Supporting Information

2D Fluorinated Graphene Oxide (FGO)-Polyethyleneimine (PEI) Based 3D Porous Nanoplatform for Effective Removal of Forever Toxic Chemicals, Pharmaceutical Toxin and Waterborne Pathogens from Environmental Water Samples

Avijit Pramanik[1], Olorunsola Praise Kolawole[1], Kaelin Gates[1], Sanchita Kundu[1], Manoj K. Shukla[2], Robert D Moser[2], Mine Ucak-Astarlioglu[2], Ahmed Al-Ostaz[3], Paresh Chandra Ray[1] *

[1]Department of Chemistry and Biochemistry, Jackson State University, Jackson, MS, USA; E-mail: paresh.c.ray@jsums.edu; Fax: +16019793674

[2]US Army Engineer Research and Development Center, 3909 Halls Ferry Road, Vicksburg, MS.

[3]Department of Civil Engineering, University of Mississippi, University, Mississippi, 38677 United States

Methods

All the chemicals such as Polyethylenimine (PEI), 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide hydrochloride (EDC), N-hydroxyl succinimide (NHS), N,N-Dimethylformamide anhydrous (DMF), Hydrofluoric Acid (HF), potassium permanganate, graphite (flakes), Sodium nitrate, sulfuric acid were purchased from Sigma-Aldrich and were used without further purification. Perfluoro butanoic acid (PFBA) and perfluoro butane sulfonic acid (PFBS), tetracycline and moxifloxacin antibiotics were purchased from Sigma-Aldrich and were used without further purification. *Salmonella* and *Escherichia coli* waterborne pathogens were purchased from the American Type Culture Collection (ATCC, Rockville, MD).

Synthesis of graphene oxide from graphite

For this purpose, in the first step we have developed water-soluble graphene oxide from graphite using improved Hummer's method, as we and others have reported before[20-29, 34-49,47]. In detail, 0.501 g of graphite, 0.503 g of sodium nitrate, and 23 mL of concentrated sulfuric acid were mixed and stirred at room temperature for 30 minutes. Next, 3.0 g of potassium permanganate power was added to the mixture by 5 times (~30 min space for 1-3, ~0.5 g and ~60 min space for 4-5, ~0.7 g). When the 1st batch of permanganate was added, the solution turned to green. The next batch of permanganate was added after the green color disappeared. 3 mL of concentrated sulfuric acid was used to wash down the graphite on the inside of the beaker after each addition. Then 100 ml of deionized water was added to the above solution. Finally, H_2O_2 (50 %) was added to the suspension till there was no gas evolving (~ 3 mL). The mixture turned from almost black to yellow. Then, the yellow mixture was washed with 5% HCl followed by ethanol to remove the metal impurities. The yellow suspension was centrifuged at very high speed several times till the pH was near neutral. The final solution was dried by lyophilization.

Synthesis of fluorinated graphene

We have developed fluorinated graphene oxide from GO using HF [24-27,33]. In detail, 80 mg of Graphene Oxide (GO) was dispersed in 40 ml of double distilled water. Then 417 µl (48 wt%) of HF was added to the above solution. The solution mixture was autoclaved and maintained 180°C for 30 h. The resultant solution was naturally cooled to room temperature. Then the fluorinated graphene semisolid compound was obtained and filtered and followed by being washed with water to remove excess acid. The pure fluorinated graphene was finally freeze dried through lyophilization for future use.

Synthesis of two dimensional (2D) fluorinated graphene oxide and PEI based three dimensional (3D) porous nanoplatform.

We have developed 2D fluorinated graphene oxide and polyethyleneimine (PEI) based 3D porous nanoplatform (FGO-PEI). In details, at first, 40 mg of fluorinated graphene was

dispersed in 40 ml DMF and then the mixture was sonicated for 1h. Then under the nitrogen atmosphere, 250 µl of EDC (1mg/ml) and 150 µl of NHS (1mg/ml) were added to the DMF suspended fluorinated graphene solution. Then 400 mg of PEI was added to the resulting mixture and stirred it for 5 days at room temperature. Finally, the solid Fluorinated Graphene-PEI nanocomposite was obtained by vacuum drying.

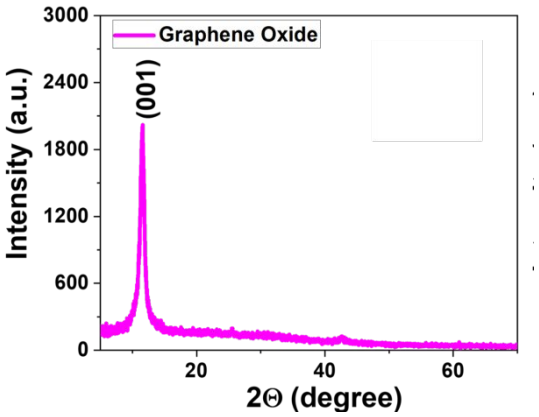

Figure S1: XRD data from GO shows the presence of (001) reflection peaks.

Determining perfluoro butanoic acid (PFBA) and perfluoro butane sulfonic acid (PFBS) removal amount using LC-MS

For the determination of the removal amount for perfluoro butanoic acid (PFBA) and perfluoro butane sulfonic acid (PFBS) using two dimensional (2D) fluorinated graphene oxide and PEI based three dimensional (3D) porous nanoplatform, we have used LC-MS (Agilent technologies) [11-21]. For this purpose, we have used the X Bridge-C18 column (4.6mm×250mm) from Agilent Technologies [11-21]. For the analysis we have used negative ionization (ESI-) mode[11-21]. For processing the data, we have used a workstation.

Surface Enhanced Raman Spectroscopy (SERS) analysis for the determination of the concentration of tetracycline and moxifloxacin antibiotics from environmental sample

For the measurement of SERS, we have used confocal Raman system with laser excitation of 670 nm gold nanoparticle as plasmonic surface [27-30]. Experimental details have been reported before [27-30]. For Raman data collection, we have used 100× magnification, and a numerical aperture of 0.9. for our experiment [27-30]. For Raman data collection we have used 10-second acquisition time and 5-scan averaging, so that we could achieve a very good signal-to-noise ratio[27-30].

***Salmonella* and *Escherichia coli* pathogens sample preparation**

Different waterborne pathogens like *Salmonella* and *Escherichia coli* were cultured according to the ATCC protocol, as we have reported before [34-49,47]. Once different waterborne pathogens like *Salmonella* and *Escherichia coli* grew to 10^5 CFU/ml, we used them for separation experiment. For this purpose, from the stock solution of *Salmonella* and *Escherichia coli*, we have diluted to vary the concentration of MDR bacteria from 10^2 -10^5 CFU/mL. After that, 10^3 CFU/mL of *Salmonella* and *Escherichia coli* were mixed with environment samples. After that the mixture was gently shaken for over 40 min.

After that we used two dimensional (2D) fluorinated graphene oxide and PEI based three dimensional (3D) porous nanoplatform for effective removal of *Salmonella* and *Escherichia coli*.

Finding number of bacteria captured during filtration using colony counting technique.

At the end, the number of live bacteria was counted in the water sample with a colony counter after 24 h at 37 °C incubation, as we have reported before[34-49,47]. After that, a single colony of bacteria from the tryptic agar plate was inoculated into 10 ml of Tryptic Soy Broth for 12 hours. The colony number for each plate was counted with a colony counter (Bantex, Model 920 A) [34-49,47].