Supplementary material *for the article:*

A facile and green synthesis for reduced graphene oxide-metal/metal oxide nanocomposites and their applications in water purification

T. S. Sreeprasad[a], Shihabudheen M. Maliyekkal[b], K. P. Lisha[c] and T. Pradeep[d]*

DST Unit on Nanoscience, Department of Chemistry and Sophisticated Analytical Instrument Facility, Indian Institute of Technology Madras, Chennai - 600 036, India

[d]*Email: pradeep@iitm.ac.in; Fax: 91-44-2257-0545/ 0509*

Authors 'a' and 'b' have contributed equally.

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Supplementary data S1. Detailed procedure for the oxidation of graphite to GO and preparation of RGO

a) Preoxidation of graphite

In a 250 mL beaker, concentrated H_2SO_4 (25 mL), 5 g of $K_2S_2O_8$ and 5 g of P_2O_5 were taken and heated to 90 °C with constant stirring. After all the reactants are completely dissolved, the reaction temperature was decreased to 80 °C. To this reaction mixture, 6 g of graphite powder was added slowly. Bubbling was observed initially and subsequently subsides over a period of 30 min. Temperature of the reaction mixture was maintained at 80 °C for 5 h. Subsequently, heating was stopped and the mixture was diluted with 1 L of distilled water and left undisturbed overnight. The resultant solution was then filtered and washed to remove excess acid. The solid product was dried in air overnight.

b) Oxidation to graphite oxide (GO)

230 mL of concentrated H_2SO_4 was maintained at 0 °C using an ice bath. Pre-oxidized graphite was then added to this and stirred. 15 g of $KMnO_4$ was added slowly making sure that the temperature never goes beyond 10 °C. Then, the temperature was raised to 35 °C and allowed to react for 2 h. Subsequently, 1 L of distilled water was added carefully keeping the temperature below 50 °C. The reaction mixture was again stirred for 2 more hours and then 1.5 L of distilled water and 25 mL of 30% H_2O_2 were added. The mixture was kept at room temperature for a day and the supernatant was decanted. The remaining solution was centrifuged and washed with 10% HCl followed by distilled water. This was repeated several times. The resultant solid was dried and 2 % (w/w) dispersion was prepared in distilled water. This dispersion was put for dialysis for 3 weeks to remove all unwanted contaminants like salts and acid. Then the dispersion was diluted to 0.1 % (w/w).

c) Chemical reduction of GO to graphene nanosheets

The GO reduction was carried out similar to a procedure reported by Li et al.[11] In a typical synthesis procedure, GO was sonicated for 45 min. (CREST TRU-SWEEP 27D, 50 Hz) to exfoliate the suspension completely and centrifuged at 5000 rpm to remove the unexfoliated GO. About 8.1 mL of the purified exfoliated GO solution was diluted to 90 mL using high pure distilled water. Into this solution, 45 µL of hydrazine solution (35 wt% in water) was added and stirred for one min. To the above mixture, 315 µL of ammonia solution (28 wt% in water) was added and the whole solution was stirred well for five min. After three steps, the mixture was kept at 90 °C without stirring for 2 h. Then, the solution was put for dialysis against 0.5 % ammonia for 2 days to remove excess hydrazine which

may be present in the solution. Dialysis was repeated against distilled water for two weeks to remove all the residual contaminants that may be present in the reaction mixture. This sample will be referred to as purified reduced graphene oxide sheets (RGO).

Parameters	Influent water (Kotlapalli Panchayath Puttaparthi, Andhra Pradesh, India)
pH @ 25 ^{0}C	8.01
Fluoride	2.34
Total Alkalinity (as $CaCO_3$)	217
Total Hardness (as $CaCO_3$)	178
Conductivity @ 25 ^{0}C	565 μmhos cm^{-1}
Organic carbon	< 0.5
Total Dissolved Solids	384
Calcium (as Ca^{2+})	36
Magnesium (as Mg^{2+})	22
Sulphate (as SO_4^{2-})	25
Chloride (as Cl^{-1})	13
Phosphate (as PO_4^{-3})	<0.02
Iron (as Fe^{3+})	<0.001
Nitrate (as NO_3^-)	38.9
Turbidity (as NTU)	0.2 NTU
Silica (as SiO_2)	47.4
Manganese	<0.001

