Supporting Information

Single-step exfoliation and chemical functionalisation of graphene and hBN nanosheets with nickel phthalocyanine

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1-step Preparation Method (Graphene)

This involved mixing raw Ni-Pc and graphite powders together in a ratio 1:10 in weight. NMP was successively added to achieve an initial graphite concentration of 1 g{l}^{-1}. This was followed by bath sonication for 1 hour and centrifugation at 1000rpm for another hour.

The 1-step samples show strong absorption between 530-660nm. This is from the Q-band absorption from Ni-Pc and is due to the much high concentration of Ni-Pc present when compared to the 2-step (~1000 times more). There was also some evidence of the new peak at 702nm that appeared for the 2-step preparation method although it is much less clearly defined.

![Absorption Spectrum](image-url)
Raman spectroscopy of these samples gave no evidence of interaction between Ni-Pc and the graphene flakes. EDX failed to show any evidence of Ni on the flaked surface. Instead large aggregates of Ni-Pc were seen in the TEM, often near the graphene flakes (see figure). It is likely that the phthalocyanine molecules preferentially segregate to these larger aggregates instead of the graphene flakes. These are able to form due to the greater concentration of Ni-Pc in these samples.

We believe that there is some interaction between the flakes and the Ni-Pc clumps which accounts for some new features in the absorption spectrum, but there is no chemical functionalisation of the flake surface/edges.

Complimentary results were also obtained for h-BN flakes prepared by the one step method, with strong absorption between 530-600nm due to the presence of Ni-Pc clumps.
Figure 5.22 TEM BF diffraction contrast images. A-B are from the 1-step sample, C-D are from the 2-step. A) Circular BN flake near two Ni-Pc rods. B) ‘Square’ BN flake near Ni-Pc rods. C) BN flake showing some folding. D) Cluster of circular BN flakes. Flake at bottom of picture has lateral size measurement lines.