Supplementary Information

High temperature structural transformations of few layer graphene nanoribbons obtained by unzipping carbon nanotubes

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PXRD and Raman spectra of FWNTs and FLGn after unzipping

The bulk unzipping of FWNTs is also followed by powder X-ray diffraction. The reflection centered at 3.53 Å corresponding to the interwall spacing is shifted to about 9 Å upon unzipping. Major differences in the raman spectra before and after unzipping are related to the broadening of all D, G and 2D bands, along with the loss of the 2D bands and the increase of the D/G ratio.

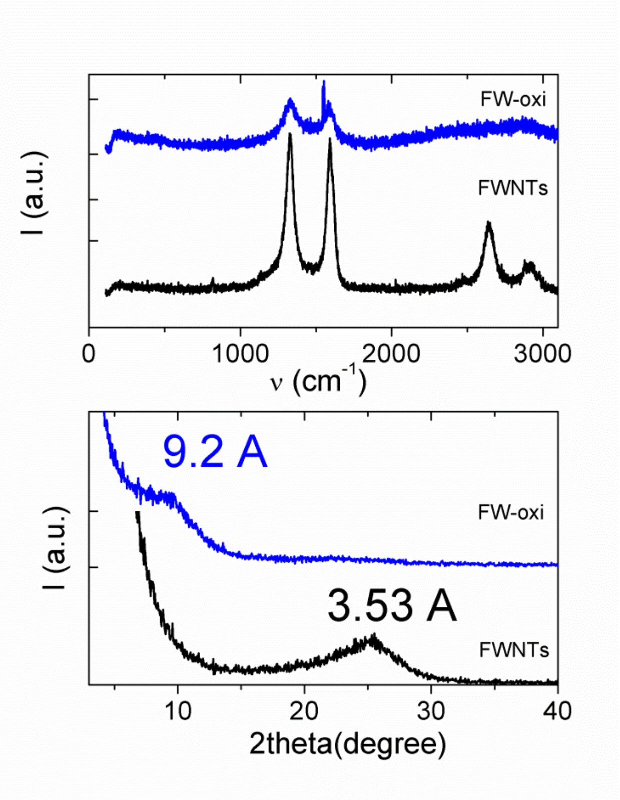

Figure S 1. a) Raman spectra of initial FWNTs and the resulting graphene oxide nanoribbons after oxidation with 700 wt% $KMnO_4$ in H_2SO_4 showing shifts in the region of the D, G and 2D bands b) XRD patterns for the same samples**.**

Raman spectra after high temperature annealing

Raman spectra after high temperature annealing at 1050ºC are shown in Figure S 2.

An additional shift on the raman frequencies is observed upon annealing. For Multilayer graphene nanoribbons the D band shifts to higher wavenumber after annealing whereas the G band shifts to lower wavenumbers in the reduced form of MLG.

In the case of Few Layer graphene nanoribbons there is hardly any shift after annealing in Argon at 1050ºC but there are changes in the relative intentisties of the D, G and 2D bands as well as in their width.

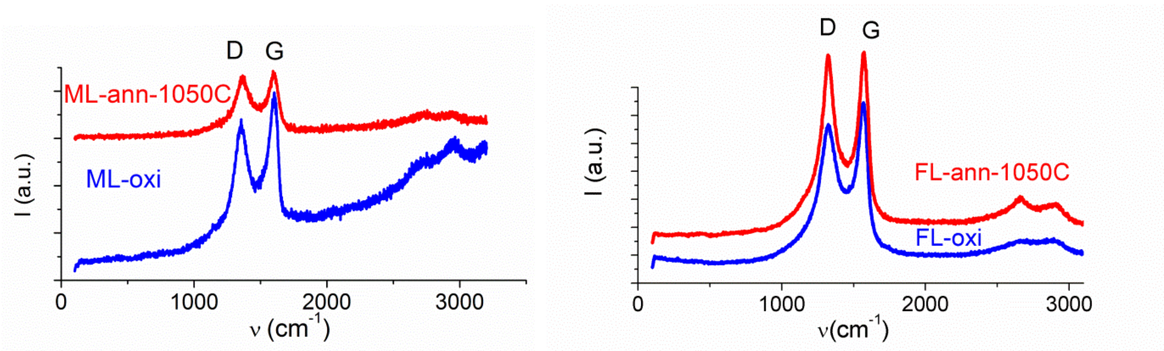

Figure S 2. Raman spectra of unzipped graphene nanoribbons before and after high temperature annealing at 1050ºC in Argon atmosphere for 2 hours. (a) MLGr (b) FLGr.

Comparison of pristine MWNTs with unzipped and annealed MLGnr

As discussed in the introduction these carbonaceous materials are very sensitive to the electron beam and therefore it is difficult to capture good high resolution images of them. It is nonetheless possible to distinguish between the layered edge of initial MWNTs, the unstructured edge of unzipped MWNTs and the layered edge of high temperature annealed MGNr.