Figure S2. Table1: The water quality characteristics of the Hg(II) spiked groundwater collected from Kotlapalli, Puttaparthi, Andhra Pradesh, India.
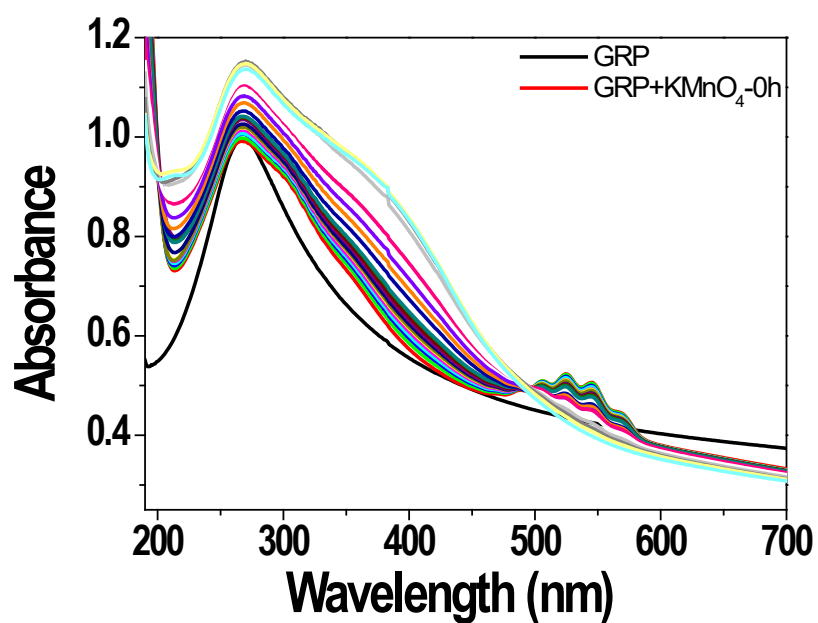
Figure S3. Time-dependent UV/Vis spectra of the reaction between RGO and KMnO$_4$.
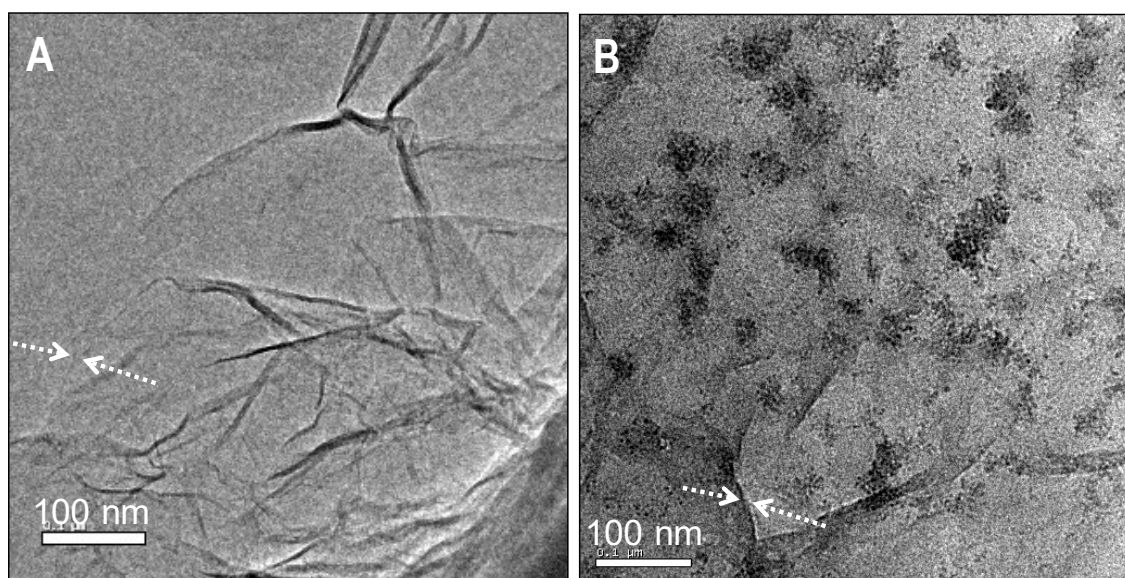
Figure S4. Large area TEM image of A) RGO and B) RGO-MnO$_2$ showing the characteristic wrinkles and edges of ~1 nm confirming the graphenic nature.
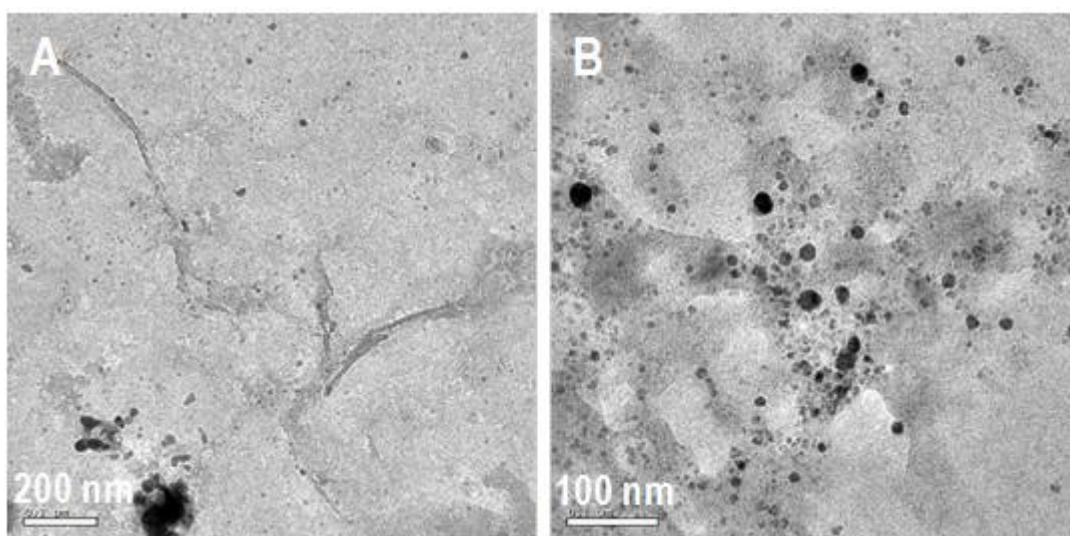
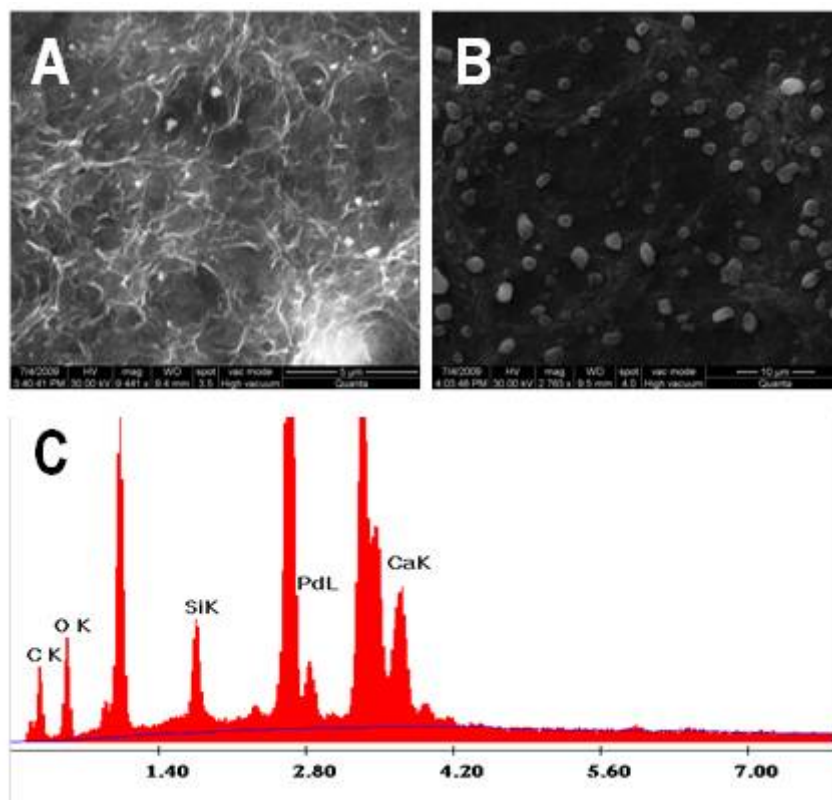Figure S5. Large area TEM Images showing the polydispersity found in GRP-Ag sample.

