Electronic Supplementary Information

van der Waals pressure assisted synthesis of solid state nanomercury from mercury salts at ambient conditions: A sustainable approach for effortless Hg(II) removal from waste water and safe storage thereof

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1. Experimental section

1.1. Materials

Analytical grade reagents were used for the study and was used as-received.Ortho phenylenediamine was purchased from Thermo Scientific. Mercury (II) chloride was obtained from Hi Media Laboratories Pvt. Ltd. Deionized water was used to make all of the solutions.

1.2. Synthesis of NGOQDs

In a typical procedure, 0.2 g of ortho phenylenediamine was dissolved in 60 mL of water and the mixture was subjected to hydrothermal treatment at 200 °C for 6 h in a Teflon-lined autoclave. The insoluble portion was filtered off after cooling to ambient temperature and the NGOQDs were then recovered by centrifugation at 3500 rpm for 30 min, followed by dialysis. The NGOQDs was kept at 4 °C and used for future studies.

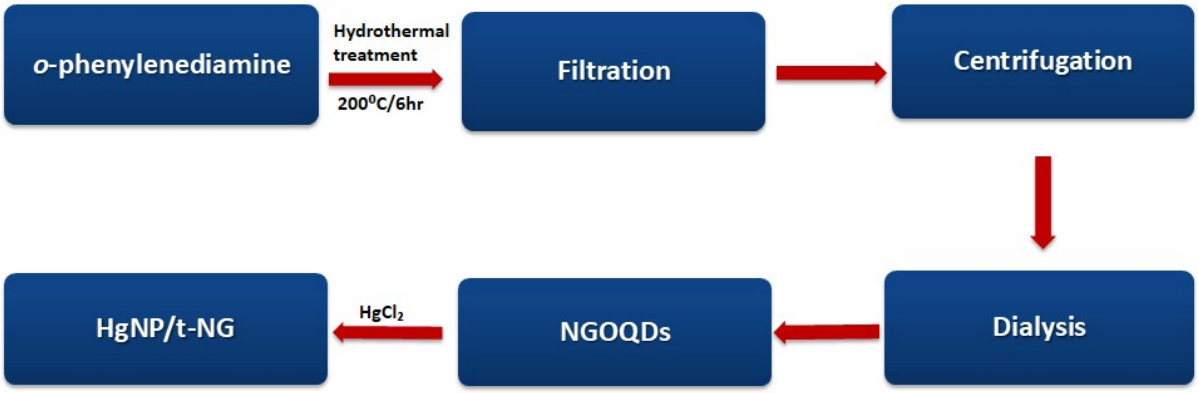
1.3. Fabrication of solid mercury nanoparticles supported on nitrogen doped turbostratic graphene (t-NG) matrix

A rather simple procedure was adopted for the preparation of mercury nanoparticles, which involved the addition of 2 mL of mercury(II) chloride aqueous solution (1 mM) to 10 mL of NGOQDs aqueous dispersion (1.9 mg/mL). Orange coloured turbid solution initially formed is turned into orange precipitate within ~ 20 minutes. The solid was separated from the solution, washed thoroughly with deionised water, and dried at room temperature to yield HgNP/t-NG composite. The flowchart of the synthesis path to t-NG framework supported mercury nanoparticles is given in this supplementary information (Scheme S1, ESI†).

2. Characterisations

The fluorescence of the material was monitored using LZC-4X photoreactor. The UV-vis absorption spectra were measured with a JASCO V-550 spectrometer. The fluorescence spectrometer Cary Eclipse (Agilent Technology) was used to record the fluorescence spectra of NGOQD solution. HORIBA time-correlated single-photon-counting (TCSPC) with 370 nm wavelength excitation light source was used to measure fluorescence lifetime decay. The elemental analysis of the samples was examined by EDX supported in the FE-SEM SIGMA 300. The morphology and particle size were analyzed by JEOL JEM 2100 TEM running at an accelerating voltage of 200 kV, with minimum exposure time, taking care to reduce the electron beam impact on the sample . XRD of the systems were recorded using a Rigaku Miniflex-II diffractometer with Cu K_α radiation in the scan range of 2θ 5-90°. JASCO FTIR-4100 spectrometer was used to obtain the Fourier transmission infrared (FTIR) spectrum using the

KBr pellet technique. Raman spectra were recorded using WITec alpha 300RRA (WITec GmbH, Ulm, Germany) having 532 nm DPSS laser. Omicron spectrometer was used to perform XPS with Mg K_α radiation having an energy of 1253 eV.



Scheme S1. Flowchart illustrating the synthesis of t-NG framework supported mercury nanoparticles.

