Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

High-quality functionalized few-layer graphene: Facile fabrication and doping with nitrogen as a metal-free catalyst for the oxygen reduction reaction

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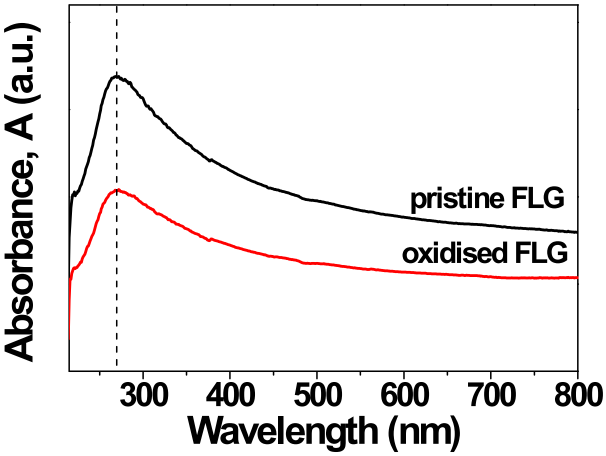

Fig. S1 UV−vis absorption spectra of pristine FLG and oxidised FLG at 225 ºC dispersed in a cationic surfactant cetyltrimethylammoniumbromide aqueous solution. The strong peak at about 269 nm arises from the π-plasmon resonance that is commonly observed in graphitic materials. The absorption spectra are featureless up to the visible region (800 nm), with a steady decrease in absorbance with the increase of wavelength.

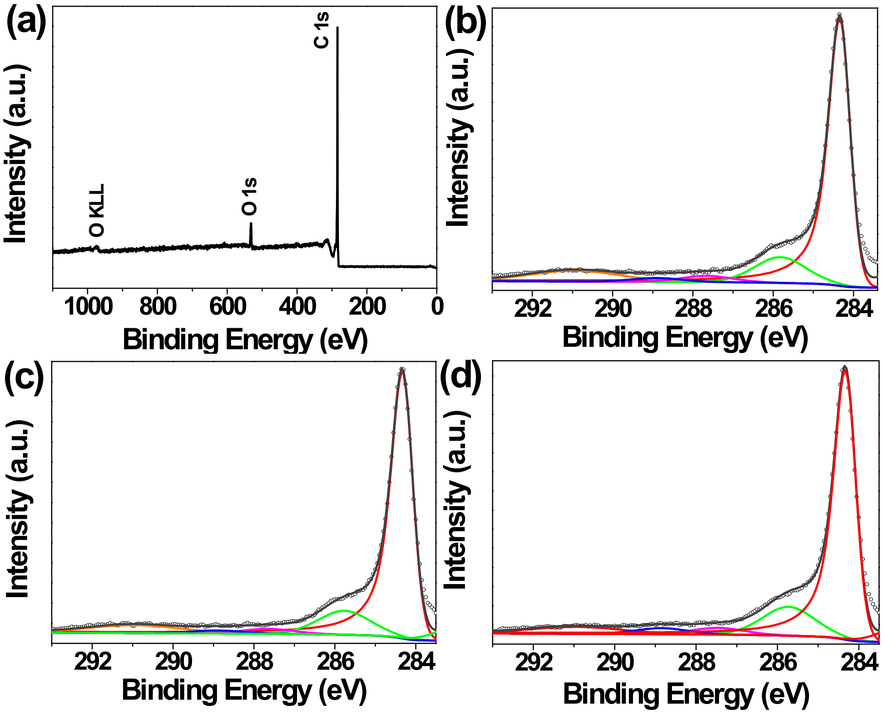

Fig. S2 (a) Wide–survey XPS spectrum. C 1s XPS spectra of functionalized FLG at (b) 125 ºC, (c) 175 ºC and (d) 275 ºC.

