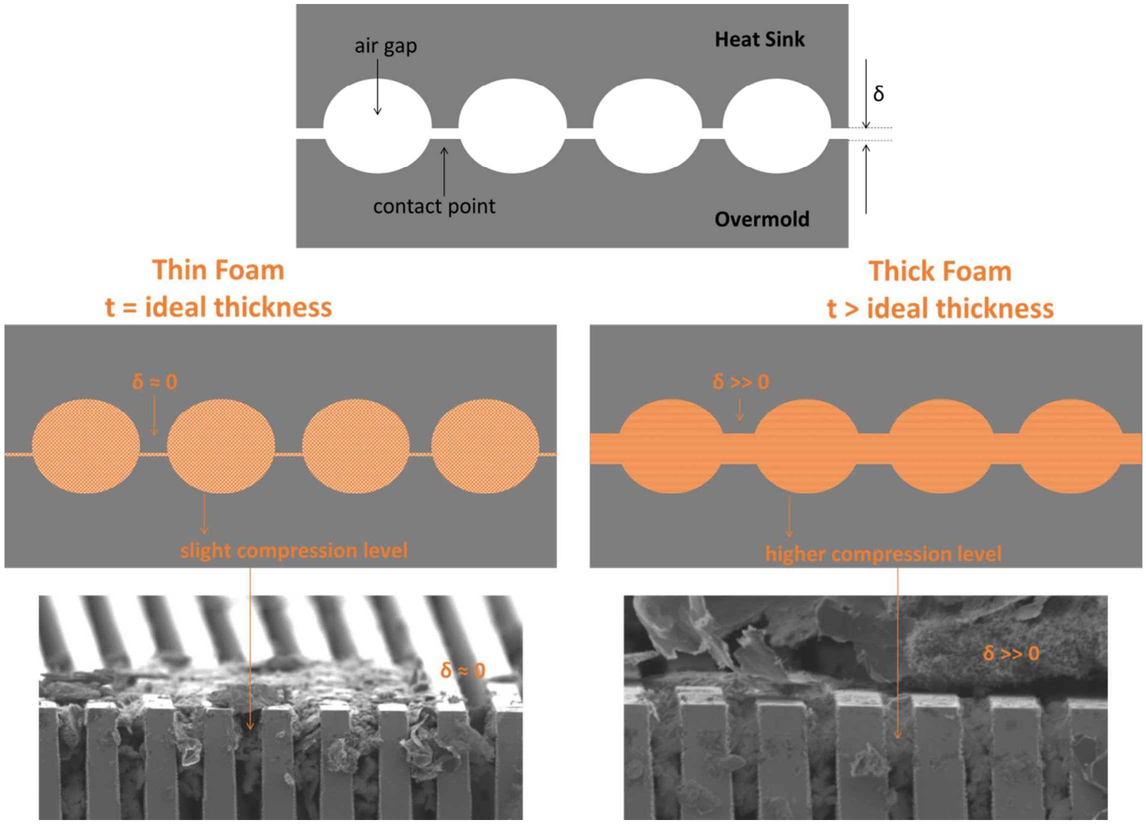


Figure S5. Schematic of the gap adaptability of the 3D-foams to illustrate the effect of using 3D-foams with ideal thickness and larger thickness.