Supplementary Materials

Fluorescent Moieties Through Alkaline Treatment of Graphene Oxide: A Potential Substitute to Replace CRM in wLEDS

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Supplementary S1. Synthesis Method

1 g of commercial graphite (Aldrich CAS Number:7782-42-5) was placed in a flask of 250 ml adding 23 ml of concentrated H_2SO_4 (96% acq.) at room temperature under stirring for 10 minutes. Then, the flask was placed into an ice bath and $KMnO_4$ was added in little aliquots every 5 minutes for 1 hour into the flask in order to prevent strong reaction. $KMnO_4$ (3 g) was employed in order to obtain the GO sample (see Table 1 in the main text).

After removal of the flask from the ice bath, 140 ml of deionized H_2O was slowly added; then, 10 ml of H_2O_2 (10%) was added in order to stop the reaction. The addition of H_2O_2 and H_2O was performed after the introduction of $KMnO_4$, under continuous stirring at room temperature and without sonication. H_2O_2 was added dropwise in small aliquots, and the resulting mixture was allowed to settle overnight at room temperature.

The brown-yellowish powder was separated from supernatant by centrifugation (4000 rpm for 10 minutes) and the powder was washed three times in HCl (10% v/v, 10 ml) and deionized water (30 ml), removing the washing water by centrifugation. Subsequently, the powder was dried in oven at 90 °C for 24 hours. An aliquot of the GO powder was treated in alkaline solution (KOH 10 %, 10 ml): the powder (100 mg) was placed in a falcon with conical bottom adding H_2O double distilled (10 ml) and KOH solution 0.1 M (15 ml). Subsequently, the solution was centrifuged (4000 rpm, 15 min) and after removing the supernatant, the powder was dried in oven for 24 h. In this way samples named GO_0.04_KOH have been obtained (see Table 1 in the main text). Other KOH treatments were applied in the same conditions with respect to the previous alkaline washing but with increasing KOH concentration, equal to 0.1 M (labelled as GO_0.1_KOH) and 0.2 M (labelled as GO_0.2_KOH). A third treatment was done in hot condition (1h at boiling point under reflux) by using a solution of KOH 1.78 M (labelled as GO_1.5_KOH).

Aqueous solutions were prepared dissolving the GO and KOH-treated GO powders in distilled water at a concentration of 1 mg/ml, then sonicating the solutions in an ultrasound bath for 30 minutes (120 W, 40 kHz), trying to maintain the bath at room temperature; clear solutions were obtained removing the undissolved solid and keeping temperature at 5°C. In this way, sonicated solutions reported in Table 1 in the main text have been obtained. Drop-casting was used to deposit GO, dispersed in an aqueous solution

after sonication, on Si substrate for AFM and Raman analyses. The substrates were left drying in air.

Supplementary S2. Characterization of the bw-GO (R1) and surnatants (S1) recovered by the KOH treatment

FTIR Analysis

Table S1. Spectral positions of the FTIR peaks recorded for sample base-washed using different KOH concentrations: 0.04 M (GO_0.04_KOH), 0.1 M (GO_0.1_KOH), 0.2 M (GO_0.2_KOH), 1.78 M (GO_1.5_KOH) and their corresponding supernatants.

Sample	νO-H	νCH	νC=O	νC=C	δ C-H	δ O-H	δ C-O	δ C-OH	C-H Ar.	C-C-C ‖
GO_0.04_KOH_SURN	3378	2842	-	1624	1343	1407	-	1112,1014	830,692	595
GO_0.04_KOH	3378	2842	-	1592	1343	1407	-	1083	835	-
GO_0.1_KOH_SURN	3180	3032 aryl	1734	1590 (low int.)	1345	1390	-	1112 1059	880,806,706,670	617
GO_0.1_KOH	3386	2927 alkyl 2850 alk.	1734	1627/ 1590 (lower int. shoulder)	1345	1390	1244	1112	979,830,706	-
GO_0.2_KOH_SURN	3176	3010 ar.	-	1570 (low int.)	1345	1390	-	1137 1059	879, 806,701	617
GO_0.2_KOH	-	2919 alk. 2845 alk.	1718	1570	1345	-	1215	1059	-	-
GO_1.5_KOH_SURN	3174	-	1730	1582 (low int.)	1352	1392	-	1055	985,880,806,702	595
GO_1.5_KOH	3295	2912 alk. 2842 alk	1730	1624 1582 similar int.	1352	1392	1255	1055	985	-

