

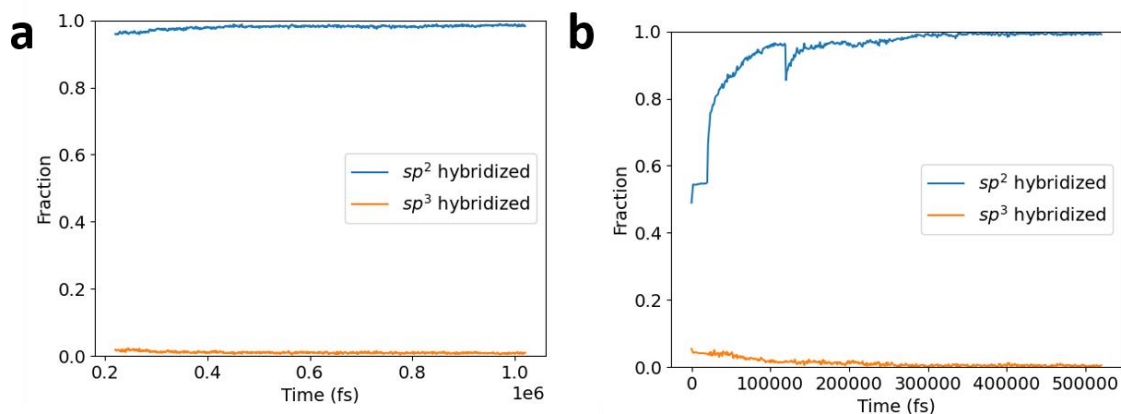
## Supporting Information

for *Macromol. Mater. Eng.*, DOI 10.1002/mame.202300230

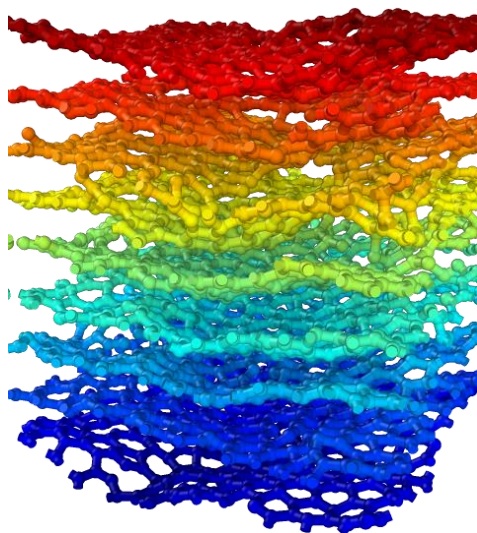
Graphitizability of Polymer Thin Films: An In Situ TEM Study of Thickness Effects on Nanocrystalline Graphene/Glassy Carbon Formation

*C. N. Shyam Kumar\**, *Clemens Possel*, *Simone Dehm*, *Venkata Sai Kiran Chakravadhanula*, *Di Wang*, *Wolfgang Wenzel*, *Ralph Krupke* and *Christian Kübel\**

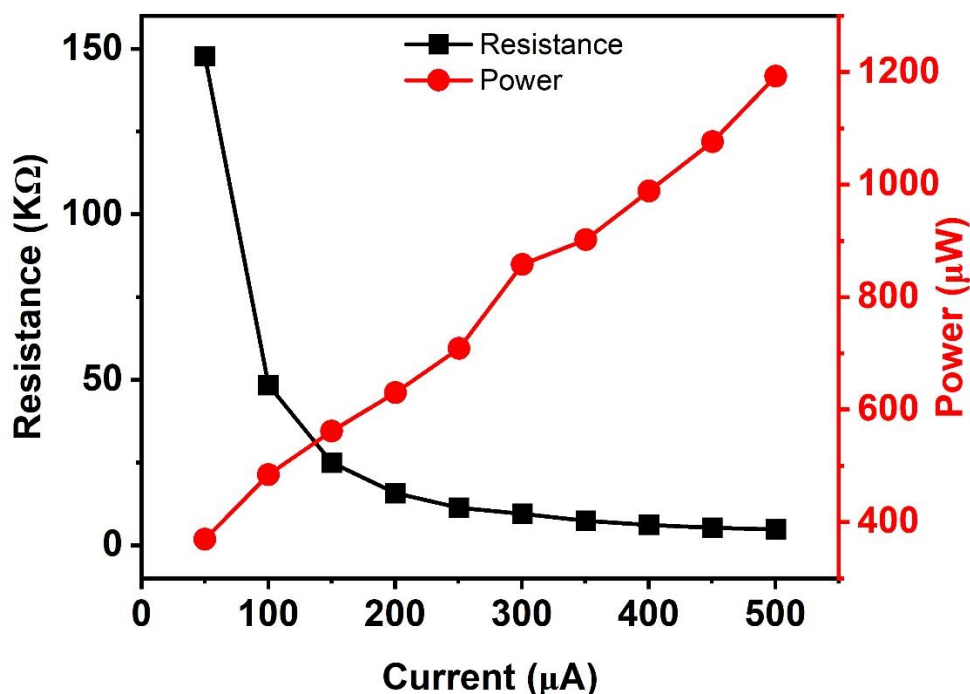
## Supplementary



Supplementary Figure 1: Fraction of  $sp^2$  and  $sp^3$  hybridized atoms. While  $sp^2$  hybridized bonds indicate graphene-like structures,  $sp^3$  hybridized bonds indicate diamond-like structures. Here the hybridization fraction for the thin layer simulation from Figure 8c/d is shown (a) as well as for the thin layer simulation from Figure 8 (b). Regardless of the initial structure, almost all atoms form  $sp^2$  hybridized bonds, i.e. graphene-like structures allowing to neglect those atoms with less/more than 3 bonded neighbours for analysis. This observation is in accordance with the expected structure from carbon phase diagrams for the observed temperature and pressure and thus, also confirms the quality of the force field.



Supplementary Figure 2: Final structure of annealing simulation of initially amorphous 2D periodic thin layer carbon structure with thickness of 35 Å. The 9 carbon graphene layers, that formed during annealing, are nicely aligned parallel to the surface. For better visualization the carbon atoms are colored depending on their z position (z axis perpendicular to graphene layers).



Supplementary Figure 3: Change in resistance and dissipated power during current annealing of the sample in Figure 3. The value will be a combination of contact resistance and film resistance, but reflects the increase in conductivity of the film with increasing carbonisation.

Supplementary table -1

Composition of S1805

Component	Concentration
Electronic grade propylene glycol monomethyl ether acetate	70.0 - 80.0 %
Mixed cresol novolak resin	10.0 - 25.0 %
Diazo Photoactive Compound	1.0 - 10.0 %
Cresol	< 1.0 %
Nonionic surfactant	< 1.0 %
Methoxy-1-propanol acetate	< 1.0 %

Supplementary video 1: Current annealing video

Supplementary video 2: Formation of cage-like structures in the thick samples.

Supplementary video 3: Formation of cage-like structure from bonded bi layers.

Supplementary video 4: Formation of CNT like structures from bonded bilayers.