Figure S6. SEM images of GRP-Pd samples: A) 0.025 mM and B) 0.1 mM of PdCl$_2$. C) EDS spectrum taken from the sample containing 0.1 mM PdCl$_2$.
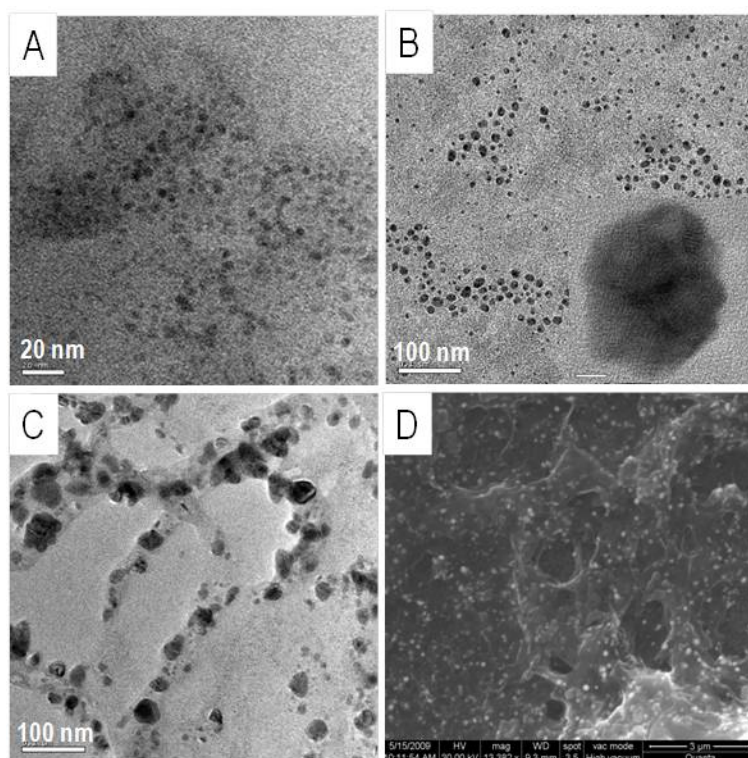
Figure S7. TEM images A) 0.01 mM B) 0.02 mM C) 0.07 mM GRP-Au sample. Inset of B shows a high magnification lattice resolved image of the 0.02 mM sample with slightly non-spherical structure. D) SEM image of an aggregated sample (0.1 mM), of GRP-Au.
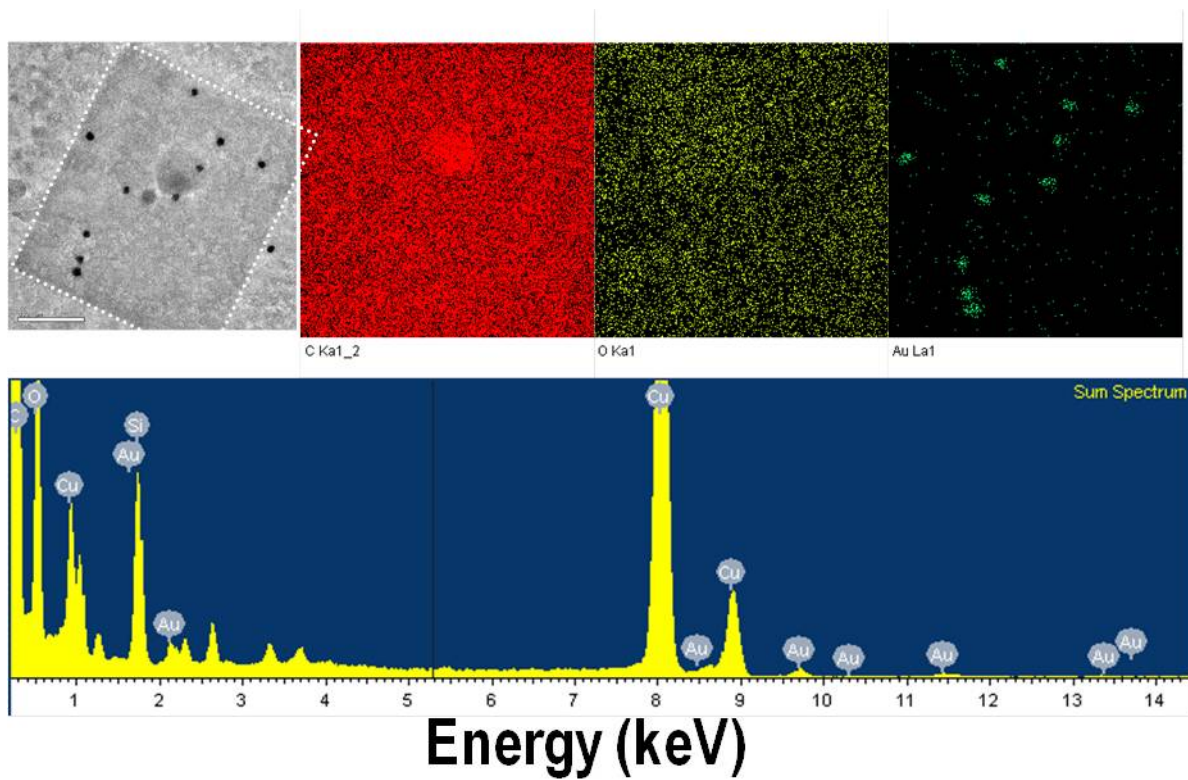
Figure S8. EDS spectrum and elemental maps taken from the GRP-Au sample containing 0.05 mM HAuCl$_4$. The EDs scan was at a different angle to the TEM camera. The scanned area is marked in the figure.
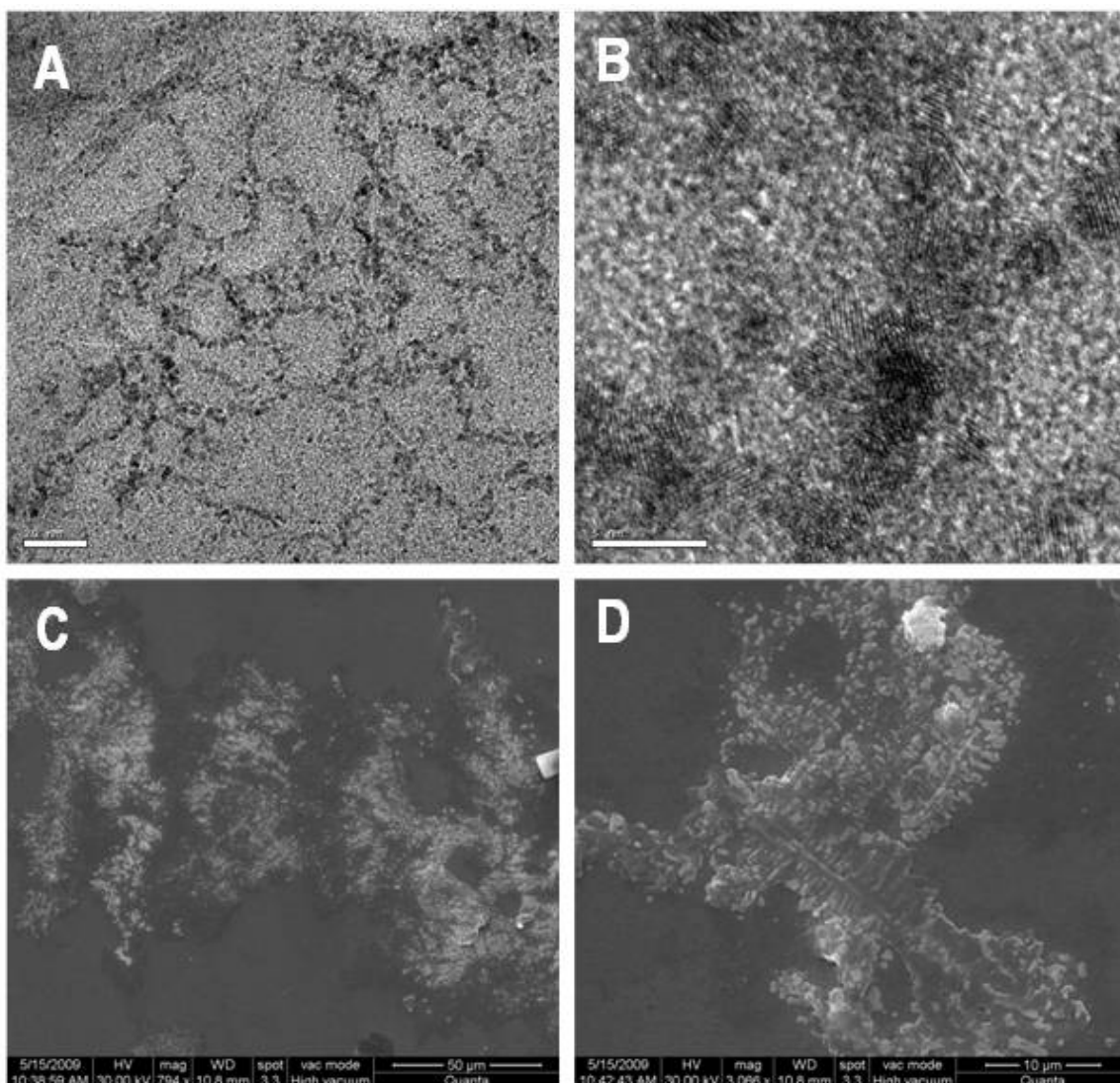
Figure S9. A) TEM image of the GRP-Pt sample containing 0.02 mM H$_2$PtCl$_4$. B) Lattice resolved image of the same sample showing the lattice structure of Pt nanoparticles. C) and D) SEM images taken from the aggregated sample of higher concentration (0.1 mM).
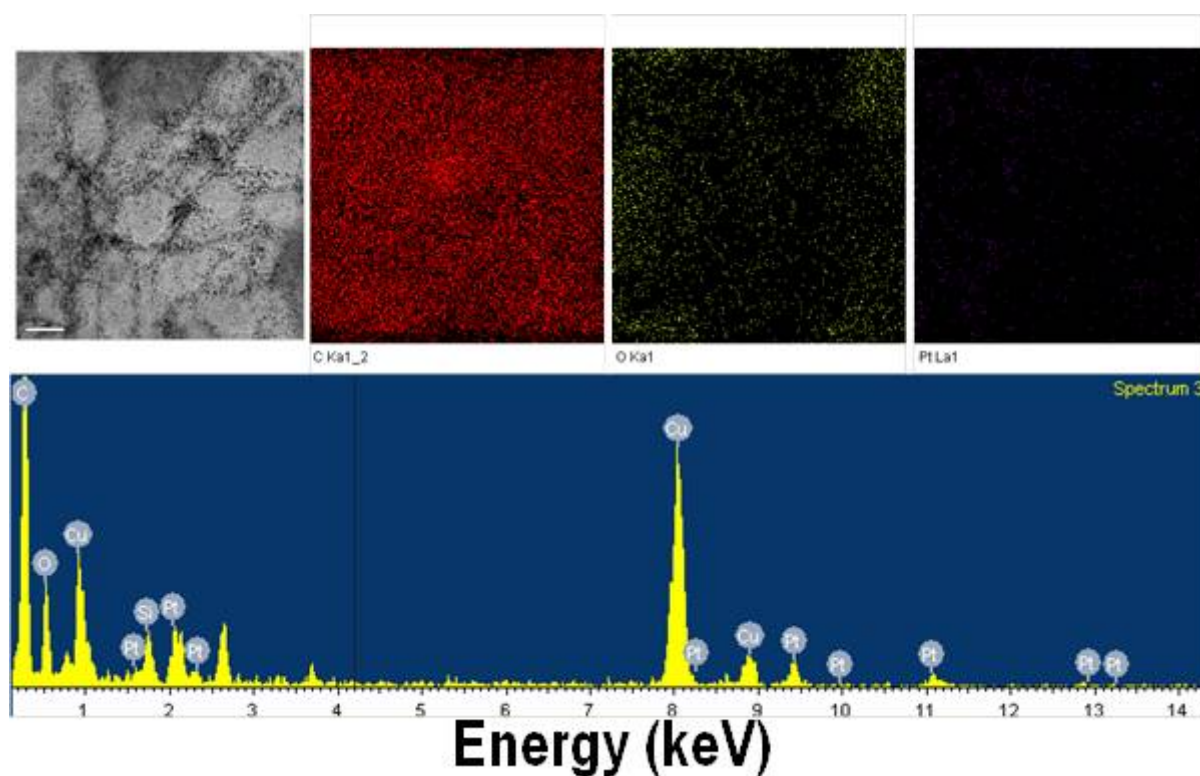
Figure S10. EDS spectrum and elemental maps taken from the GRP-Pt sample containing 0.02 mM H_2PtCl_4 showing the presence of Pt. The particle size being small, mapping did not reveal exact match, but a contour of the structure shown in the microscope can be seen in the Pt map also.
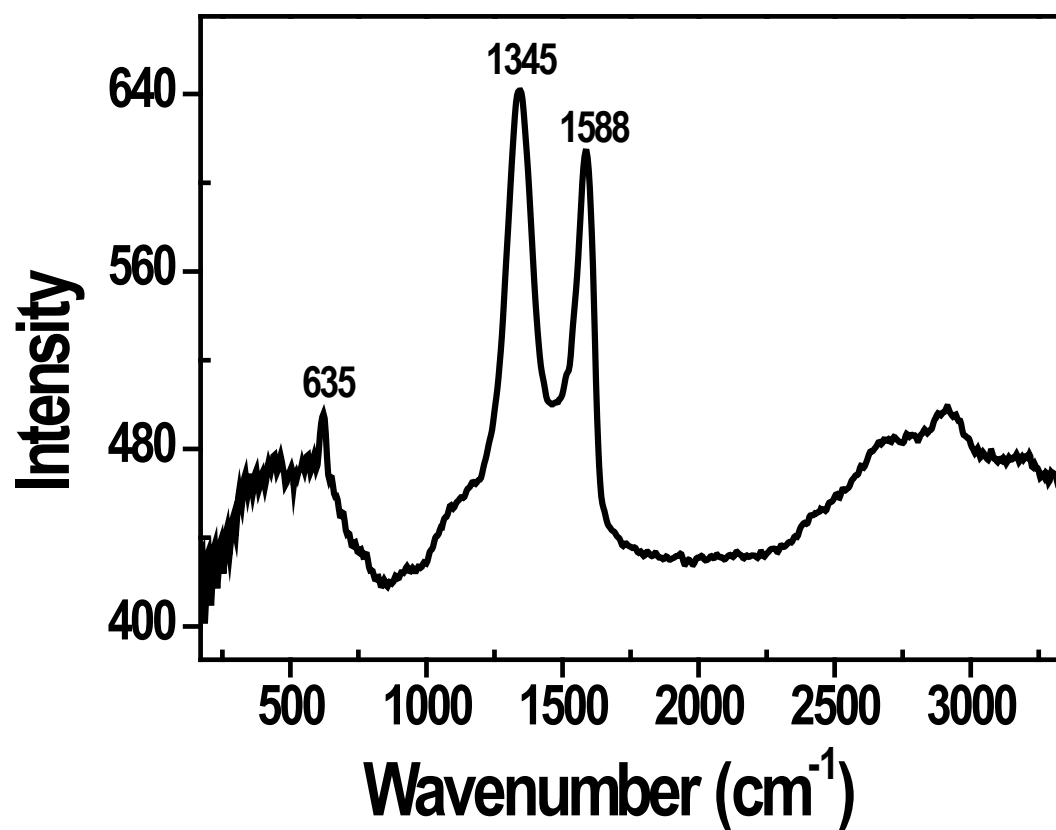
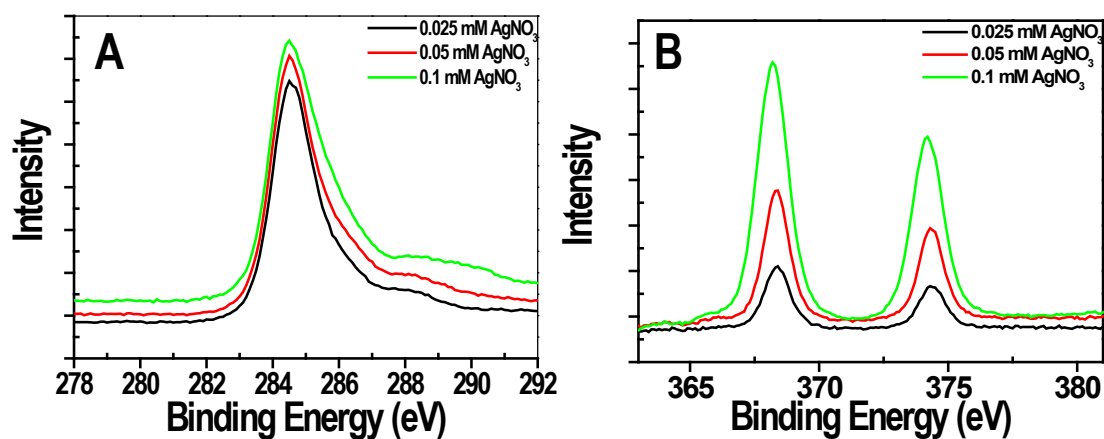Figure S11. Raman spectrum of GRP-MnO$_2$ sample showing the presence of MnO$_2$ in the composite.

