<div align="center">**Supplementary Information**</div>

Magnetite decorated graphite nanoplatelets as cost effective CO_2 adsorbent

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1. Results and Discussion

1.1 FTIR study of *f*-GNP

FTIR study was performed to confirm the attachment of functional groups at the surface of GNP by acid treatment. Figure-S1 shows the FTIR spectra of graphite nanoplatelets (GNP) and functionalized graphite nanoplatelets (*f*-GNP). This spectra suggests the attachment of oxygen containing functional groups (-COOH, >C=O etc.) at the surface of GNP due to acid treatment. These functional groups at the surface of *f*-GNP act as anchoring sites for magnetite nanoparticles and hence better decoration over the surface.[1]

1.2 XPS study

Formation of Fe_3O_4-*f*-GNP nanocomposite was also confirmed with XPS analysis. Magnesium K_α radiation was used for X- ray generation. Figure-S2 is the wide scan spectrum of the sample in which the photoelectron lines at binding energy of about 285, 532, 711.5 and 725eV are attributed to C1s, O1s, $Fe2p_{1/2}$ and $Fe2p_{3/2}$ respectively. These peaks confirm the formation of Fe_3O_4-*f*-GNP nanocomposite.[2-4]
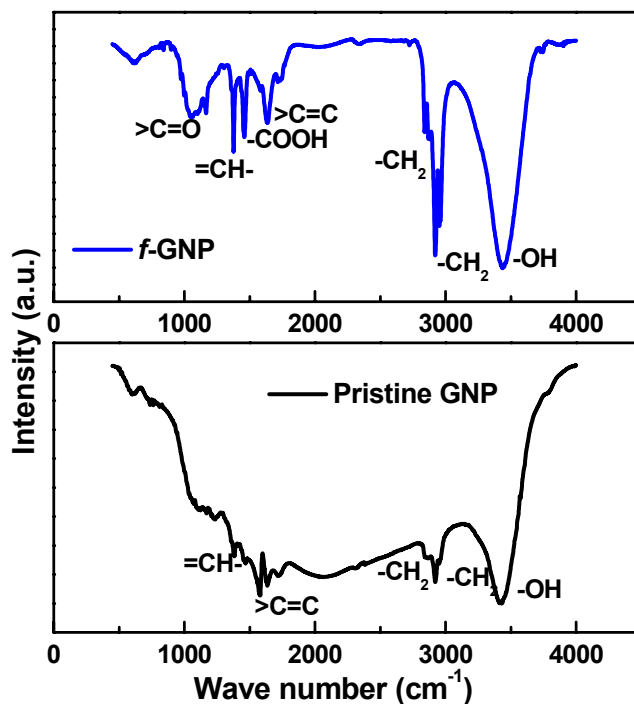
Figure-S1 FTIR spectra of pristine GNP and functionalized GNP
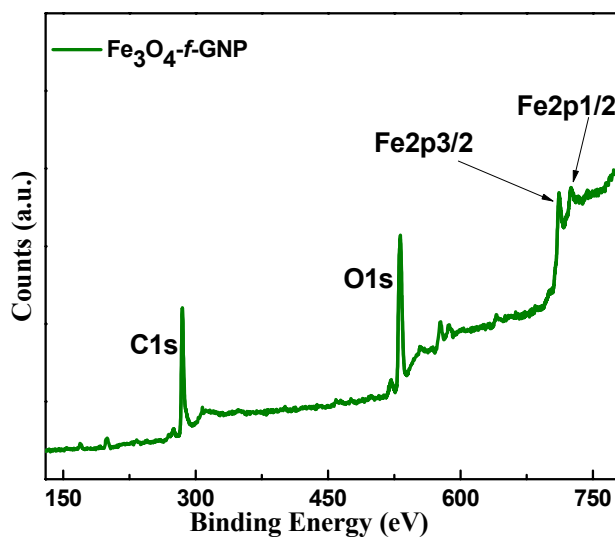