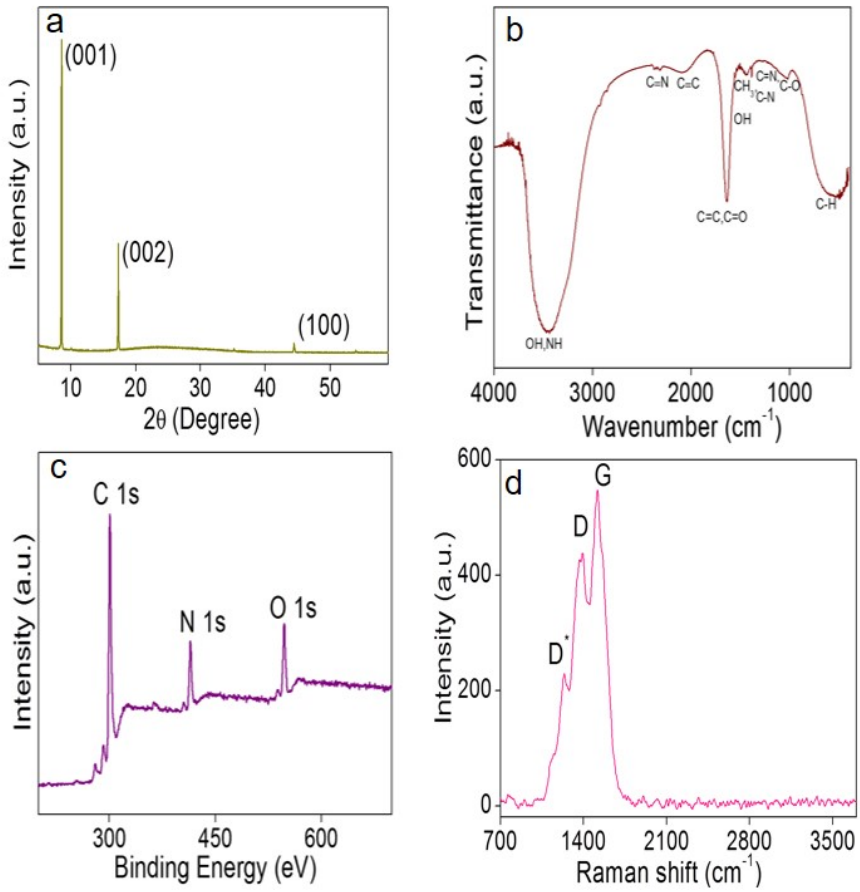


Figure S1. **a)** XRD pattern of NGOQDs. **b)** FTIR spectrum of NGOQDs. **c)** XPS spectra of NGOQDs. **d)** Raman spectrum of NGOQDs.

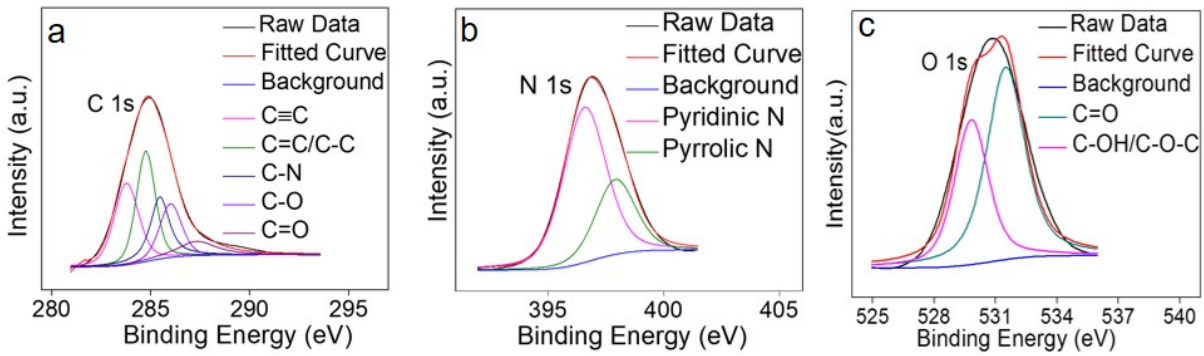


Figure S2. Deconvoluted **a)** C 1s **b)** N 1s **c)** O 1s XPS spectra of NGOQDs.