Figure S12. XPS spectra of GRP-Ag composite. A) C 1s and B) Ag 3d. In the C 1s spectra, we can see that as the concentration of AgNO$_3$ increased, the extent of oxidation of carbon is also increased. Ag 3d$_{5/2}$ is at the metallic value of 368 eV.
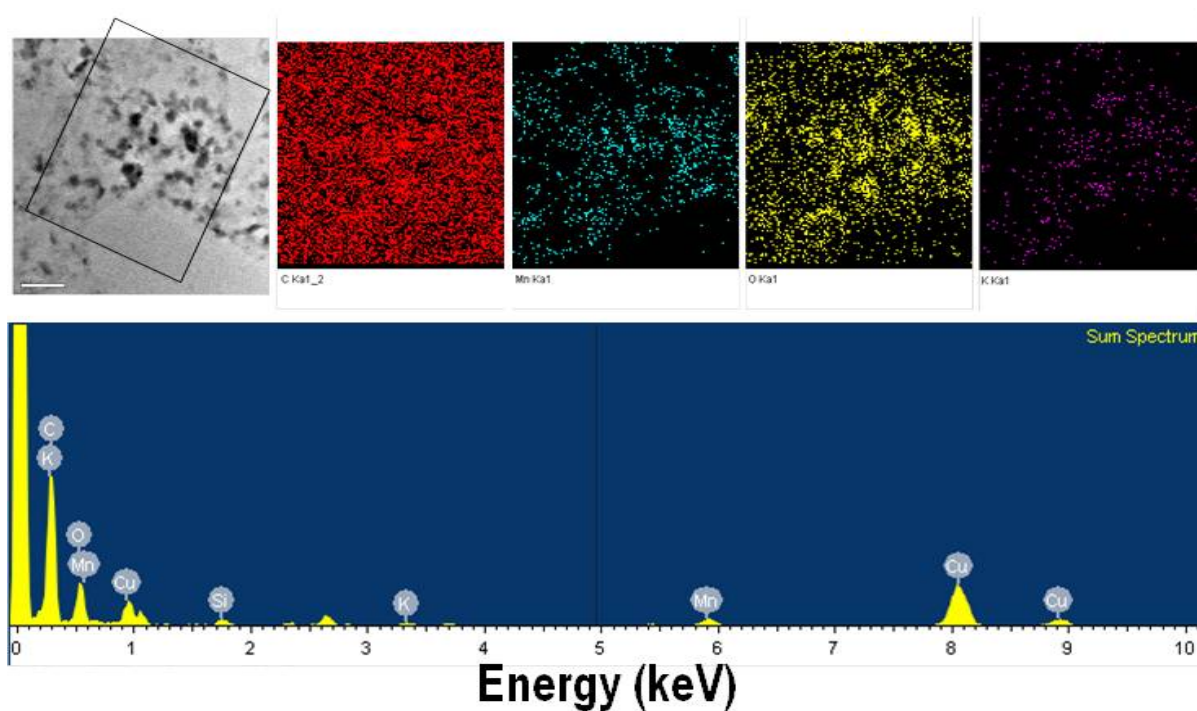
Figure S13. EDS spectrum and elemental maps taken from the GRP-MnO_2 sample containing 0.02 mM $KMnO_4$. The EDs scan was at a different angle to the TEM camera. The scanned area is marked in the figure.