Figure- S2 X-ray photoelectron spectroscopy of Fe_3O_4-*f*-GNP nanocomposite

1.3 Surface area measurement

Nitrogen adsorption-desorption has been performed to analyze the textural nature of nanocomposite. Figure-S3 shows the N_2 adsorption-desorption isotherm for Fe_3O_4-*f*-GNP nanocomposite. It shows hysteresis for nanocomposite suggesting the high surface area of nanocomposite. The specific surface area of Fe_3O_4-*f*-GNP nanocomposite, calculated using BET equation was found to be $102.35 m^2/g$. Pore diameter was found to mainly vary from 3nm to 10nm for nanocomposite, suggesting the possible mesoporous nature of nanocomposite. The total pore volume using BJH method was found about $0.1509 cm^3/g$ for nanocomposite.
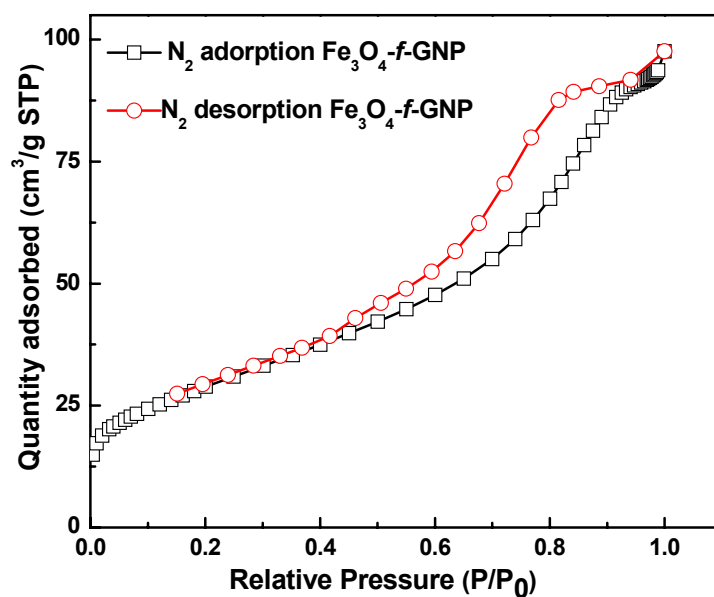

Figure- S3 N_2 adsorption-desorption isotherm for *f*-GNP and Fe_3O_4-*f*-GNP nanocomposite

1.4 Raman spectroscopy analysis

Figure-S4 shows the Raman spectrum of pristine and CO_2 adsorbed Fe_3O_4-*f*-GNP nanocomposite. It exhibits the D-band ($1334.4 cm^{-1}$) and G-band ($1563.4 cm^{-1}$), along with extra

peaks were observed at lower Raman shift values.[5] G-band corresponds to the tangential modes

of vibrations, while D-band corresponds to the defect of GNP.[6] Extra peaks arise due to the

formation of Fe_3O_4 nanoparticles over the surface of *f*-GNP. Peaks at lower Raman shift (218.6,

284.5, 391 and 588.6cm^{-1}) values may correspond to vibration modes of Fe-O bonds of Fe_3O_4

nanoparticles and Fe-C bonds at the surface of *f*-GNP. [7,8] Hence Raman study again confirms the

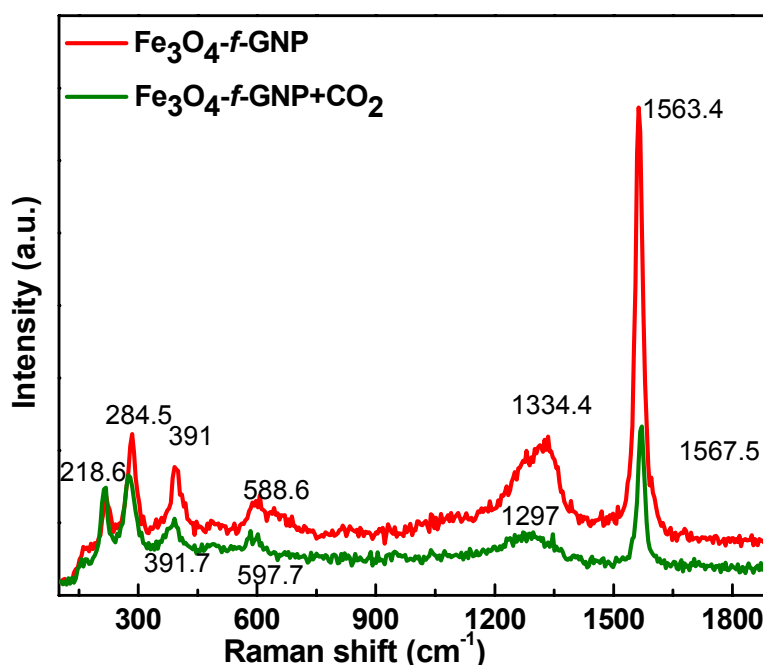formation of magnetite nanoparticles over surface of *f*-GNP.