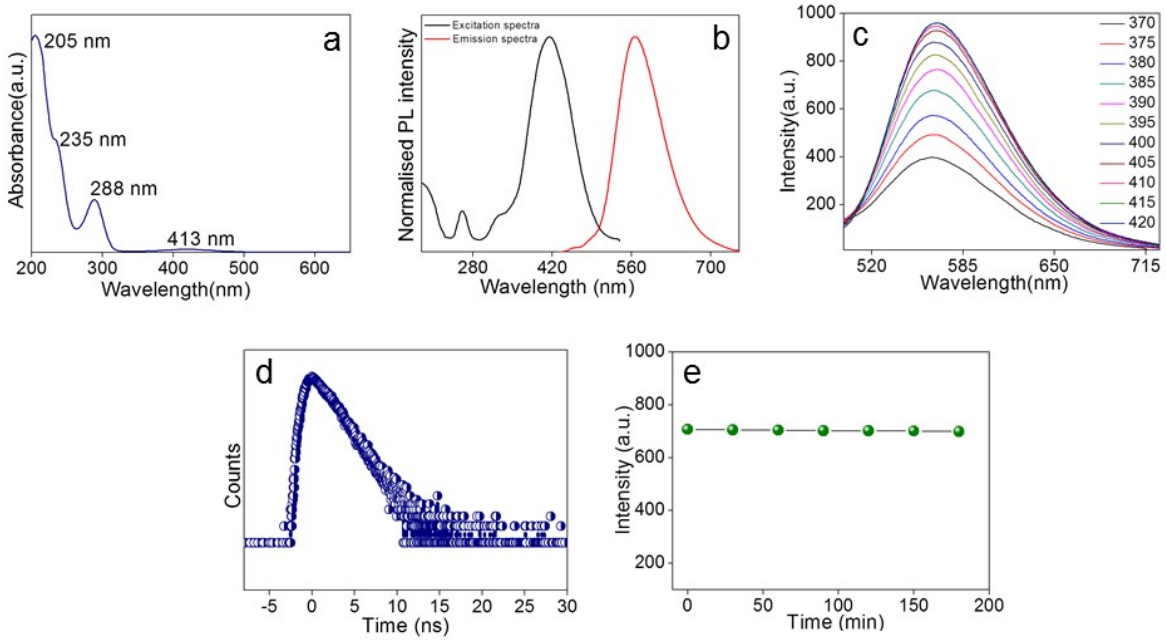


Figure S3 **a)** UV-vis absorption spectrum of the particles in aqueous solution. **b)** Excitation and emission spectrum of NGOQDs. **c)** Excitation independent emission spectra of NGOQDs**. d)** PL decay curve at 370 nm excitation and emission monitored at 562 nm. **e)** Time-dependent luminescence intensity of NGOQDs in aqueous dispersion.

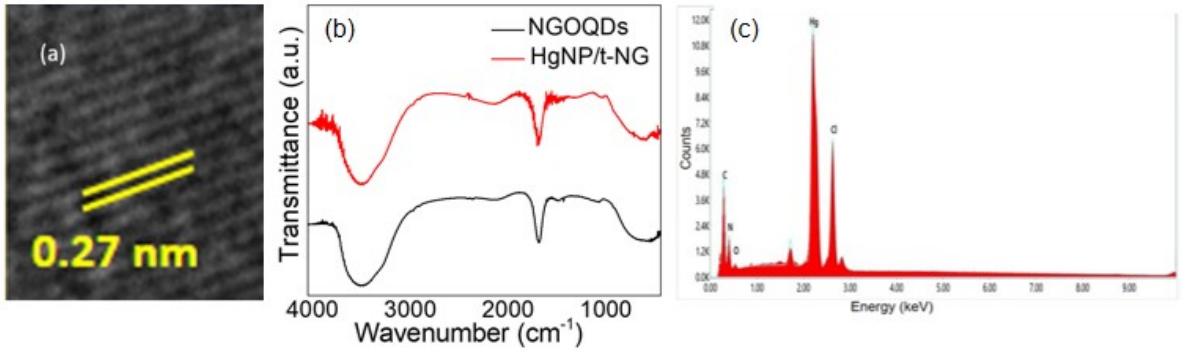


Figure S4. **a)** Lattice fringes corresponding to solid mercury. **b)** Comparison of normalised FTIR spectra of NGOQDs and HgNP/t-NG. **c)** EDX analysis of HgNPs on t-NG matrix.

