

Electronic Supplementary Information

**Cost-effective fabrication of graphene-like nanosheets from
natural microcrystalline graphite minerals by liquid
oxidation-reduction method**

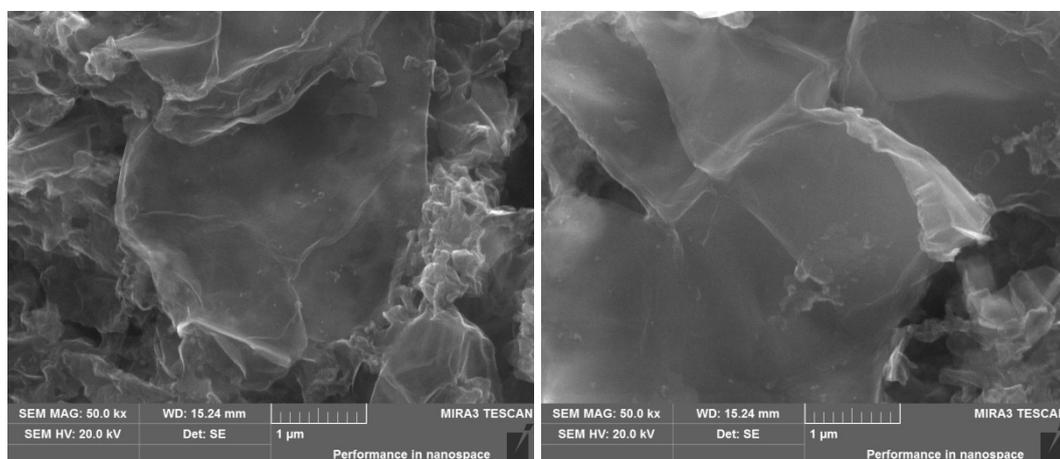
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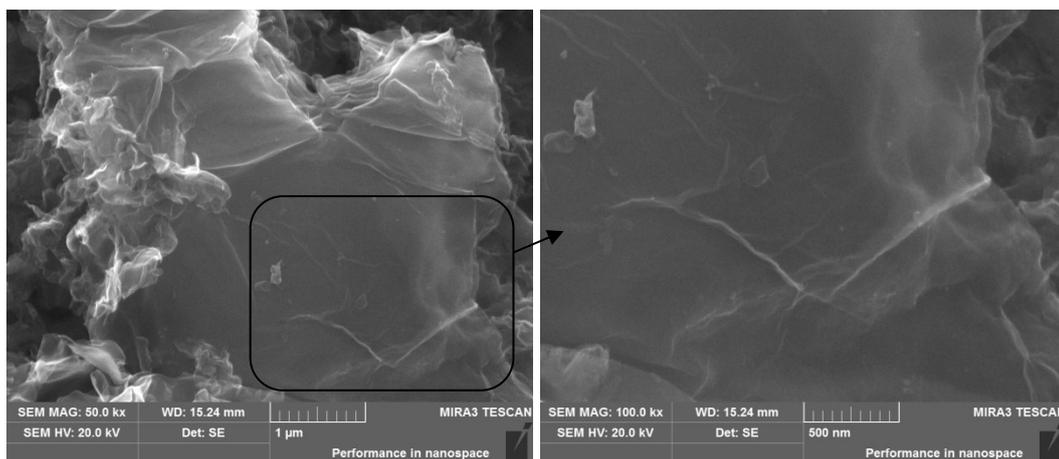