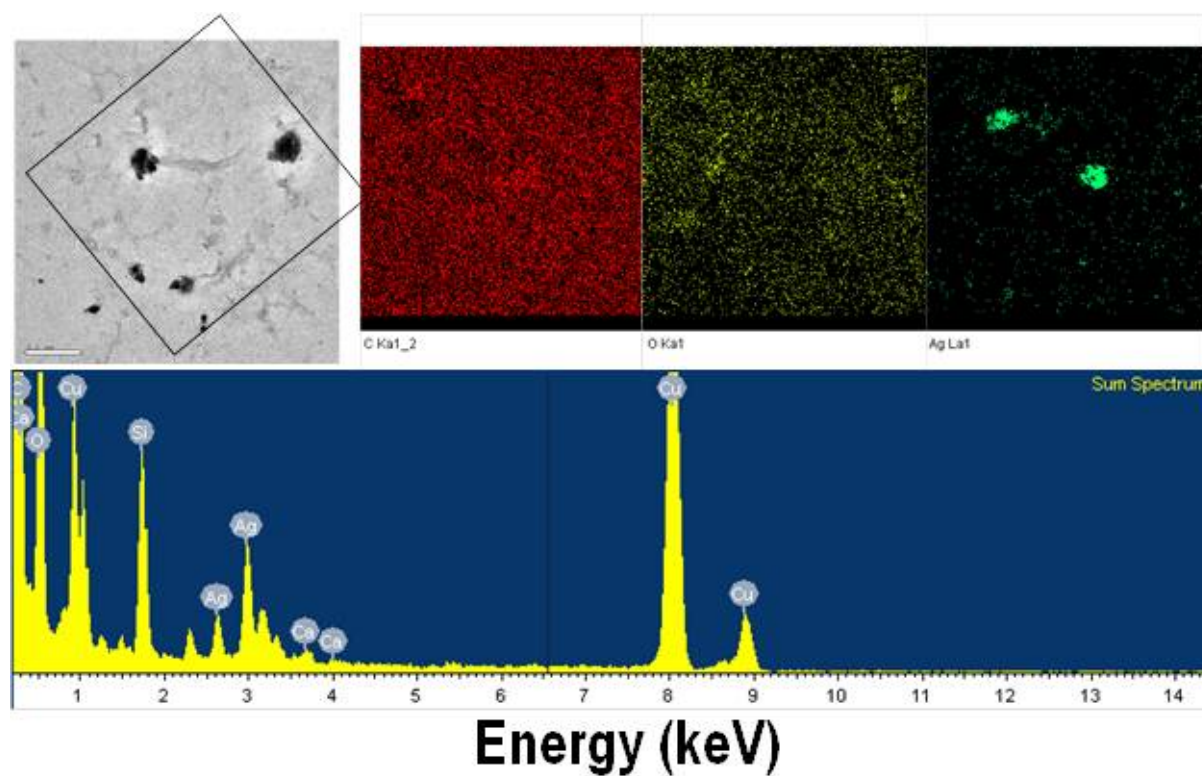Figure S14. EDS spectrum and elemental maps taken from the GRP-Ag sample containing 0.02 mM AgNO₃. The EDS scan was at a different angle to the TEM camera. The scanned area is marked in the figure.
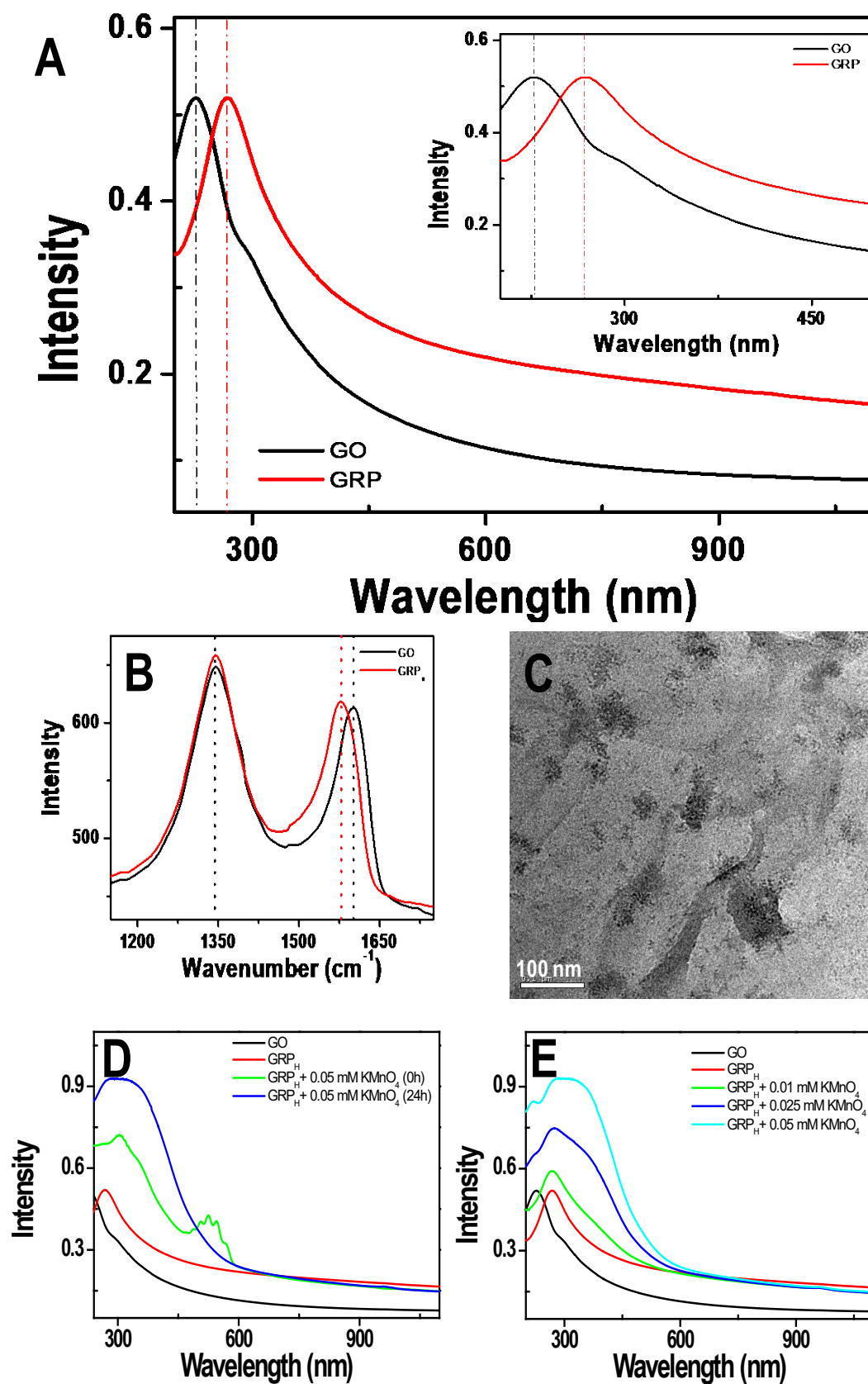
Figure S15. A) UV/ Vis spectra of GO and hydrothermally synthesized GRP (GRP$_H$). Inset the expanded view showing the shift of peak position. B) Raman spectra of GO and GRP$_H$ showing the shift in G-band position accompanying the reduction. C) Large area TEM image of GRPH-MnO2 (0.02 mM) composite and UV/Vis spectra of composites formed using GRP$_H$ D) time-dependent spectra and E) concentration dependent spectra
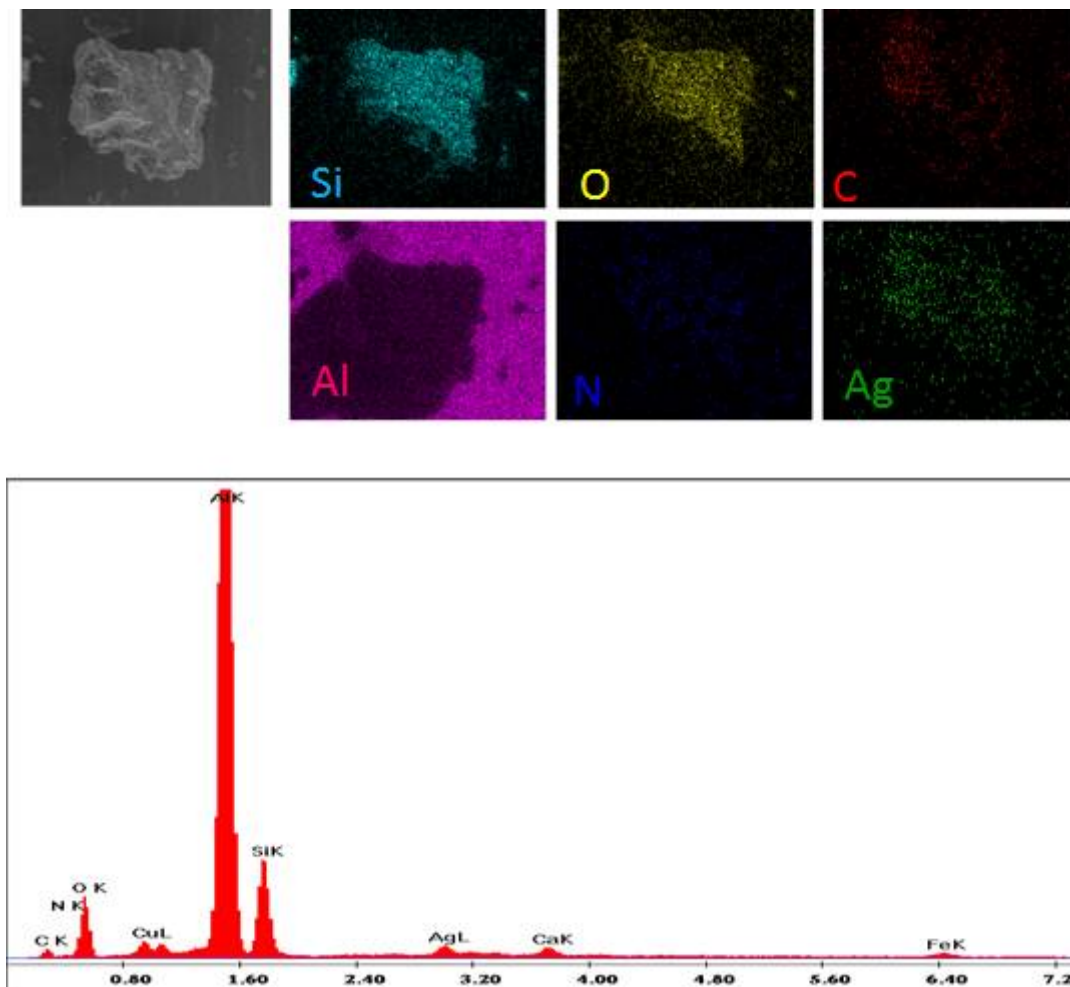