Raman Analysis

A detailed interpretation of Raman spectra of graphite, GO and KOH-treated GO is given in Ref. [Protopapa et al., 2022]. We can notice in Figure 3 of that paper that Raman spectra of the GO samples significantly differ from that of graphite since the oxidation process generates much more structural defects. In particular, the D band becomes much more intense with respect to the original commercial graphite due to the increase of defects. Moreover, the broadening of the G and D′ peaks in the GO samples spectra are such that the two bands collapse in a single G line (indicated as G+D′ in Figure 3 of Protopapa et al., 2022 and in Figure S1). We also notice the emergence of a broad feature at about 1500 cm^{-1} that is referred to as D_3. The D_3 band does not produce any maximum in the observed spectra but its introduction is necessary to correctly fit the spectral shape between the D and the G bands. The D_3 band is present in highly defective carbon materials like carbon black and is assigned to the presence of amorphous carbon [Jawhar et al., 1995, Ferrari et

al., 2007]. Figure S1 shows the Raman spectra of KOH-treated samples at increasing KOH concentration. As one can note, the height of the "saddle" between the D and G bands increases as the KOH concentration increases, due to an increase of the D_3 band related to amorphization.

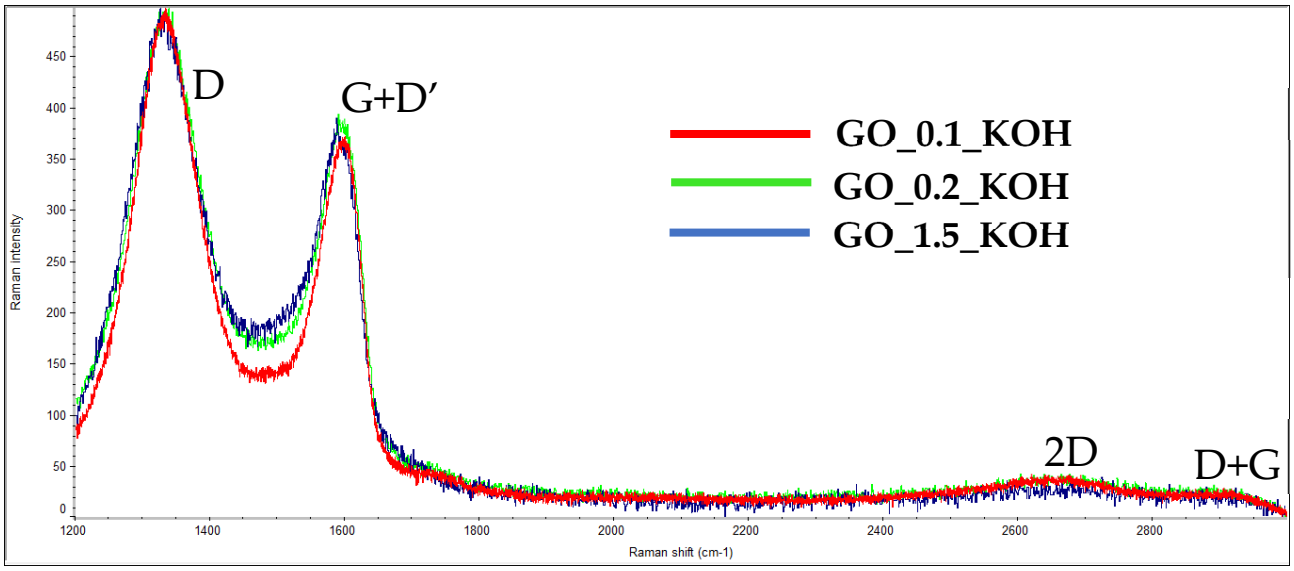


***Figure S1**: Raman spectra of KOH-treated GO at increasing concentrations of KOH.*

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AFM Analysis

GO_0.04_KOH_SURN

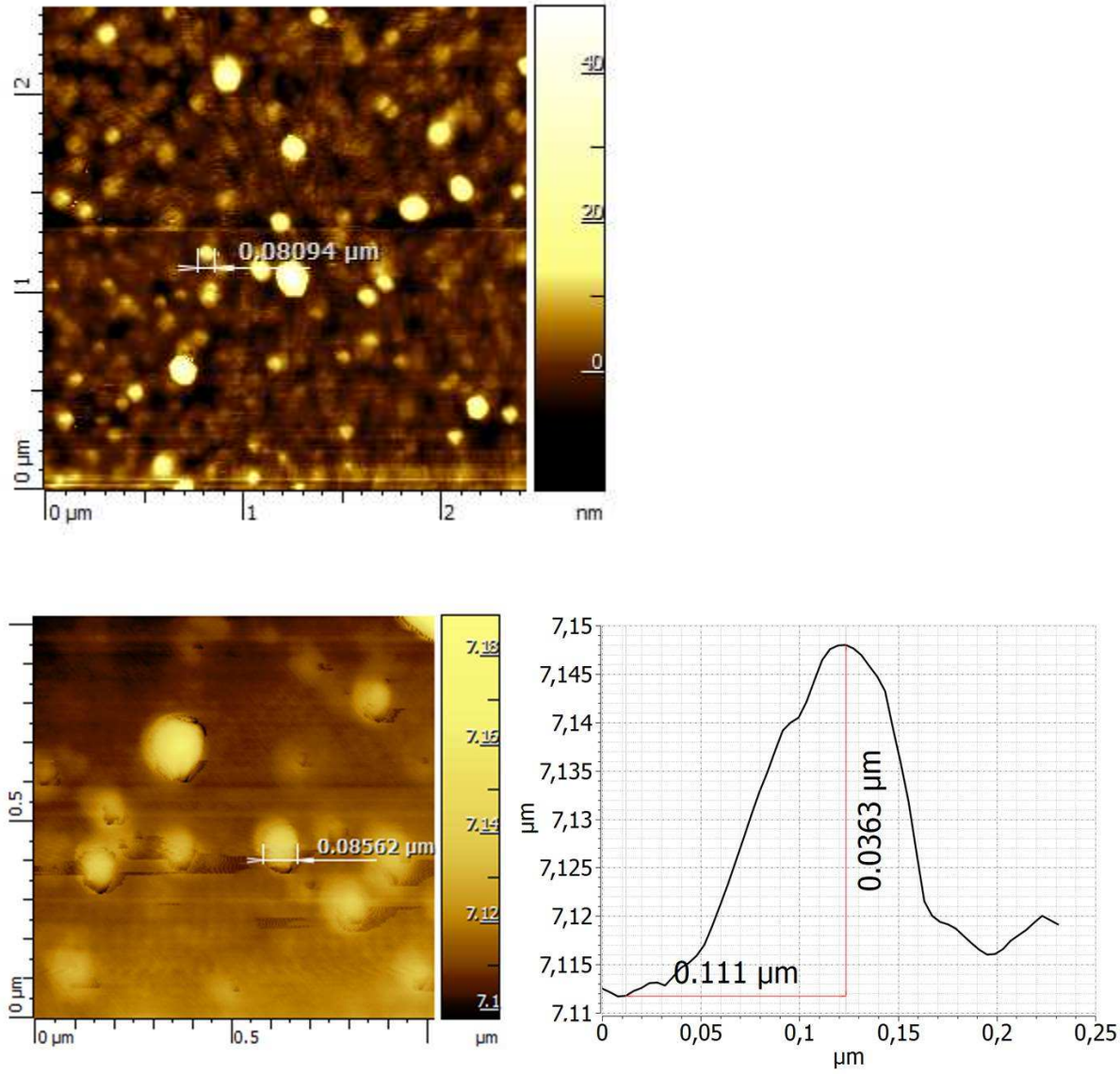


Figure S2. *AFM image and z-profile for sample GO_0.04_KOH_SURN dropped onto a silicon substrate.*