Figure- S4 Raman spectra of pristine and CO_2 adsorbed Fe_3O_4-*f*-GNP nanocomposite

In the case of CO_2 adsorbed Fe_3O_4-*f*-GNP nanocomposite G-band shifts to higher value of

1567.5cm^{-1}, which may be attributed to the physisorption of CO_2 at the surface of

nanocomposite. Adsorbed CO_2 molecules at the surface of nanocomposite possibly increase the

reduced mass of harmonic oscillator and hence shift of G-band towards higher frequency. Peaks

corresponding to the Fe-O and Fe-C bonds (391.7 and 597.7cm^{-1}) also shows shift towards higher value. These peak shifts may correspond to the strong chemical interaction between magnetite nanoparticles and CO_2 molecule. Hence, Raman spectroscopy study of CO_2 adsorbed Fe$_3$O$_4$-*f*-GNP nanocomposite also suggests the physicochemical adsorption of CO_2.[9]

1.5 Dubinin-Radushkevitch (D.R.) Equation

The CO_2 adsorption isotherm at room temperature for nanocomposite was treated by the D.R. equation.[10,11] D.R. equation can be represented as follows-

$$\ln W = \ln W_0 + (R/E)^2 [T \ln(P_0/P)]^2 ----(S1)$$

Where 'W' is the amount of adsorbed CO_2, 'W_0' is the micropore volume and 'E' is the characteristic adsorption energy. 'W' is given by-

$$W = \frac{N \times M}{\rho} ----(S2)$$

Where 'N' is the amount of CO_2 adsorbed in mol/g, 'M' is the molecular weight (M=44g) and 'ρ' is the density of the CO_2 at the temperature T (ρ=0.85g/cm^3 at 298K). D.R. equation is based on the postulate that the mechanism for adsorption in micropores is that of pore filling rather than a layer-by-layer formation of a film on the walls of the pores.

Figure-S5 shows the variation of Ln(N) with $[T \times Ln(P/P_0)]^2$ and DR equation fit for Fe$_3$O$_4$-*f*-GNP nanocomposite in two different pressures ranges 3-5bar and 6-14bar at room temperature (25ºC). Micropore volume can be calculated by finding the intercept at Y-axis (N_0) and using the equation-S3 as below-
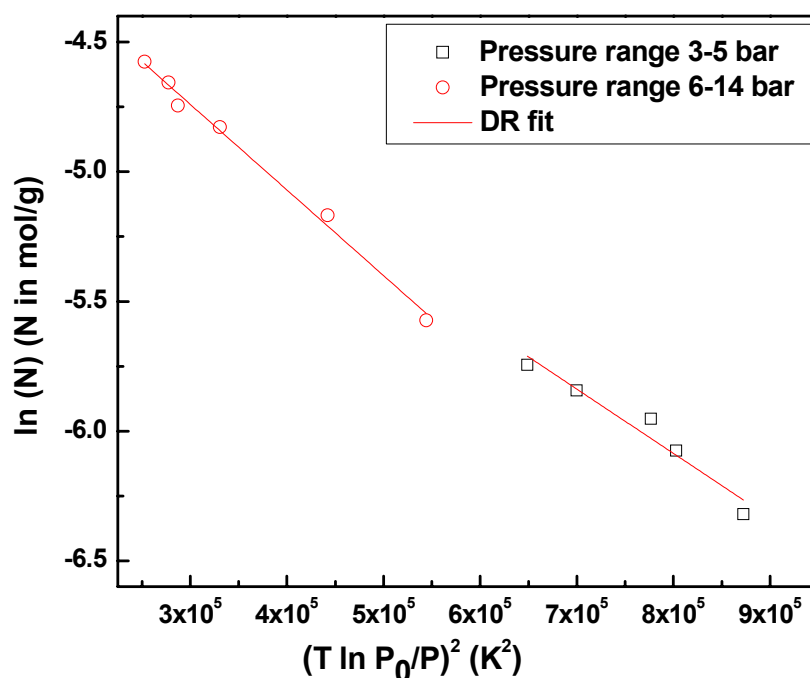
$$W_0 = \frac{N_0 \times M}{\rho} ----(S3)$$

Figure-S5 Dubinin-Radushkevitch fit for Fe_3O_4-*f*-GNP nanocomposite

Micropore volume depends upon the intercept and micropore volume for Fe_3O_4-*f*-GNP nanocomposite was found to be $0.858 cm^3/g$ with the adsorption in lower pressure range (3-5bar). Micropore volume at higher pressure range (6-14bar) was found to be $1.217 cm^3/g$. Large deviation of micropore volume for nanocomposite measured by DR fit of CO_2 adsorption with BJH method (pore volume=$0.1509 cm^3/g$) for N_2 adsorption-desorption isotherm was observed. In low pressure range isotherm deviates from DR fit, which may be attributed mainly to the interaction of CO_2 with magnetite nanoparticles and formation bicarbonates and carbonates.[24,31] In high pressure range increase in deviation was observed, which may be associated to the additional multilayer adsorption at high pressures. As DR fit suggests micropore filling, hence the large deviation of micropore volume also confirms the physicochemical adsorption behavior of Fe_3O_4-*f*-GNP nanocomposite for CO_2.

References

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