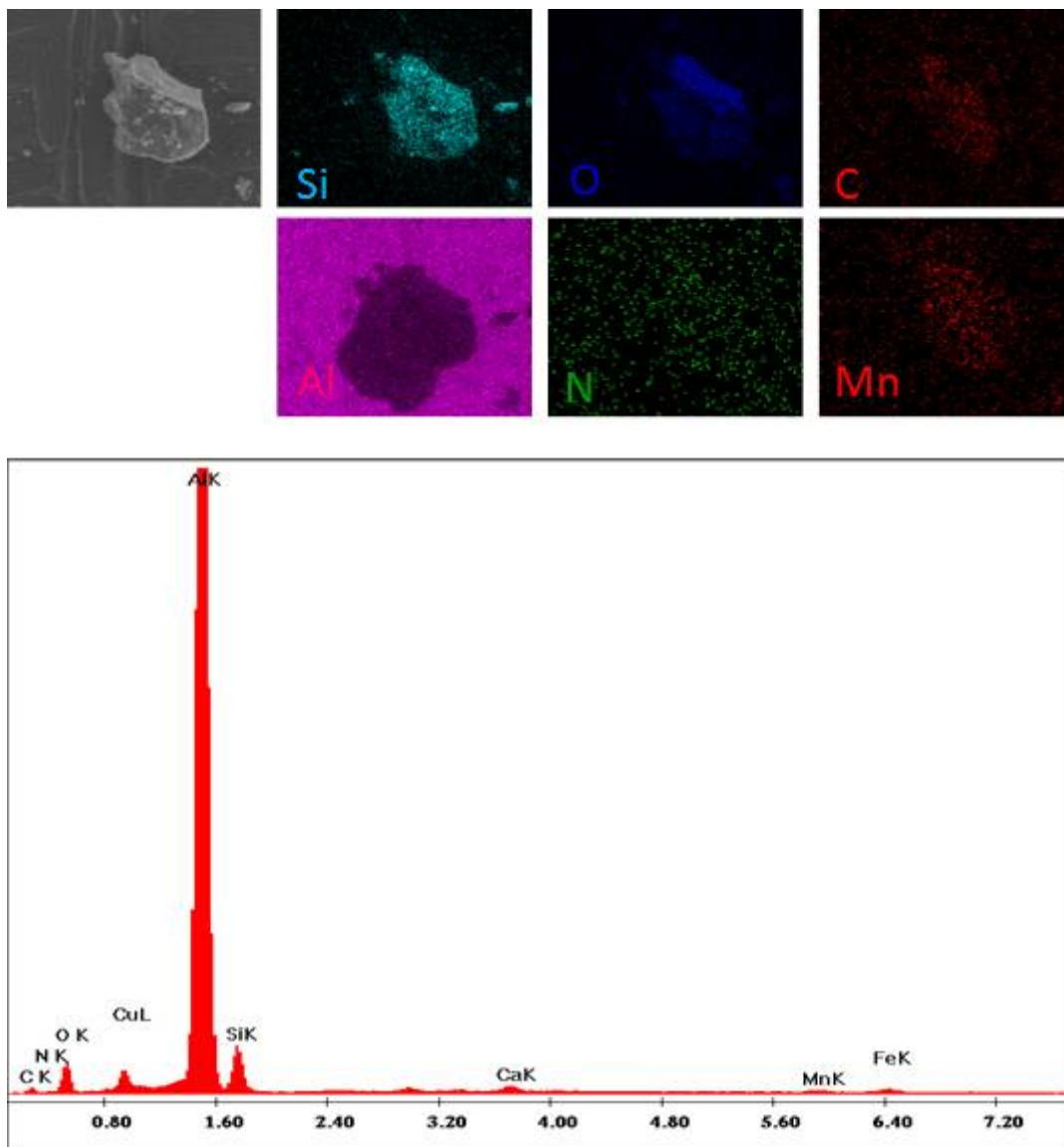Figure S16. EDAX analysis of Ch-RGO-Ag@RS composite.