GO_0.1_KOH_SURN

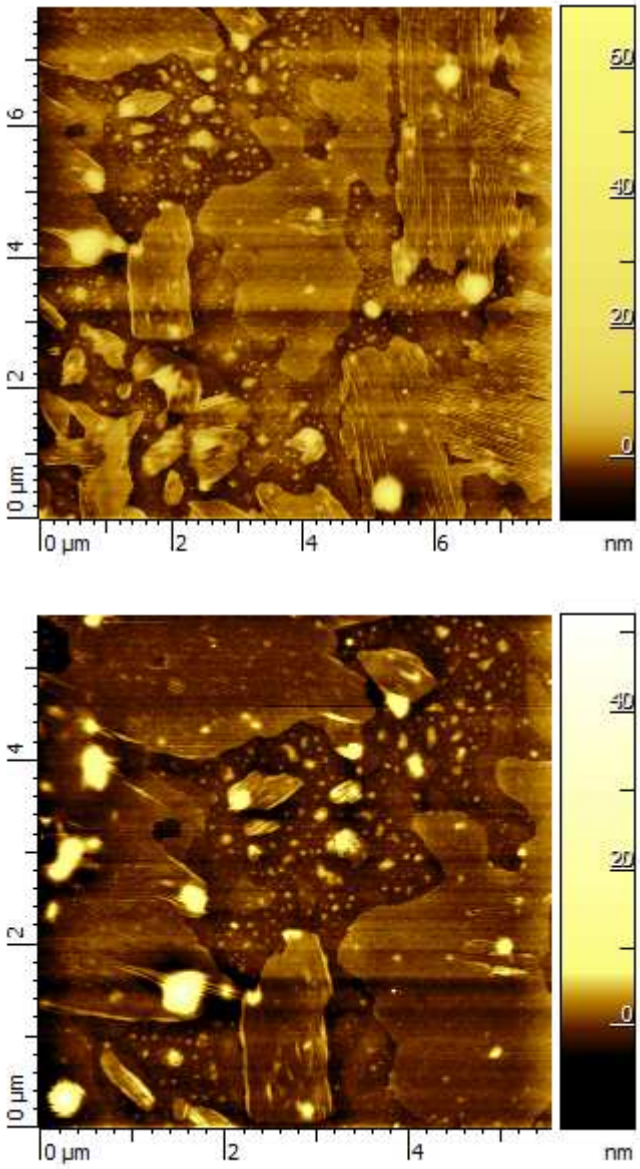


Figure S3 *AFM images for sample GO_0.1_KOH_SURN dropped onto a silicon substrate.*