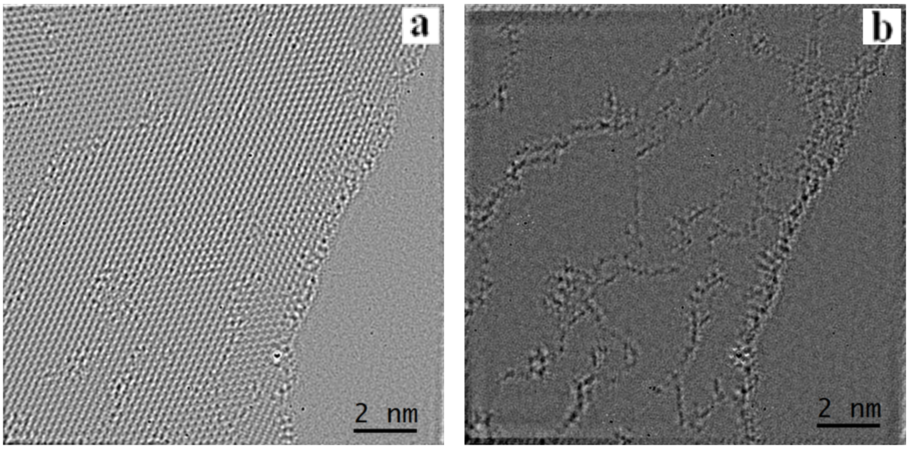

Fig. S3 (a) and (b) HRTEM images of a functionalized single–layer flake. (b) shows the disordered regions in image (a) which were highlighted by masking and subtracting the crystalline graphene identified through the FFT of image (a). Despite this, the graphitic lattices observed were substantially retained after oxidation. The flakes shown in Fig. S2 were functionalized at 125 ºC.

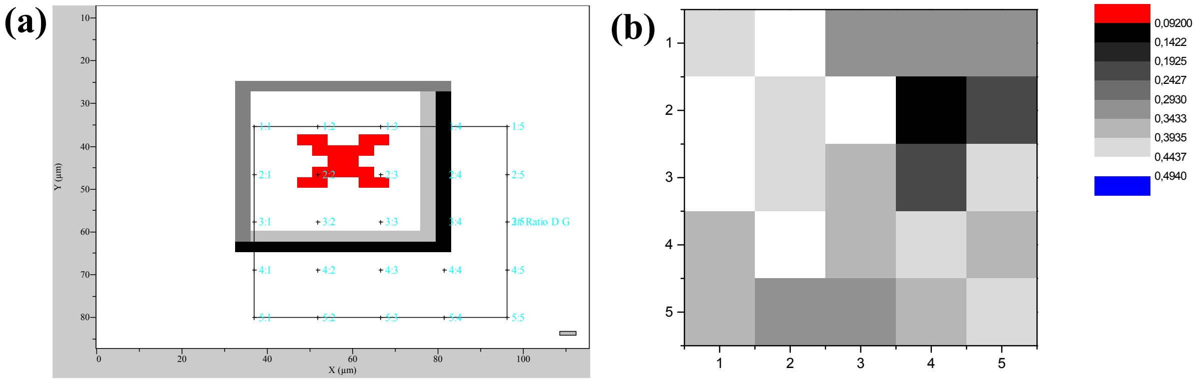

Fig. S4 (a) Optical microscopic image of the film sample prepared by vacuum filtration of the NMP dispersion of OFLG functionalized at 125 ºC. (b) Two–dimensional Raman map of the regions for the film as shown in image (a). The color gradient bar to the right of the map represents the D–to–G peak intensity ratio.