Figure S17. EDAX analysis of Ch-RGO-MnO$_2$@RS composite.

Supplementary data S18. Mathematical expression for pseudo first order and pseudo second order reaction kinetic models.

Pseudo-first-order equation:
$$q_t = q_e\left(1 - e^{-k_1 t}\right) \qquad (1)$$

Pseudo-second-order equation:
$$q_t = \frac{q_e^2 k_2 t}{1 + q_e k_2 t} \qquad (2)$$

Where q_e and q_t are the adsorption capacity at equilibrium and at time t, respectively (mg g^{-1}). k_1 is the

rate constant of pseudo-first-order adsorption (1 min^{-1}) and k_2 is the rate constant of pseudo-second-

order adsorption (g mg^{-1} min^{-1}).

Adsorbents	Pseudo first-order-rate parameters				Pseudo-second-order rate parameters			
	k_1 (l min^{-1})	q_e (mg g^{-1})	RMSE	χ^2	k_2 (g mg^{-1}min $^{-1}$)	q_e (mg g^{-1})	RMSE	χ^2
GO	0.051	7.66	0.179	0.091	0.006	9.25	0.361	0.227
RGO	0.045	7.74	0.234	0.085	0.005	9.47	0.438	0.299
RGO-Mn	0.089	9.50	0.165	0.028	0.011	10.80	0.324	0.138
RGO-Ag	0.082	9.53	0.172	0.040	0.010	10.89	0.405	0.213
Ch-RGO-Mn@RS	0.13	0.04	0.0009	0.0002	4.98	0.04	0.001	0.0002
Ch-RGO-Mn@RS	0.12	0.04	0.001	0.0005	4.21	0.04	0.0009	0.0002

Figure S19. Table 2. Pseudo-first-order and pseudo-second-order rate parameters obtained for the adsorption of fluoride by various adsorbents.
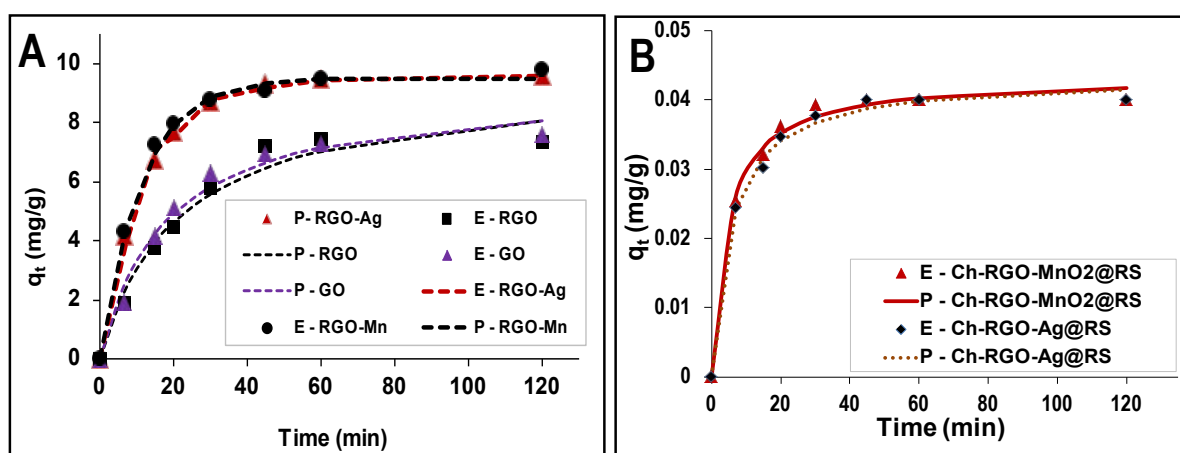

