

1. Preparation of dispersions

1.1 FLG samples

Functionalized FLG water dispersions were diluted up to 0.1 %wt in a volumetric flask.

All types of functionalized FLG samples (Figure S1**Error! Reference source not found.**) formed stable water dispersions without further treatment.



Figure S1: FLG 0.1 %wt water dispersions.

1.2 CNT samples

Functionalized CNT water dispersions were diluted up to 0.1 %wt in a volumetric flask. In this case, all types of functionalized CNTs could not form stable dispersions in water (Figure S2).

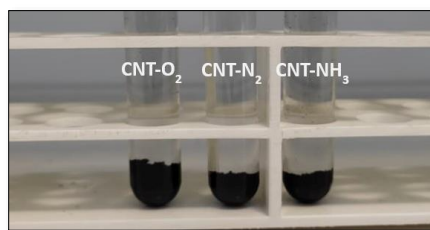


Figure S2: CNTs 0.1 %wt water dispersions

Stabilization of CNT dispersions

Preparing stable CNT dispersions is a challenging task due to their chemical structure. The delocalization of π electrons makes CNTs highly conducting and reduces their ability to absorb chemical moieties via π - π stacking interaction. Also, their high aspect ratio leads to bundling, which takes place through strong van der Waals interactions of 500 eV/ μm energy of tube-tube contact. Such a strong intermolecular attraction reduces the solubility, as solvent molecules are not able to enter between CNTs [17].

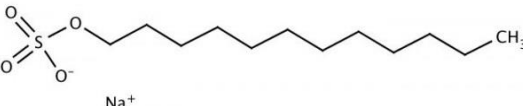
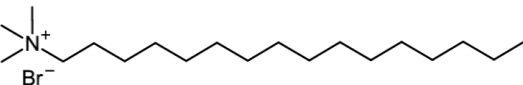
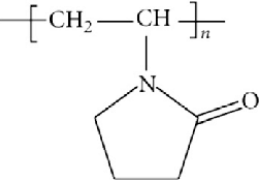
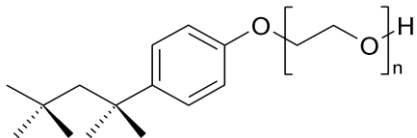
The stabilization of CNT dispersions can be approached in two ways: mechanically and chemically. The mechanical approach includes sonication and high-shear mixing. The laboratory equipment included a 3510 BRANSON sonication bath and a UP200St Hielscher ultrasonic processor (Figure S3). This mechanical process is not efficient on its own. The dispersion shows very poor stability, and it settles after a short time. Also, sonication cannot be performed many times or for a prolonged time, because it can result in CNT fragmentation, decreasing its aspect ratio [18].



Figure S3: 3510 BRANSON sonication bath (left) and Thielsen ultrasonic processor (right).

On the other hand, the chemical approach for increasing stabilization includes the utilization of surfactants. This is a non-covalent method based on the absorption of chemical moieties onto the CNT surface, either via π - π stacking interaction, in the case of uncharged surfactants, or through coulomb attraction in the case of charged surfactants, rendering them soluble in water [19,20]. Four different types of surfactants were evaluated: Sodium Dodecyl Sulphate (SDS), Cetrimonium bromide (CTAB), Polyvinylpyrrolidone (PVP), and Triton X 100. All tested surfactants are presented in **Error! Reference source not found.**

Table S1: Table of surfactants that were tested.

| Surfactant | | Chemical Formula | Chemical Structure | Type |
|----------------------------|---------|--|--|-----------|
| Sodium Sulphate (SDS) | Dodecyl | $\text{CH}_3(\text{CH}_2)_{11}\text{OSO}_3\text{Na}$ |  | Anionic |
| Cetrimonium (CTAB) | bromide | $(\text{C}_{16}\text{H}_{33})\text{N}(\text{CH}_3)_3\text{Br}$ |  | Cationic |
| Polyvinylpyrrolidone (PVP) | | $(\text{C}_6\text{H}_9\text{NO})_n$ |  | Non-ionic |
| Triton X 100 | | $\text{C}_{14}\text{H}_{22}\text{O}(\text{C}_2\text{H}_4\text{O})_n$ |  | Non-ionic |

Surfactant selection—Step 1

All available dispersions were further diluted up to 0.05 %wt. The surfactants were added to the dispersions in a surfactant-CNT ratio of 1:1. All samples prepared are presented in Table S2.