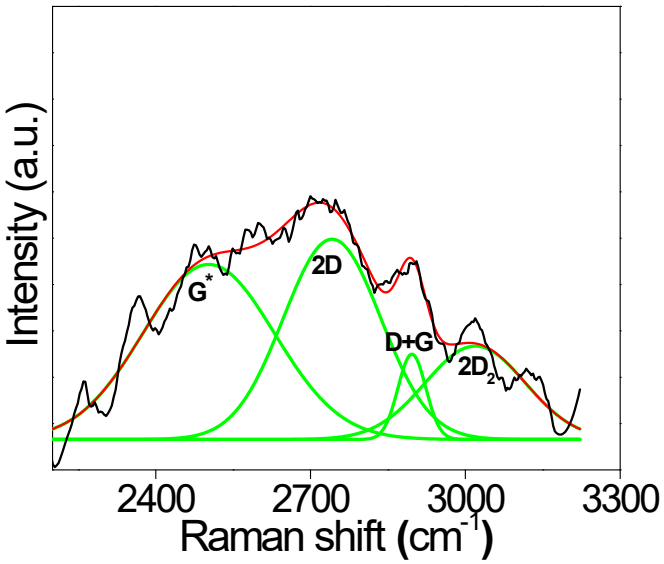


Figure S5. Curve fitted second order Raman bands

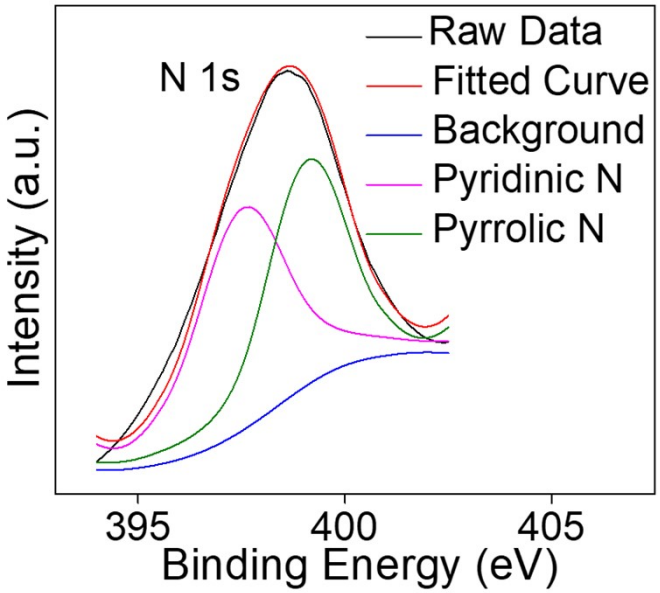


Figure S6. Deconvoluted N 1s XPS spectra of HgNP/t-NG

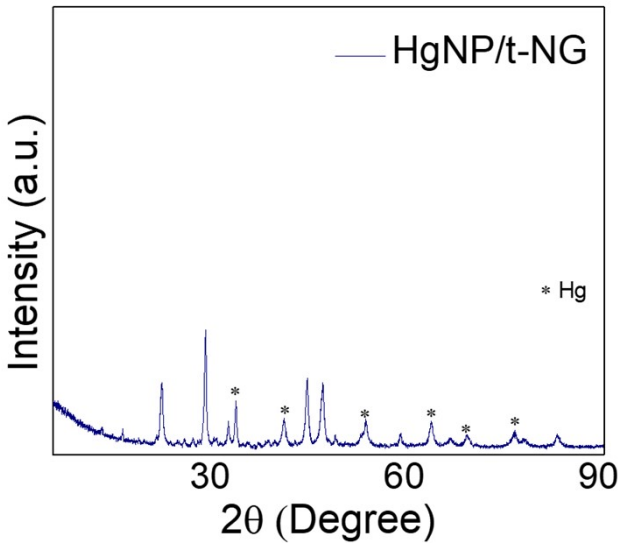


Figure S7. XRD pattern of HgNP/t-NG after 6 months, displaying peaks characteristics of solid mercury, confirming long term stability of the system.

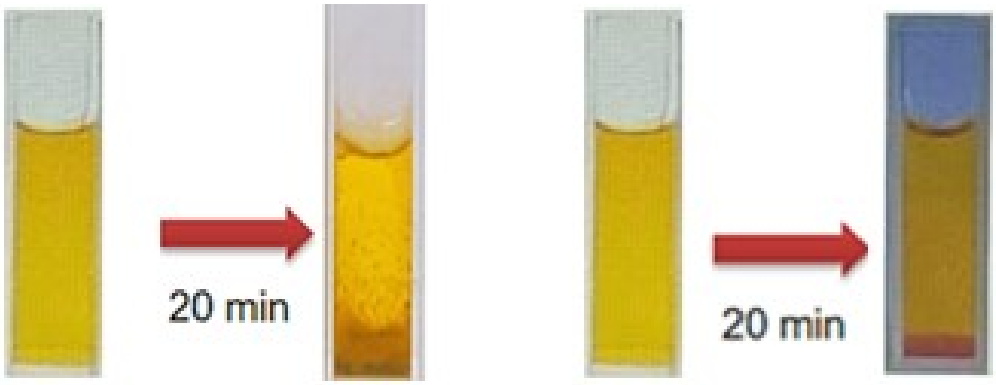


Figure S8. Removal of mercury ions from aqueous solution as solid separation by forming solid state nanomercury anchored on t-NG matrix, upon adding various mercury salts to NGOQDs solution. Hg $(NO_3)_2$ (left) and $Hg(OAc)_2$ (right).

Figure S9. Real water sample analysis with tap water (left) and well water (right) spiked with mercury. Addition of NGOQDs solution induces solid (HgNP/t-NG) separation in both the solutions in presence of mercury ions