Figure S20. Pseudo-second-order kinetic plots with experimental data for adsorption of Hg(II) by GO, RGO and various RGO composites (E – experimental, P – predicted); A).unsupported and B) supported form.
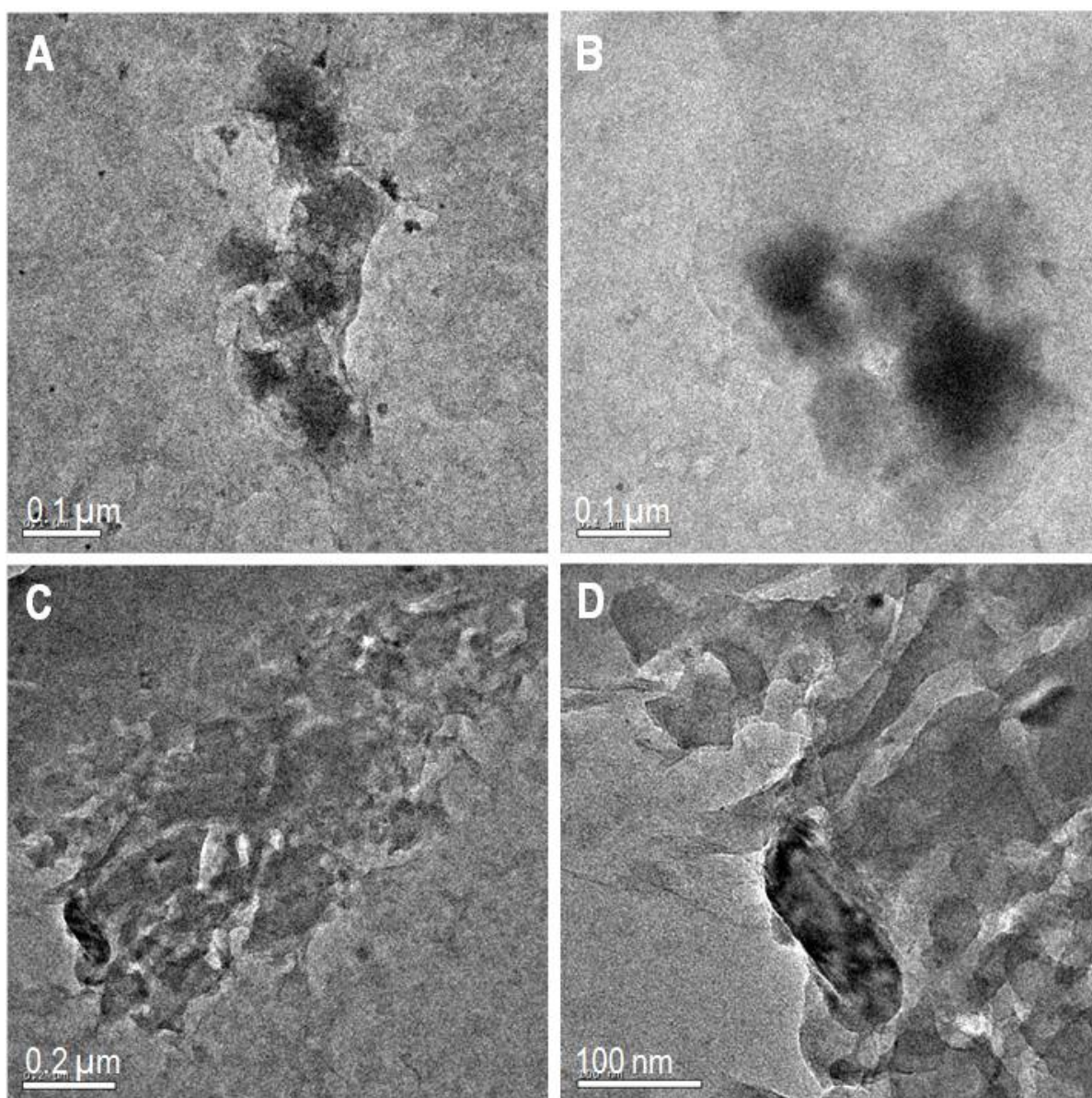
Figure S21. TEM images of GRP-MnO$_2$ composite (0.025 mM) after adsorption of Hg(II). We can see a large number of aggregated particles.
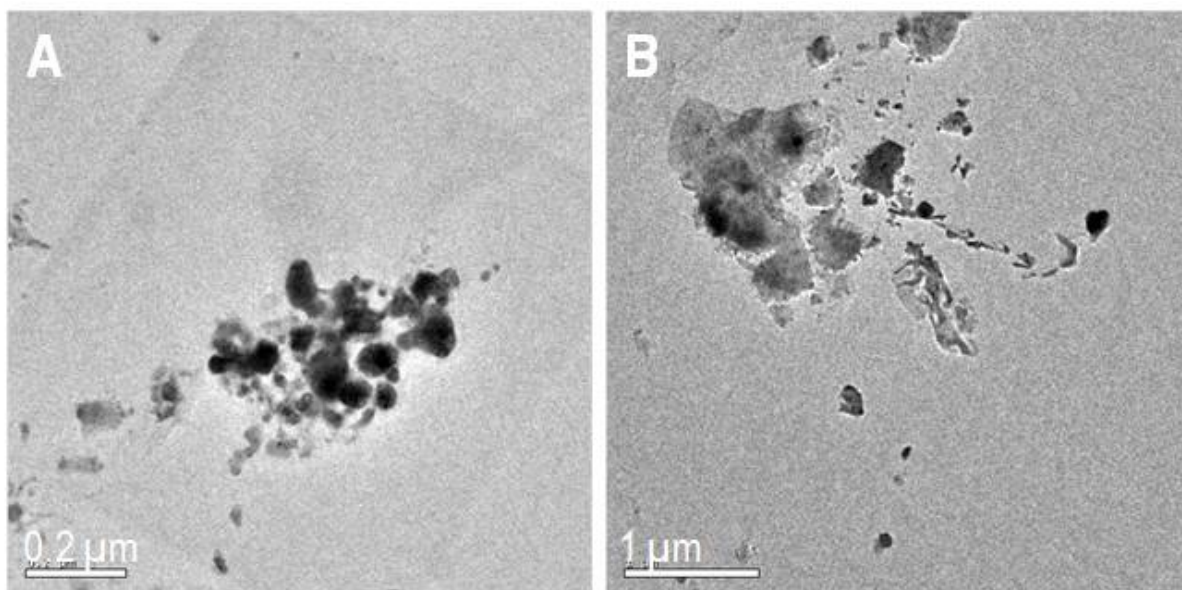
Figure S22. TEM images of GRP-Ag composite (0.025 mM) after adsorption of Hg(II). Nanoparticles are getting aggregated after Hg(II) adsorption. Inset