Table S2: Samples prepared for step 1.

| Filler type | Dispersion Concentration | Surfactant | Filler/Surfactant |
|-------------|--------------------------|------------|-------------------|
|-------------|--------------------------|------------|-------------------|

| | | | |
|---------------------|----------|---------------|-----|
| CNT-O ₂ | 0.05 %wt | No surfactant | 1:1 |
| | 0.05 %wt | SDS | 1:1 |
| | 0.05 %wt | CTAB | 1:1 |
| | 0.05 %wt | PVP | 1:1 |
| | 0.05 %wt | Triton X100 | 1:1 |
| CNT-N ₂ | 0.05 %wt | No surfactant | 1:1 |
| | 0.05 %wt | SDS | 1:1 |
| | 0.05 %wt | CTAB | 1:1 |
| | 0.05 %wt | PVP | 1:1 |
| | 0.05 %wt | Triton X100 | 1:1 |
| CNT-NH ₃ | 0.05 %wt | No surfactant | 1:1 |
| | 0.05 %wt | SDS | 1:1 |
| | 0.05 %wt | CTAB | 1:1 |
| | 0.05 %wt | PVP | 1:1 |
| | 0.05 %wt | Triton X100 | 1:1 |

All samples were sonicated for 1 h in a 3510 BRANSON sonication bath. After 24 h, the samples were observed and evaluated (Figure S4).

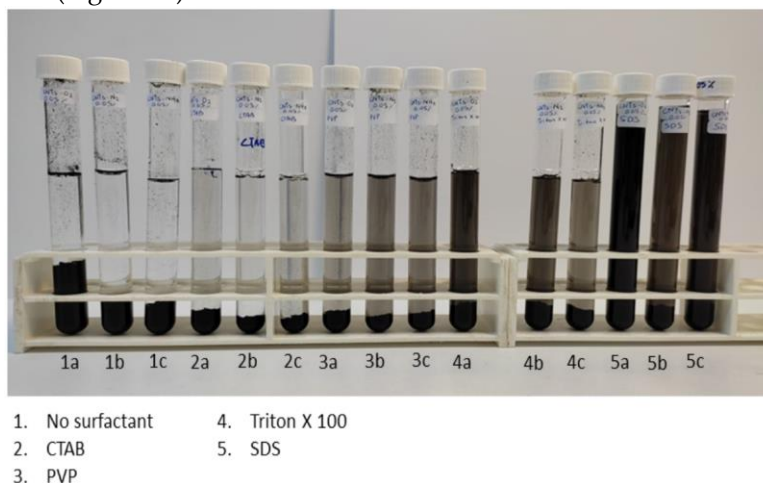


Figure S4: Samples 24 h after sonication for 1 h in 3510 BRANSON sonication bath.

As shown in Figure S4, this first test clearly shows that all samples without the surfactant and the samples with the CTAB surfactant had very poor dispersion capacities. The next steps proceeded without those samples.

Surfactant selection—Step 2

All samples tested in this step (shown in Table S3) were ultrasonicated using the Thielsen ultrasonication probe. The sonication was set to be pulsive with a 0.5/0.5 sec sonication-to-pause ratio. The amplitude was set to 40%, and the sonication lasted for 45 min for each sample. The samples after the ultrasonication are presented in Figure S5.

Table S3: Samples prepared for step 2.

| Filler type | Dispersion Concentration | Surfactant | Filler/Surfactant |
|---------------------|--------------------------|-------------|-------------------|
| CNT-O ₂ | 0.05 %wt | SDS | 1:1 |
| | 0.05 %wt | PVP | 1:1 |
| | 0.05 %wt | Triton X100 | 1:1 |
| CNT-N ₂ | 0.05 %wt | SDS | 1:1 |
| | 0.05 %wt | PVP | 1:1 |
| | 0.05 %wt | Triton X100 | 1:1 |
| CNT-NH ₃ | 0.05 %wt | SDS | 1:1 |
| | 0.05 %wt | PVP | 1:1 |
| | 0.05 %wt | Triton X100 | 1:1 |

The addition of surfactants combined with processing dispersions with the ultrasonication probe was effective; thus, all samples formed good-quality dispersions, though after more than 72 h, the most stable dispersions were those with surfactant Triton X 100. This result agrees with the literature [18].



Figure S5: Samples after ultrasonication with Thielsen sonication probe at 72 h.

As a result, the surfactant that was selected to be used for CNT dispersion stabilization was Triton X 100 in a ratio of 1:1 with the CNTs.

2. XPS analysis

X-Ray Photoelectron Spectroscopy (XPS) was performed on the individual functionalised nanomaterial powders used in each dispersion and the unfunctionalised powders to confirm that functionality had been applied and to assess the functionalisation type and degree.

The XPS element survey data detailed in

Table S4 highlight in green where significant changes to atomistic concentrations were observed for functionalised materials. Nitrogen groups were present in the functionalisation type with NH₃ and N₂, and oxygen groups in the functionalisation type with O₂, suggesting that functionalisation proceeded as intended. Furthermore, the XPS data confirm that the additional functionalisation present was chemically bonded to the carbon/graphitic material.