GO_0.2_KOH_SURN

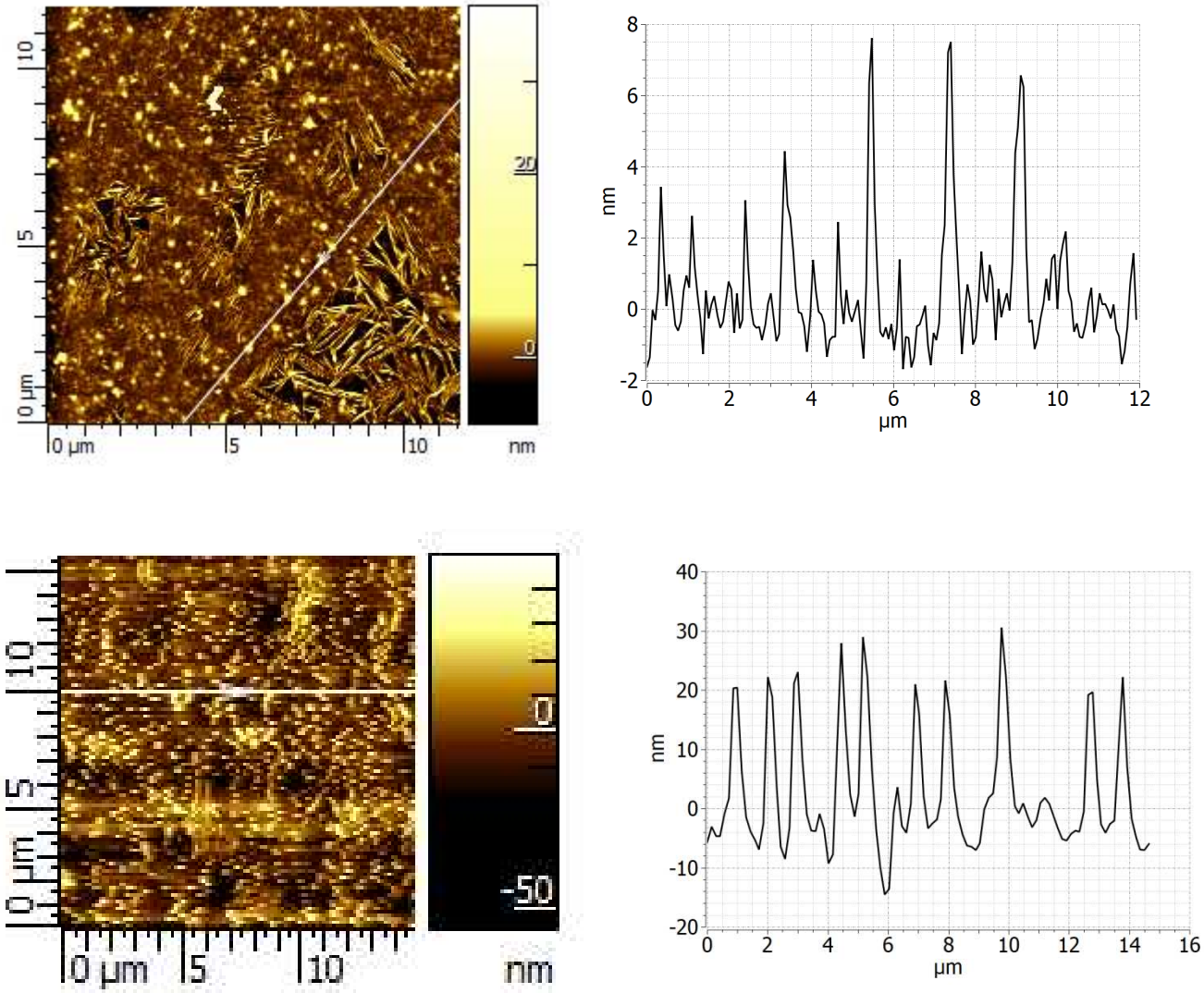


Figure S4 *AFM images and height profile for sample GO_0.2_KOH_SURN dropped onto a silicon substrate.*

GO_1.5_KOH_SURN

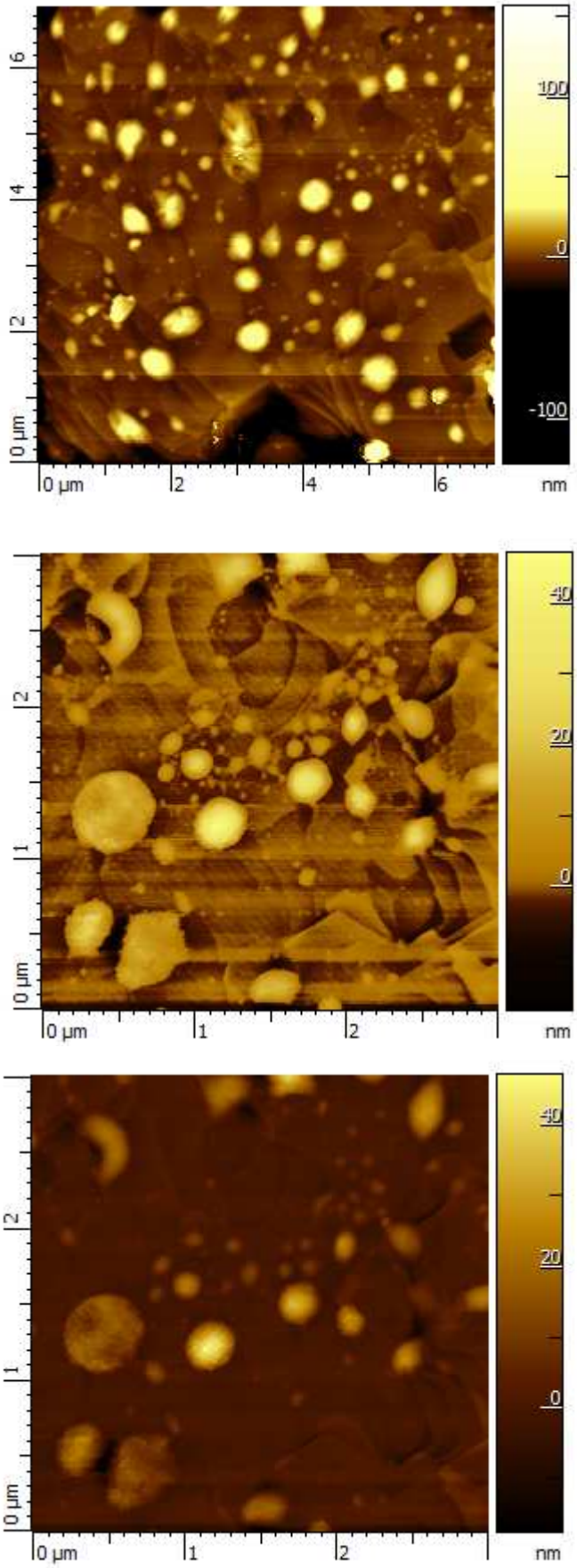


Figure S5 *AFM images for sample GO_1.5_KOH_SURN dropped onto a silicon substrate.*

References

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