Electronic Supplementary Material (ESI) for Nanoscale.

Supporting information

Electron dose during experiments

We were using low-dose conditions, keeping the illumination at an absolute minimum, taking a few selected images every 100 °C during the initial heating and blanking the beam in between. During reheating the dose was higher, with images acquired every 10 °C and blanking the beam in between. The dose for each single image varied between 2.3×10^6 e/nm^2 at 300 Kx and 4.8×10^4 e/nm^2 at 56 Kx. During the initial heating, we acquired on average 7 images at different magnifications for each temperature and with this the maximum final dose was less than 10^8 e/nm^2 during initial heating. During re heating, all images were acquired at 300 Kx and the final total dose during reheating was around 2.2×10^8 e/nm^2. High resolution images were acquired only after heating to 1200 °C and the dose was calculated to be 2.1×10^7 e/nm^2 for each image.

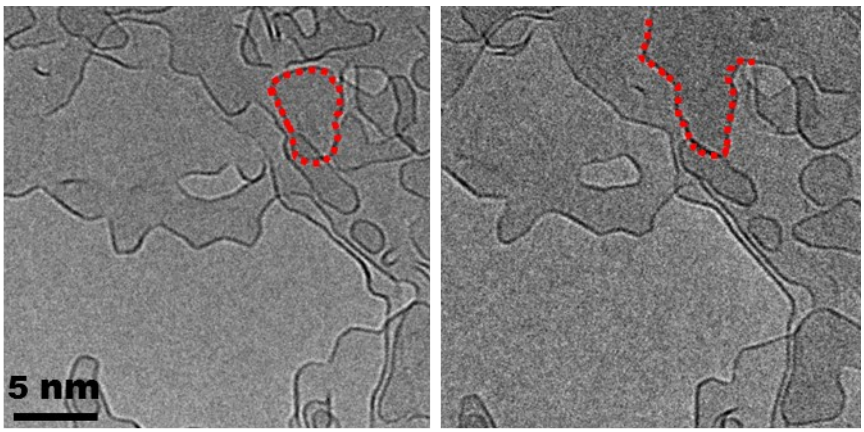

Figure S1: Merging of smaller domains to form larger ones during heating from 1100 °C to 1200 °C.

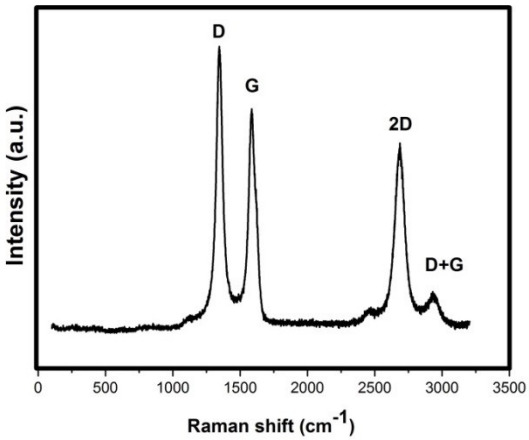

Figure S2: Raman Spectrum of graphitized sample at 1200°C

The spectrum consists of sharp D and G peak. The I_D/I_G ratio is 1.25 and the G peak position is at 1585 cm^{-1}. Also a well-defined 2D and D+G peak is visible in the spectrum indicating

the increased graphitic nature of the film. Comparing the position of the G peak and the I_D/I_G ratio to graphite, nanocrystalline graphite, diamond like carbon (DLC) with 20 % sp^3 content (a-C) and DLC with 85 % sp^3 (ta-c) following the graphitization trajectory suggested by Ferrari et al.[1], the data fits well to nanocrystalline graphite and thus confirms the nanocrystalline nature with 100 % sp^2 content. The evolution of the sharp 2D and D+G peak also confirms the highly graphitic nanocrystalline nature of the freestanding layer at 1200 °C.

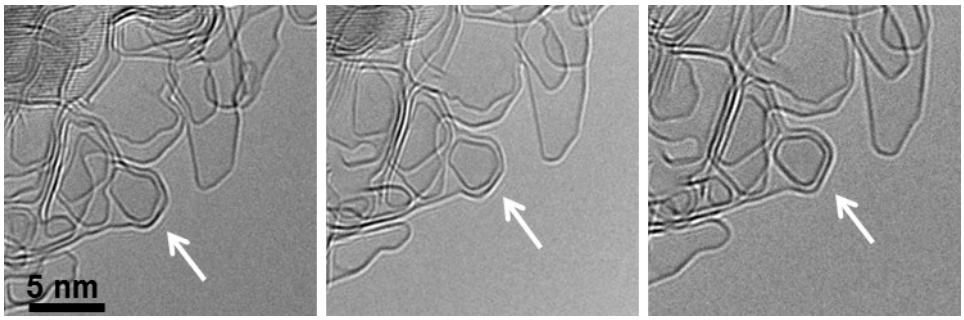

Figure S3: Transformation of a domain with disordered edges to defined faceted edges during prolonged heating. The size of the indicated domain is reducing during heating.