Table S4: XPS element survey results of unfunctionalised and functionalised nanomaterials.

| Nano Type | Functionalisation | O 1s (%) | C 1s (%) | N 1s (%) |
|-----------|-------------------|----------|----------|----------|
| FLG | None | 5.66 | 94.02 | 0.32 |
| FLG | NH ₃ | 5.88 | 92.75 | 1.37 |
| FLG | N ₂ | 6.24 | 93.06 | 0.71 |
| FLG | O ₂ | 9.33 | 90.31 | 0.36 |
| CNT | None | 1.15 | 98.85 | - |
| CNT | NH ₃ | 0.8 | 99.07 | 0.13 |
| CNT | N ₂ | 1.54 | 97.61 | 0.85 |
| CNT | O ₂ | 7.51 | 92.49 | - |

High-resolution XPS was performed on the nanomaterial powders, to ascertain a more detailed view of the functional groups being added. The XPS high-resolution dataset and graphs for plasma-functionalised nanomaterials can be found below.

Table S5: XPS high-resolution data peak assignment FLG materials.

| Name | FLG None | | FLG NH ₃ | | FLG N ₂ | | FLG O ₂ | |
|------------|----------|----------|---------------------|----------|--------------------|----------|--------------------|----------|
| | Position | %At Conc | Position | %At Conc | Position | %At Conc | Position | %At Conc |
| C 1s (SP2) | 284.44 | 59.76 | 284.43 | 57.5 | 284.46 | 60.03 | 284.44 | 53.84 |
| C 1s (SP2) | 286.89 | 7.06 | 286.88 | 5.21 | 286.91 | 4.84 | 286.88 | 4.04 |
| C 1s (SP2) | 290.75 | 9.48 | 290.74 | 8.77 | 290.77 | 9.03 | 290.74 | 8.62 |
| C 1s (SP2) | 294.25 | 1.99 | 294.24 | 1.41 | 294.27 | 1.19 | 294.24 | 1.46 |
| C 1s (SP3) | 284.94 | 9.42 | 284.93 | 12.09 | 284.96 | 9.62 | 284.94 | 12.42 |
| C=O | 289.6 | 0.82 | 289.43 | 1.28 | 289.34 | 1.26 | 288.766 | 2.13 |
| C-O/C-N | 286.55 | 3.21 | 286.54 | 3.8 | 286.58 | 4.35 | 286.66 | 5.22 |
| O-C=O | 288.49 | 2.24 | 288.21 | 2.63 | 288.26 | 2.49 | 288.5 | 2.37 |
| N 1s | 398.5 | 0.25 | 399.44 | 1 | 399.33 | 0.38 | 398.54 | 0.09 |
| N 1s | - | - | 400.93 | 0.21 | 400.7 | 0.25 | 400.3 | 0.23 |
| N 1s | - | - | 403.57 | 0.13 | 403.38 | 0.08 | 402.5 | 0.08 |
| O 1s | 531.46 | 1.77 | 531.44 | 2.08 | 531.46 | 2.01 | 531.51 | 2.95 |
| O 1s | 533.05 | 3.17 | 533.03 | 3.08 | 533.05 | 3.5 | 533.1 | 5.35 |
| O 1s | 535.28 | 0.45 | 535.26 | 0.39 | 535.28 | 0.51 | 535.33 | 0.62 |
| O 1s | 537.28 | 0.27 | 537.27 | 0.27 | 537.28 | 0.29 | 537.34 | 0.37 |
| O 1s | 539.39 | 0.13 | 539.37 | 0.13 | 539.39 | 0.18 | 539.44 | 0.2 |

Table S6: XPS high-resolution data peak assignment CNT materials.

| Name | CNT None | CNT NH ₃ | CNT N ₂ | CNT O ₂ |
|------|----------|---------------------|--------------------|--------------------|
|------|----------|---------------------|--------------------|--------------------|

| | Position | %At Conc | Position | %At Conc | Position | %At Conc | Position | %At Conc |
|------------|----------|-------------|----------|-------------|----------|-------------|----------|-------------|
| C 1s (SP2) | 284.45 | 70.86 | 284.44 | 71.45 | 284.43 | 69.98 | 284.44 | 61.94 |
| C 1s (SP2) | 286.89 | 8.67 | 286.89 | 7.52 | 286.88 | 6.6 | 286.88 | 5.37 |
| C 1s (SP2) | 290.76 | 10.5 | 290.76 | 10.27 | 290.74 | 10.07 | 290.74 | 9.37 |
| C 1s (SP2) | 294.26 | 2.23 | 294.25 | 2.05 | 294.24 | 1.85 | 294.24 | 1.72 |
| C 1s (SP3) | 284.96 | 6.16 | 284.94 | 6.34 | 284.93 | 6.66 | 284.94 | 6.53 |
| C=O | 289.45 | 0.09 | 287.52 | 0 | - | - | 289.46 | 0.47 |
| C-O/C-N | 286.2 | 0.09 | 286.38 | 0.49 | 286.35 | 1.74 | 286.36 | 4.05 |
| O-C=O | 289.14 | 0.49 | 288.7 | 0.68 | 288.57 | 0.87 | 288.44 | 2.75 |
| N 1s | - | - | 399.21 | 0.14 | 398.3 | 0.14 | - | - |
| N 1s | - | - | 400.81 | 0.07 | 399.91 | 0.42 | - | - |
| N 1s | - | - | - | - | 402.1 | 0.11 | - | - |
| O 1s | 531.99 | 0.41 | 531.3 | 0.41 | 531.47 | 0.62 | 531.69 | 2.54 |
| O 1s | 533.58 | 0.4 | 532.89 | 0.44 | 533.06 | 0.76 | 533.28 | 4.16 |
| O 1s | 535.81 | 0.1 | 535.12 | 0.13 | 535.29 | 0.19 | 535.51 | 0.66 |
| O 1s | - | - | - | - | - | - | 537.52 | 0.26 |
| O 1s | - | - | - | - | - | - | 539.62 | 0.16 |