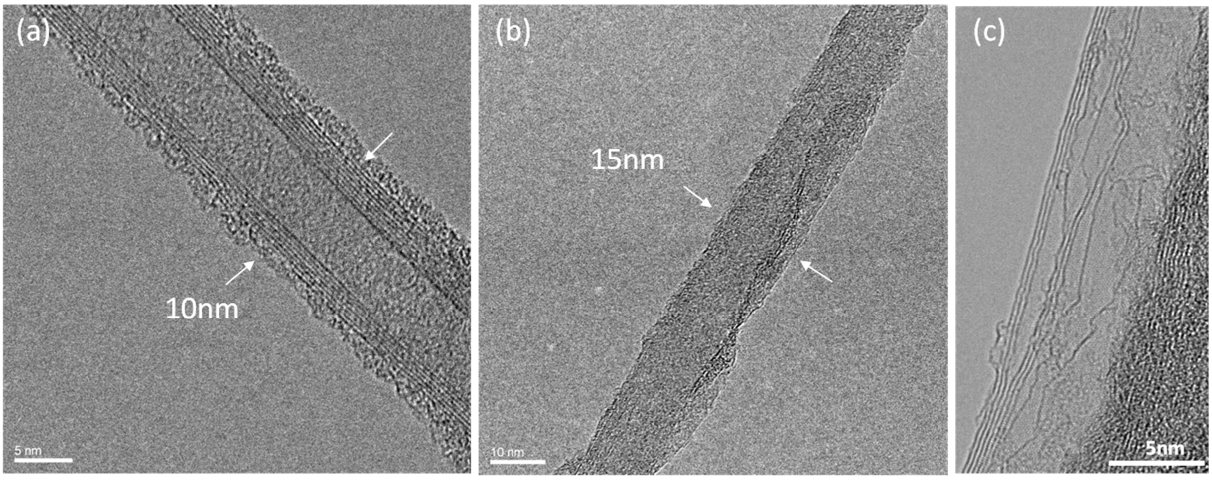

Figure S 3. TEM images comparing the edges of MWNTs during the unzzipping and high temperatura anneal process. (a) starting MWCNT (b) Unzipped MWNT with 800% $KMnO_4$ oxidant and (c) Unzipped MWNT with 800% $KMnO_4$ oxidant after annealing at 1300 ºC in vaccum.

TEM image of starting FWNTs bundle

FWNTs used for unzipping consisted of high purity nanotubes with no apreciable amount of amorphous carbon bridging the nanotubes. Due to the large lenght of pristine nanotubes the presence of catalytic nanoparticles is also scarce.

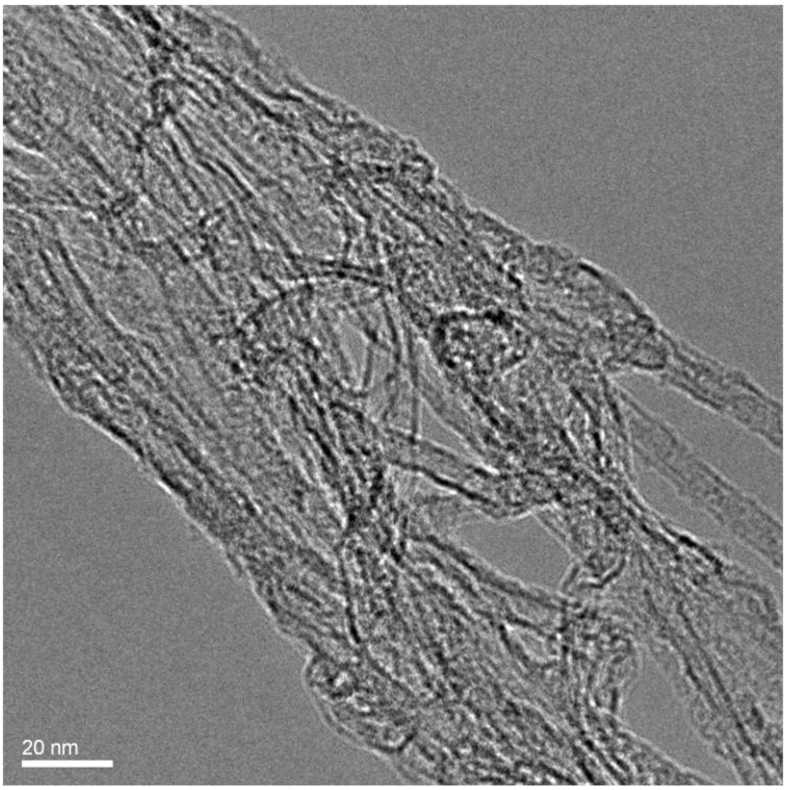

Figure S 4. TEM image of a bundle of starting FWNTs showing the sample purity.

TEM images of FLG after annealing at 1050ºC in argon

Aggregates of microcrystalline particles composed most of the sample as it can be observed in the low magnification image.

