

## Materials and Methods

### Preparation of CVD Graphene Film.

Graphene was synthesised by the chemical vapour deposition (CVD) method on a highly pure copper foil (Alfa Aesar, 99.95 %) according to the previous work.<sup>13</sup> The growth was performed with flowing 40 sccm H<sub>2</sub> and 5 sccm CH<sub>4</sub> gas at 1000 °C. After coating a poly methyl methacrylate (PMMA) layer on a single side of as-grown graphene on Cu foil, the uncoated graphene was removed by oxygen plasma. The Cu foil was subsequently etched in ammonium persulfate (APS) solution (20 g/ 1L) for 5 hours. The PMMA/Gr layer was then transferred onto SiO<sub>2</sub> substrate, followed by baking at 80 °C for 24 hours to improve the adhesion. Finally, the PMMA layer was removed by acetone.

### Characterisation.

Optical microscopy (OM) images were captured by a NIKON ECLIPSE LV 100 ND OM installed with NIS Elements D 4.20.00 software. A RENISHAW Raman spectrometer was used to characterise the quality and uniformity of the prepared graphene films. The Raman spectra were obtained using an Ar laser (514 nm) with a spot size of 1 mm. The samples were analysed by high resolution – transmission electron microscope (HR-TEM, JEM-3010, JEOL Ltd.) and Cs corrected STEM with Cold FEG (JEM-ARM200F, JEOL Ltd.) and the images were collected by Gatan Digital Camera (MSC-794) coupled to the microscope. XPS spectra for Gr/SiO<sub>2</sub> were obtained by K-Alpha XPS system (Thermo-Scientific) with an X-ray mono-chromator (Al K $\alpha$  Micro-focused depth profiling EX06 Ion Source) with a spot size of 30 – 400  $\mu$ m.

### DCFH-DA Fluorescence Measurements.

The generation of secondary reactive oxygen species (ROS) was measured with the fluorescence of 2',7'-dichlorofluorescein diacetate (DCFH-DA) probe (Sigma Aldrich, Ex/Em: 495 nm/529 nm, 2  $\mu$ M).<sup>16</sup> The measurements for all test groups were performed after designated amounts of incubation periods with 5 mL of total volume.

### Ion-exchange HPLC Analysis.

For the analysis of oxidative destruction of phenols, a hydrogen column (Aminex 87H column, 300x10mm, Bio-Rad, USA) ion-exchange high performance liquid chromatography (HPLC, Dionex Ultimate3000) was utilised. Total injection volume was set to 10  $\mu$ L in all test samples with 0.01 N  $\text{H}_2\text{SO}_4$  eluent and the flow rate of 0.5 mL/min. Time-resolved HPLC analyses were performed over 90 minutes of injection time, and the intermediate structures were identified with the RI detector (ERC, RefractoMAX520, Japan). The identification of produced organic acid was carried out by the organic acid analysis coupled to the HPLC, based on the reference peaks and the retention times.

### **pH and Temperature Measurements.**

During graphene-based phenol degradation process, the pH and temperature changes were monitored by a pH meter equipped with a digital thermometer (Trans instruments, BP3001). The changes were measured 0, 1, 3, 6, 12, 24 hours post-incubation. The values were recorded after 5 minutes of each time point for stabilisation.

### **X-ray Photoelectron Spectroscopy Analysis**

XPS spectra for Gr/SiO<sub>2</sub> were obtained by K-Alpha XPS system (Thermo-Scientific) with an X-ray mono-chromator (Al K $\alpha$  Micro-focused depth profiling EX06 Ion Source) with a spot size of 30 – 400  $\mu$ m.