FLG unfunctionalised:

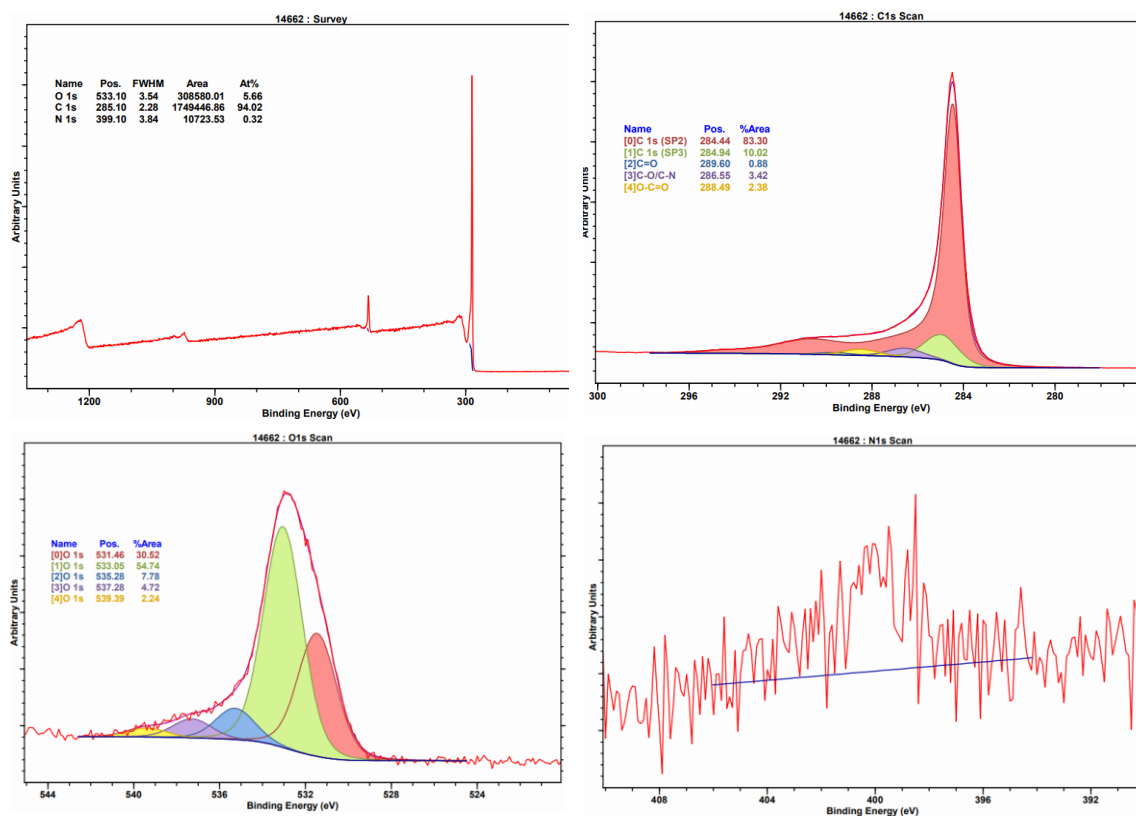


Figure S6: FLG unfunctionalised.

FLG NH₃:

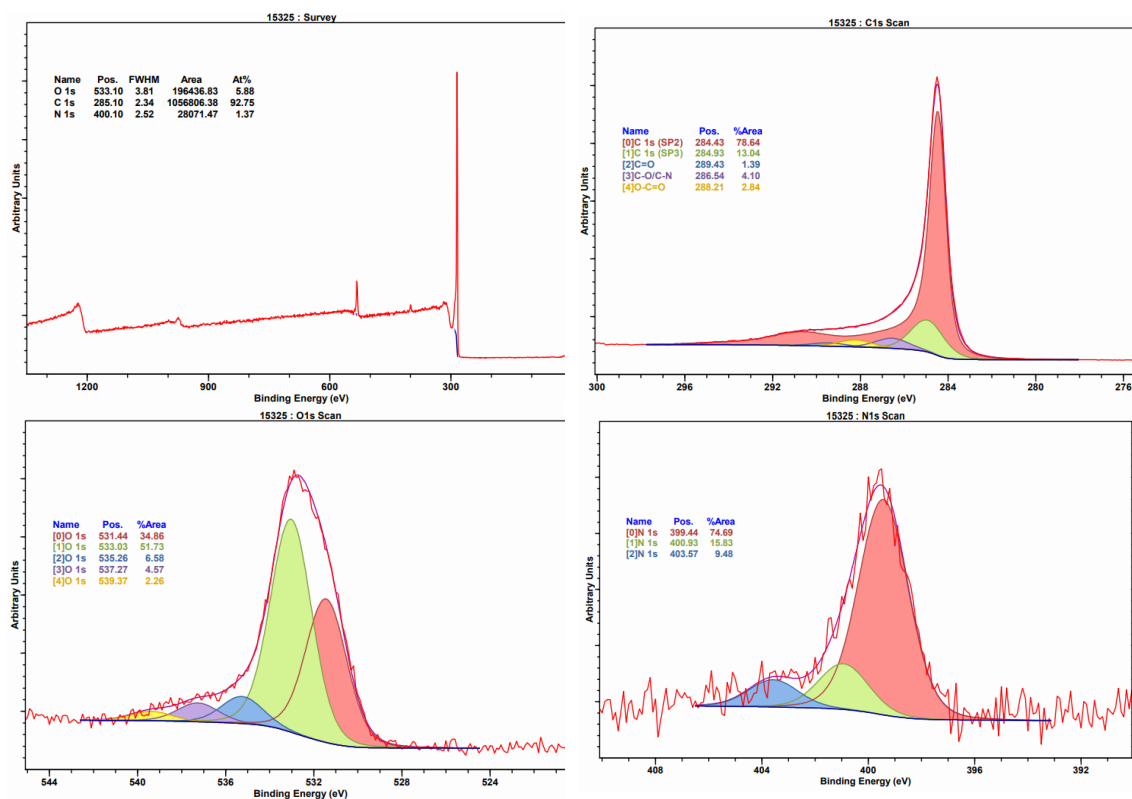


Figure S7: FLG NH3.

FLG N₂:

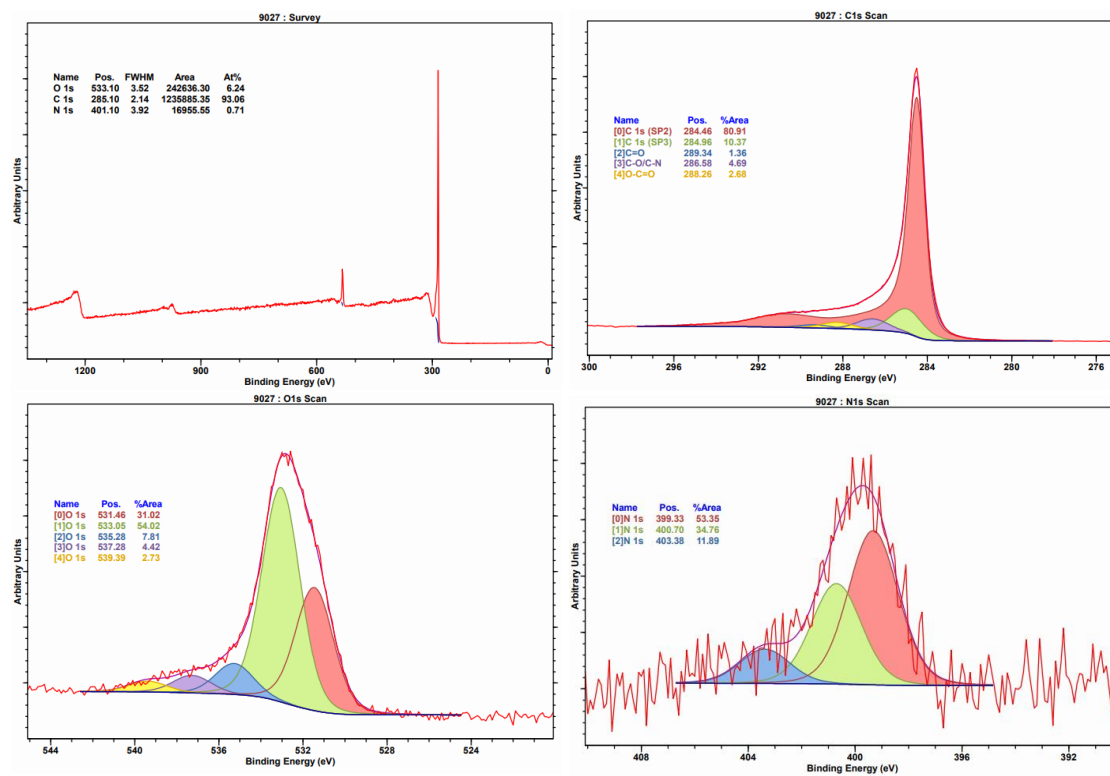


Figure S8: FLG N2.