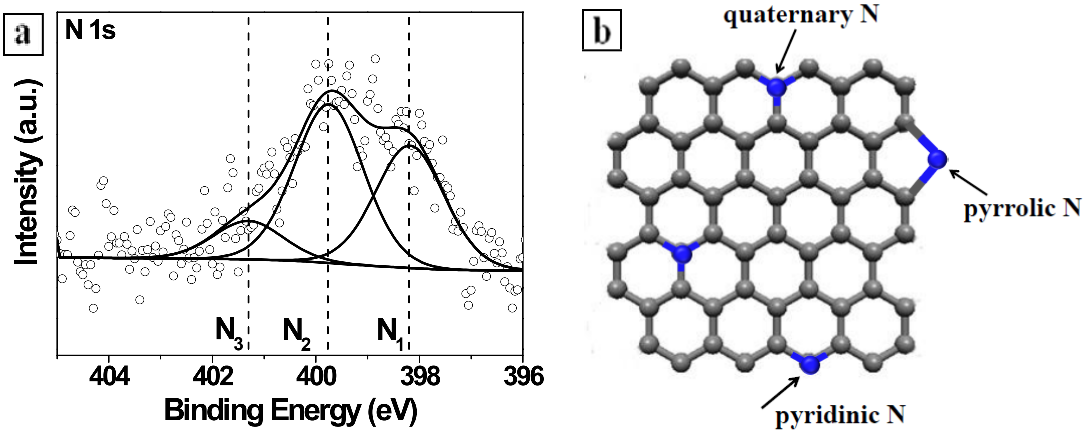

Fig. S5 (a) N 1s XPS spectrum of the N–doped FLG pretreated with HNO_3 vapour at 225 ºC. The peak deconvolution suggested three nitrogen functionalities bound to carbon centered at 398.6, 399.8 and 401.3 eV, corresponding to pyridinic N (N_1), pyrrolic N (N_2) and quaternary N (N_3), respectively. Pyridinic N atoms are located at the edges of graphene planes and each N atom is bound to two carbon atoms, donating its one p-electron to the aromatic π system. Pyrrolic N atoms bond with two carbon atoms in five-membered heterocyclic rings and to one hydrogen atom, contributing two p-electrons to the π system. Quaternary N atoms substitute carbon atoms and are involved in the graphene plane. The N doping level in the sample was estimated around 0.52 at.% by XPS measurements. The percentages of pyridinic N, pyrrolic N and quaternary N are 38.2, 49.7 and 12.1 at.%, respectively. (b) Schematic of bonding configuration of N functionalities in N–doped FLG.

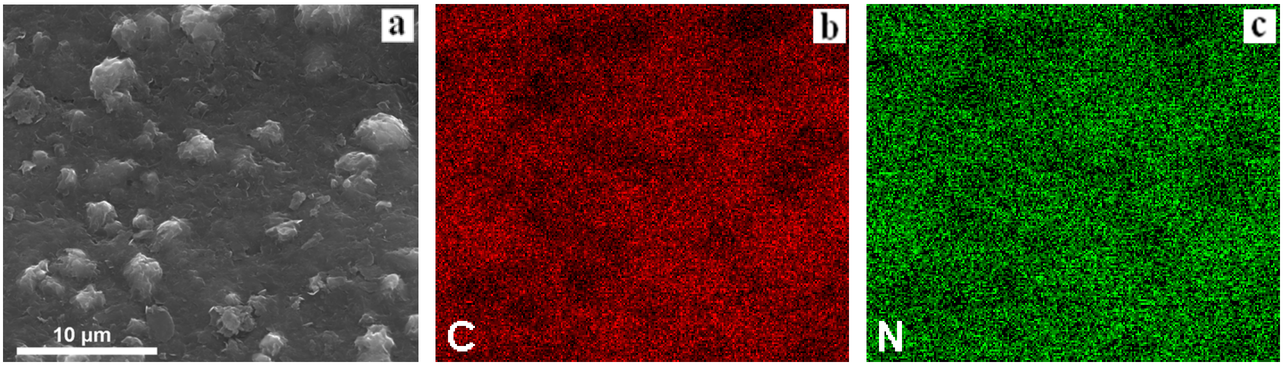

Fig. S6 (a) SEM image of the N–doped FLG pretreated with HNO_3 vapour at 225 ºC. (b)–(c) EDX mapping images of C and N, respectively, taken from the region shown in image (a).