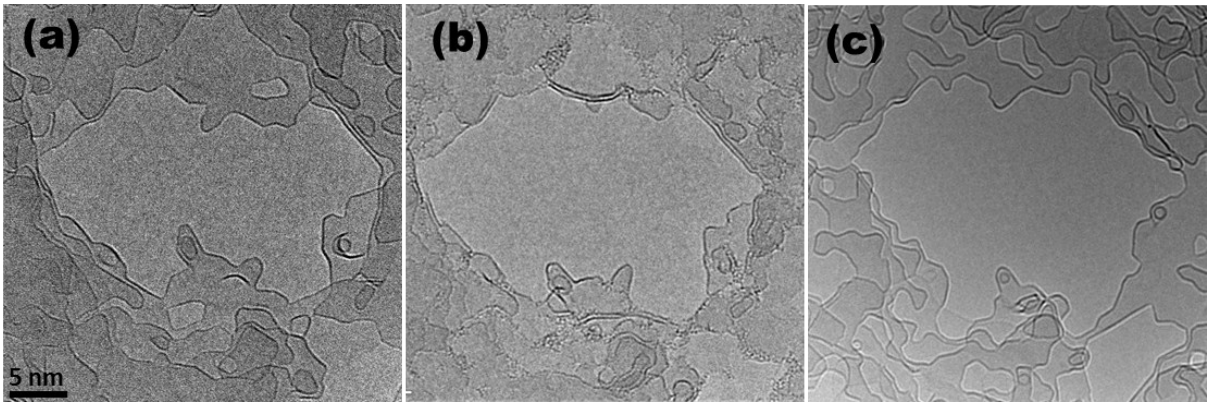

Figure S4 : Deposition of amorphous carbon on the sample (a) sample before adsorption of amorphous carbon (b) sample with the adsorbed amorphous carbon and (c) after reheating to 1200 °C. After re heating the sample, the amorphous carbon completely graphitizes increasing the dimensions of the domains.

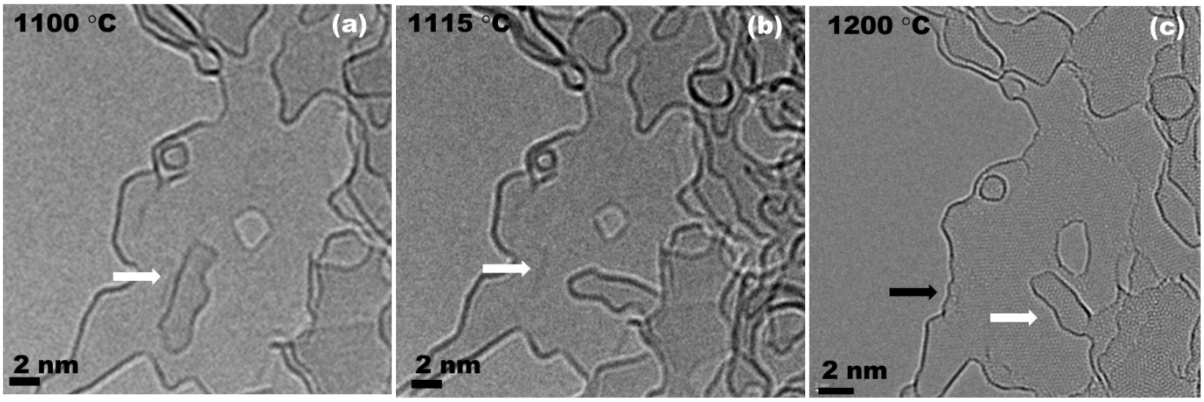

Figure S5: Migration of a small graphitic structure during heating (a, b). (c), high resolution images showing merging of the graphitic structure with a domain edge (marked by white arrows).The black arrow shows an area where a small cage like graphitic nuclei merged without discontinuity.

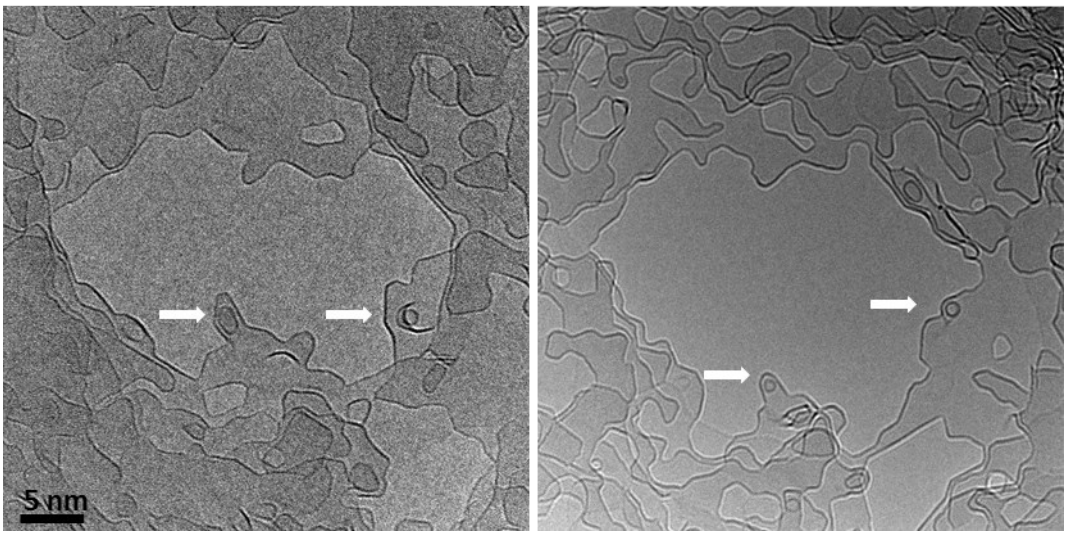

Figure S6: Trapped structures before and after reheating.

References

1. A. C. Ferrari and J. Robertson, *Philos. Trans. R. Soc. A Math. Phys. Eng. Sci.*, 2004, **362**, 2477–2512.