FLG O₂:

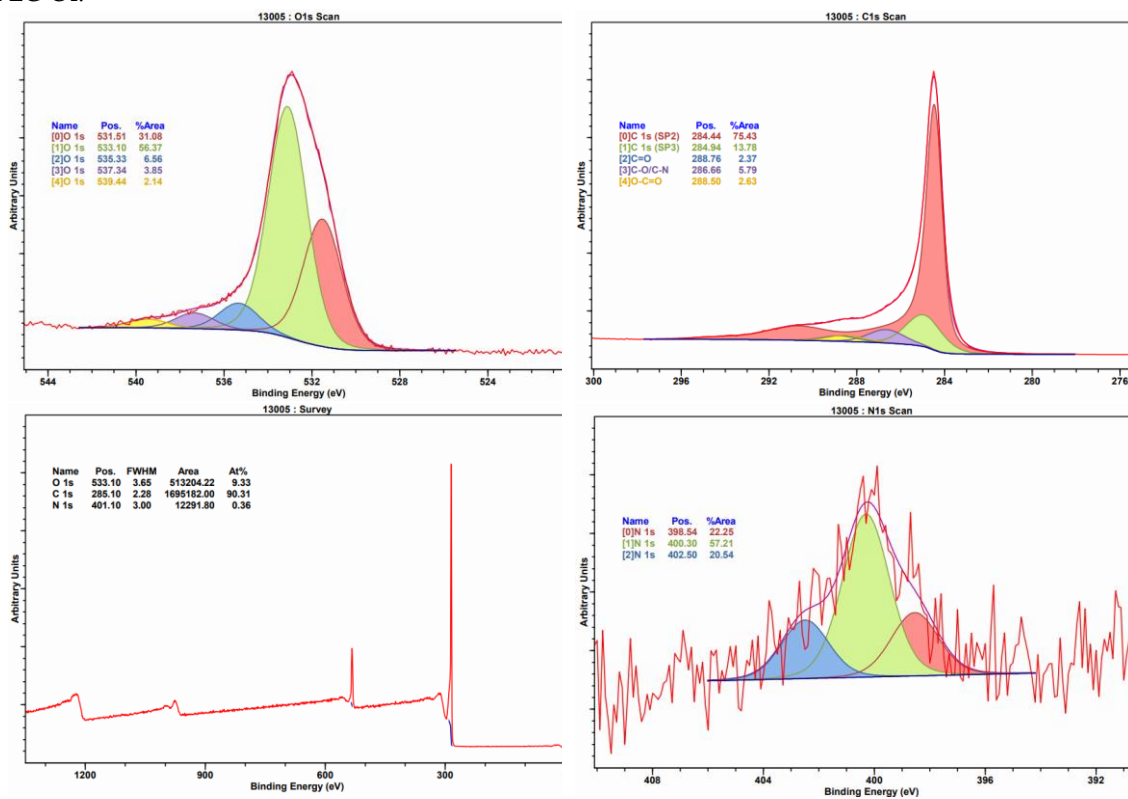
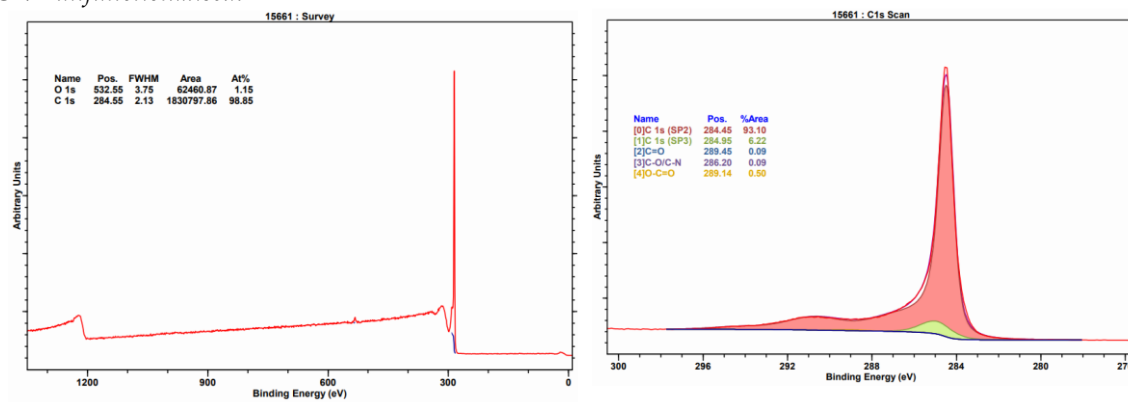


Figure S9: FLG O₂.