Fig. S1 SEM images of CRGO

Fig. S2 Low-magnification TEM images of (a) NMGM, (b)GO, (c)SRGO and (d)CRGO

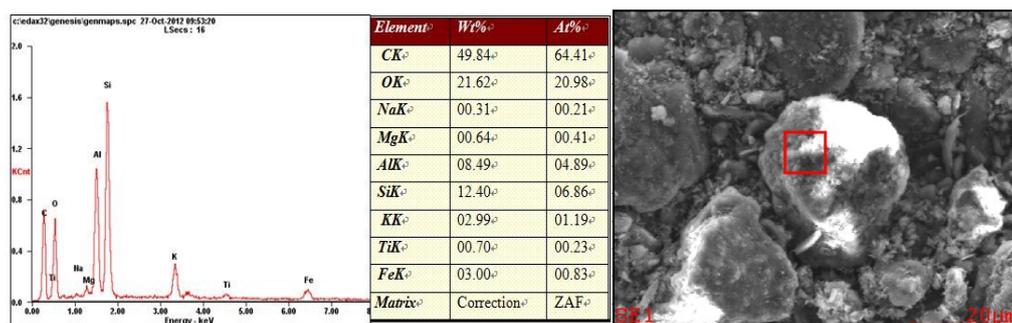


Fig. S3 EDX analysis for natural microcrystalline graphite minerals

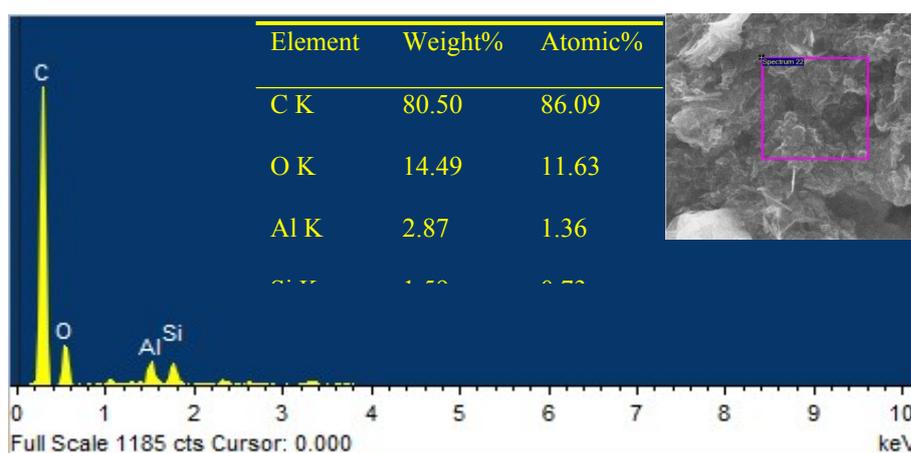


Fig. S4 EDX analysis for GNs reduced by solvothermal reduction

XPS O_{1s} spectra in Fig. S5 depict the detail of the oxygen functionalities for NMGM, GO, SRGO and CRGO. It can be concluded that the oxidation, and exfoliation and reduction

processes have changed the number of surface species. As shown in Fig.S5(a), the O1 peak at 532.65 eV is assigned to oxygen atoms from SiO₂, the O2 peak at 532.1 eV is assigned to oxygen atoms from C-O, and the O3 peak at 533.8 eV is assigned to oxygen atoms from O-C-Al. Combination with Table S1 content may indicate that the ore except some oxide impurities, natural MG mineral itself has also been a slight degree of oxidation. For GO, the O peak at 532.1 eV and 531.5 eV are assigned to oxygen atoms from C-O and C=O in epoxy, phenol, or carboxyl groups. In agreement with the results of the C_{1s} XPS spectra, the presence of the saturation values of C-O and C=O from C_{1s} and O_{1s} XPS spectra provides a good indication of complete oxidation of the edges that from carbonyl and carboxyl groups. For SRGO, the O peak at 532.3 eV and 530.9 eV are assigned to oxygen atoms from C-O and C=O in epoxy, phenol, or carboxyl groups. For CRGO, the O peak at 532.3 eV and 530.5 eV are assigned to oxygen atoms from C-O and C=O in epoxy, phenol, or carboxyl groups and the one at 535.0 eV to oxygen in chemisorbed oxygen species¹⁻³. The XPS results are corroborated by FTIR spectra.

Fig. S5 O_{1s} XPS spectra of NMGM, GO, CRGO and SRGO

Table S1 Chemical states of O atoms on NMGM, GO, SRGO, and CRGO with their relative atomic concentrations (atom %)

Samples	SiO ₂ (at. %)	O-C-Al(at. %)	C-O(at. %)	C=O(at. %)	Chemisorbed oxygen species(at. %)
NMGM	74.74	12.86	12.4	-	-
GO	-	1.57	76.28	22.15	-
SRGO	-	-	60.35	39.65	-
CRGO	-	-	-	97.08	2.92

Fig.S6 Formation mechanisms of GNs fabricated by the liquid oxidation-reduction method from NMGM

Fig.S7 SEM images of FG, MG, F-GO, M-GO, F-SRGO, M-SRGO, F-CRGO and M-CRGO

(The sample is too thick to measure)

Fig.S8 AFM images of FG, MG, F-GO, M-GO, F-SRGO, M-SRGO, F-CRGO and M-CRGO

Fig.S9 XRD patterns of FG, F-GO, F-SRGO and F-CRGO

Fig.S10 XRD patterns of MG, M-GO, M-SRGO and M-CRGO

1. C. Petit, M. Seredych, T.J. Bandosz, Revisiting the chemistry of graphite oxides and its effect on ammonia adsorption, *Journal of Materials Chemistry* 19(48) (2009) 9176-9185.
2. T.L. Barr, The nature of the relative bonding chemistry in zeolites: An XPS study, *Zeolites* 10(8) (1990) 760-765.
3. M. Bou, J.M. Martin, T.L. Mogne, L. Vovelle, Chemistry of the interface between aluminium and polyethyleneterephthalate by XPS, *Applied Surface Science* 47(2) (1991) 149-161.