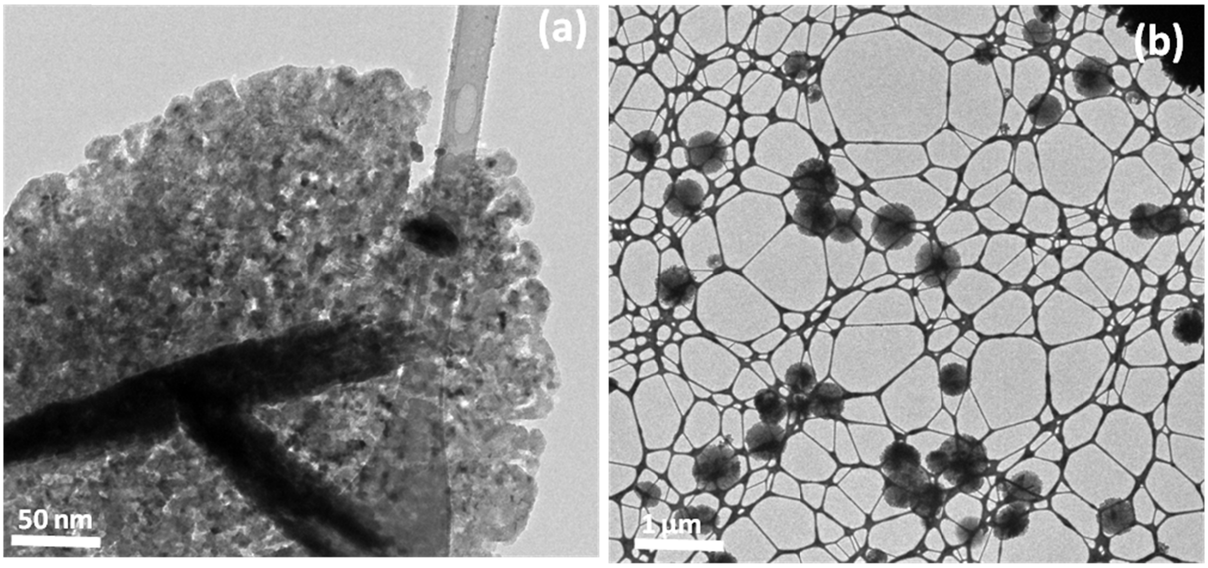

Figure S 5. TEM images of the structures formedof after annealing FLG nanoribbons in Argon at 1050ºC. (a) High magnification image of an aggregate. (b) Low magnification TEM image showing the abundance of aggregates in the TEM grid.

HRTEM image of graphitic foils after annealing FLG at 1400ºC in argon

A higher resolution image of Fig 4e is shown for clarity where the stacking of more than 30 layers can be observed.

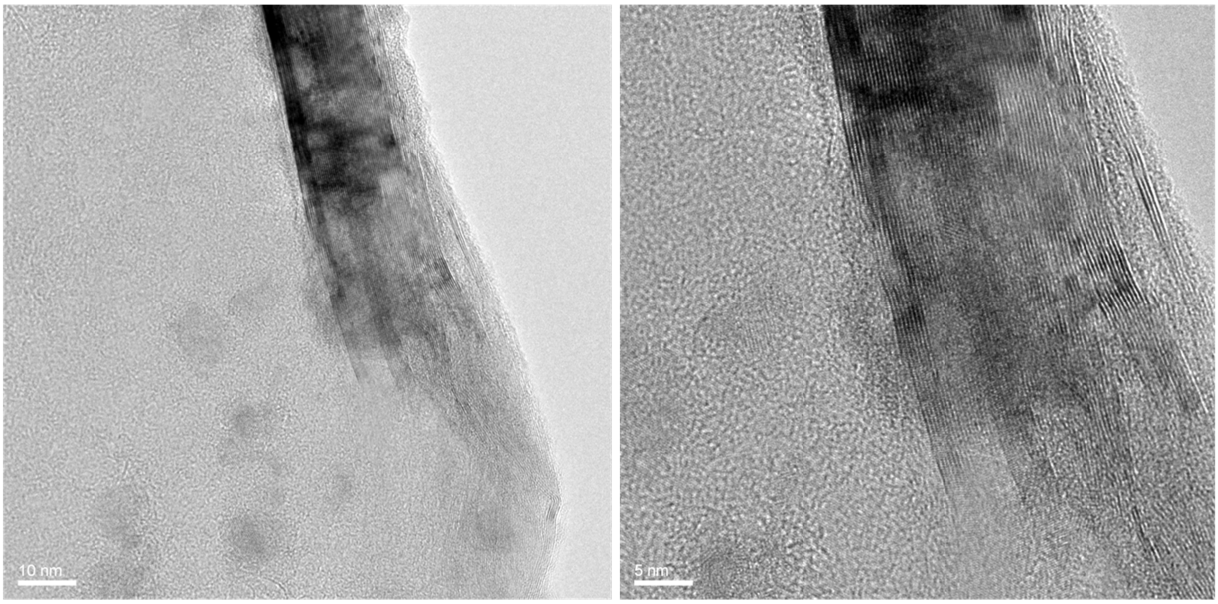

Figure S 6. TEM images of a folded edge of graphene like foil formed after annealing at 1400ºC FWGNr obtained from unzipping of FWNTs.