CNT unfunctionalised:



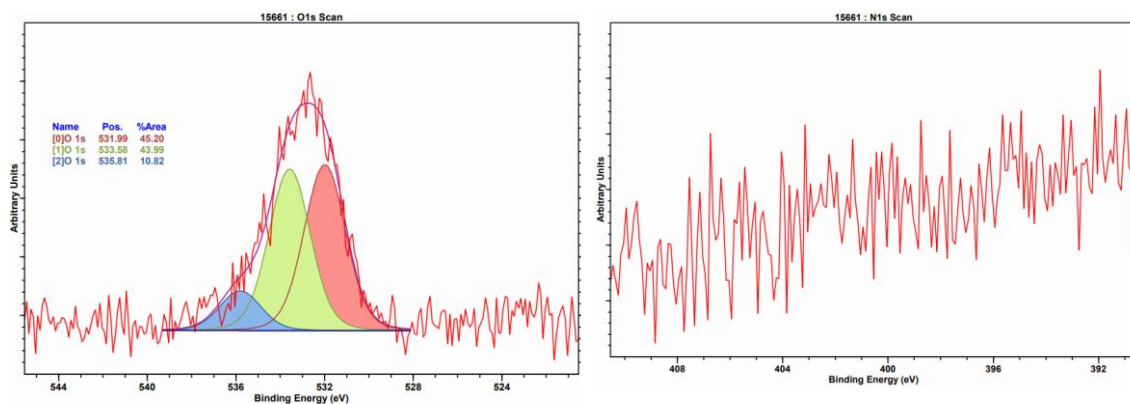


Figure S10: CNT unfunctionalised.

CNT NH₃:

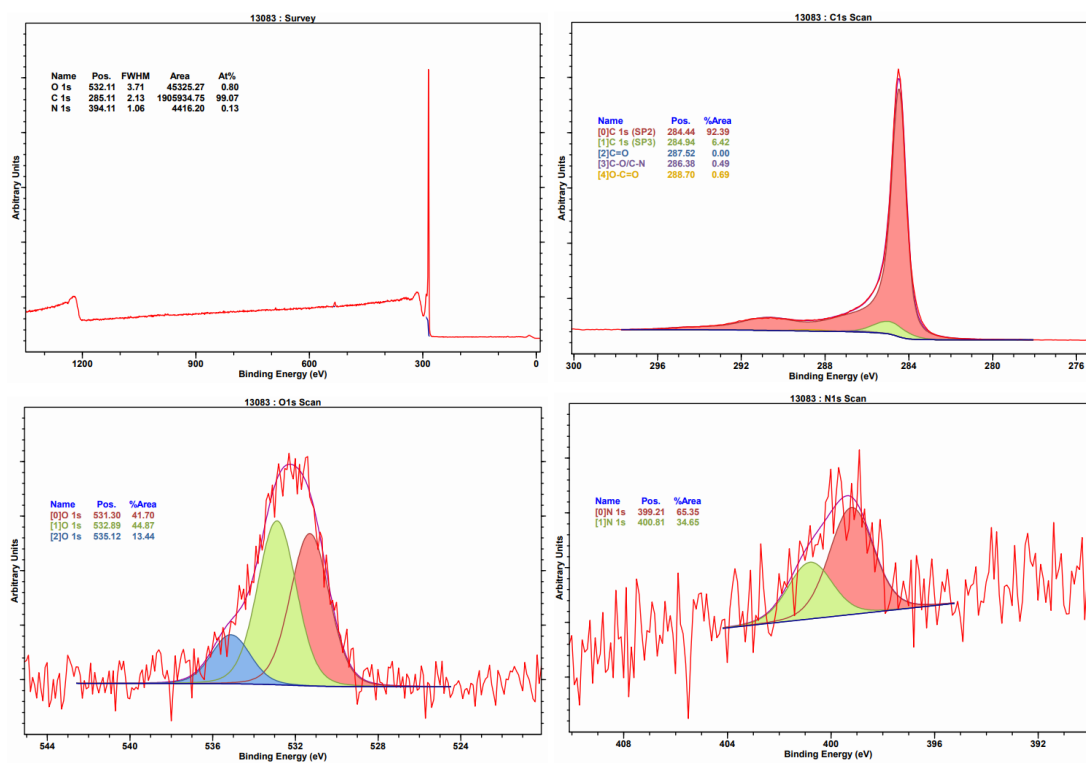


Figure S11: CNT NH₃.

CNT N₂:

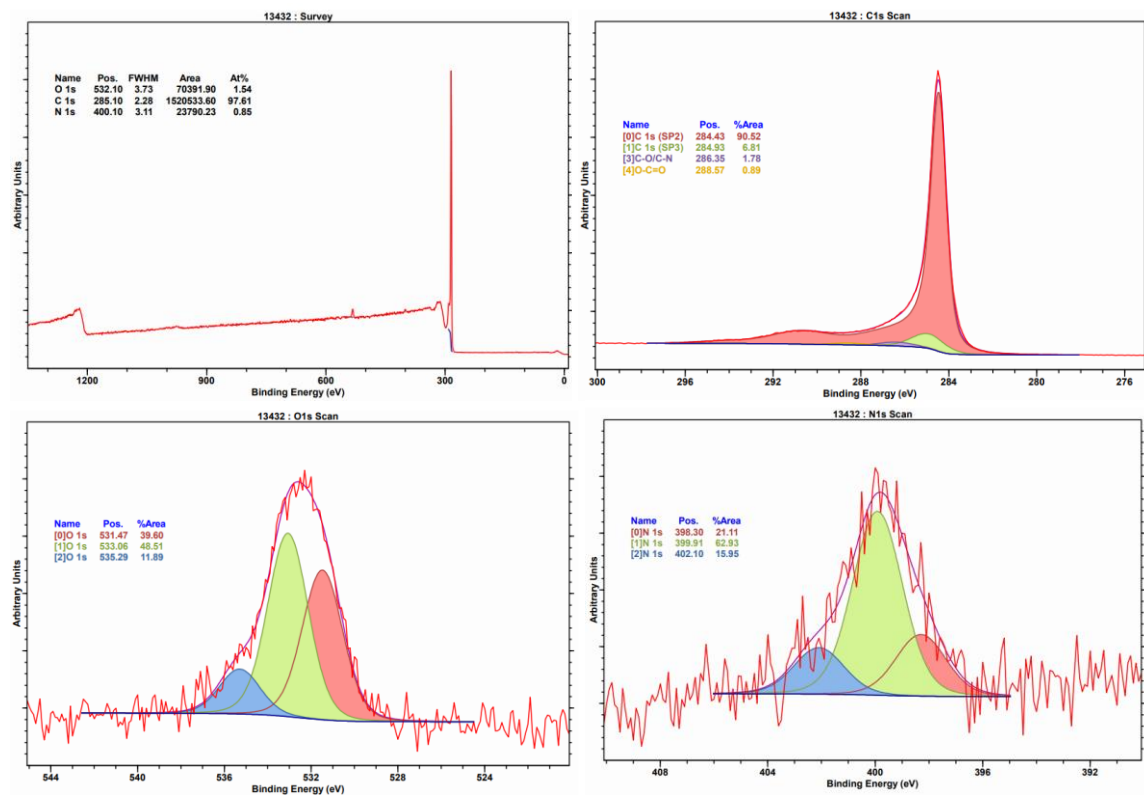


Figure S12: CNT N₂.

CNT O₂:

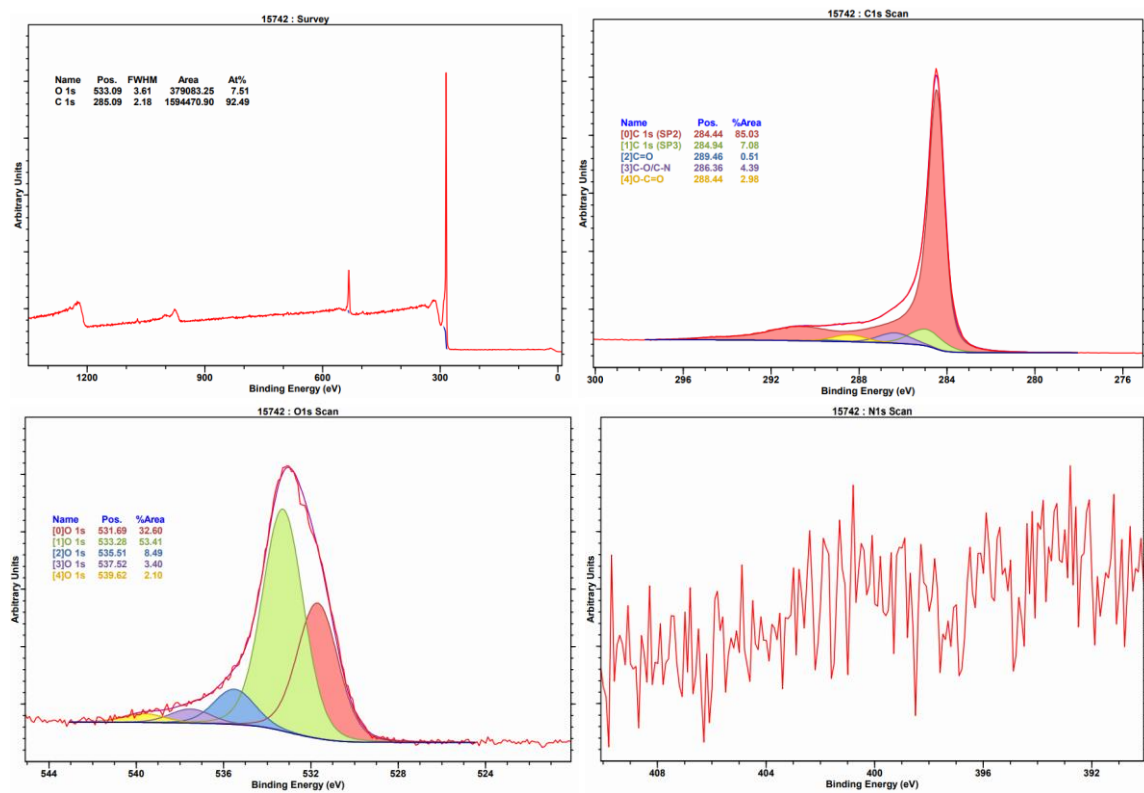


Figure S13: CNT O₂.