Supporting Information

Efficient reduction of Graphene Oxide to Graphene Nanosheets by Silica based Ionic Liquid: Synthesis, Characterization and Catalytic properties of IMD-Si/$FeCl_4^-$@GNS

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General experimental details

All solvents and chemicals used were purchased from Merck. Melting points of all synthesized compounds were taken in a Riechert Thermover instrument and are uncorrected. The IR spectra (KBr) were recorded on PerkinElmer RXI spectrometer. 1H NMR and 13C NMR spectra were recorded on a Bruker DRX-400 and Bruker Avance II 500 spectrometer using tetramethylsilane (TMS) as an internal standard and DMSOd6/CDCl3 as solvent. Mass spectra were recorded on Micromass Quattro II electrospray ionization (ESI) spectrometer. Elemental analyses (C, H and N) were conducted using the Elemental vario EL III elemental analyser. TGA data were obtained with a DSC-60 Shimadzu instrument. TGA were performed in the temperature range 0–600 °C at a constant heating rate of 20 °C min^{-1} in a nitrogen atmosphere. X-ray diffractograms (XRD) of the catalyst were recorded in the 2θ range of 5–90° with a scan rate of 8°/min on a Rigaku Minifax X-ray diffractometer with Ni-filtered Cu Ka radiation at a wavelength of 1.54060 Å. SEM-EDX characterization of the catalyst was performed on a JEOL JSM-6510 scanning electron microscope equipped with an energy-dispersive X-ray spectrometer operating at 20 kV. TEM analysis was performed on JEM-2100 F Model (ACC. Voltage: 200 kV) electron microscope. XPS was performed on PHI 5000 VersaProbe III instrument.

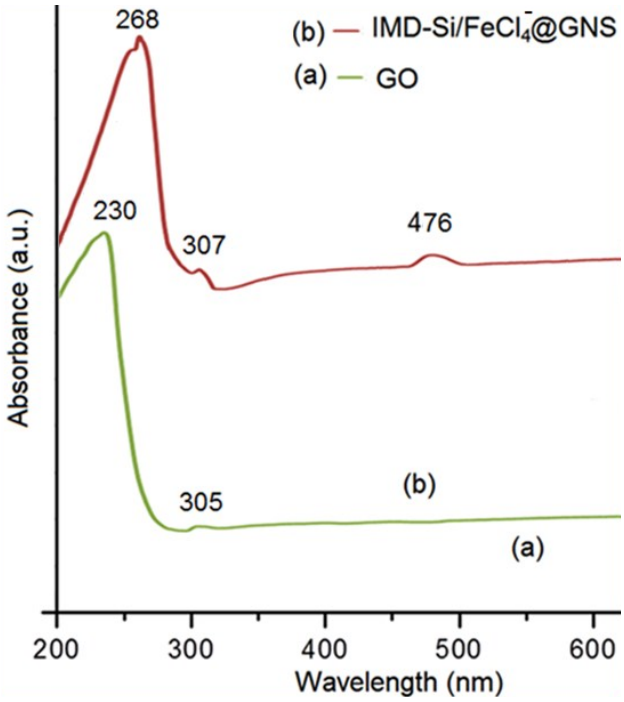


Figure S1. UV -Vis spectrum of (a) GO and (b) IMD-Si/$FeCl_4^-$@GNS

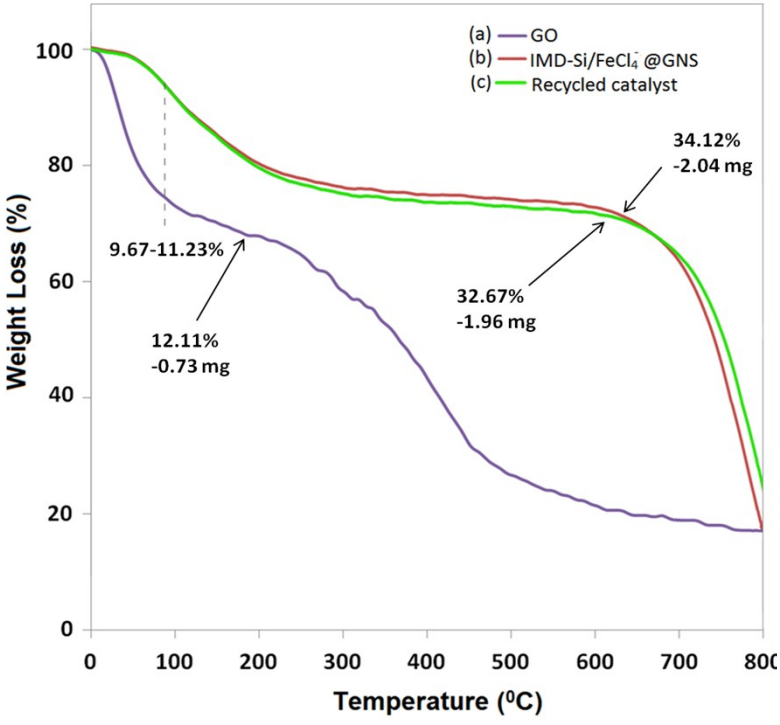


Figure S2. TG curves of (a) GO (b) IMD-Si/$FeCl_4^-$@GNS and (c) Recycled catalyst

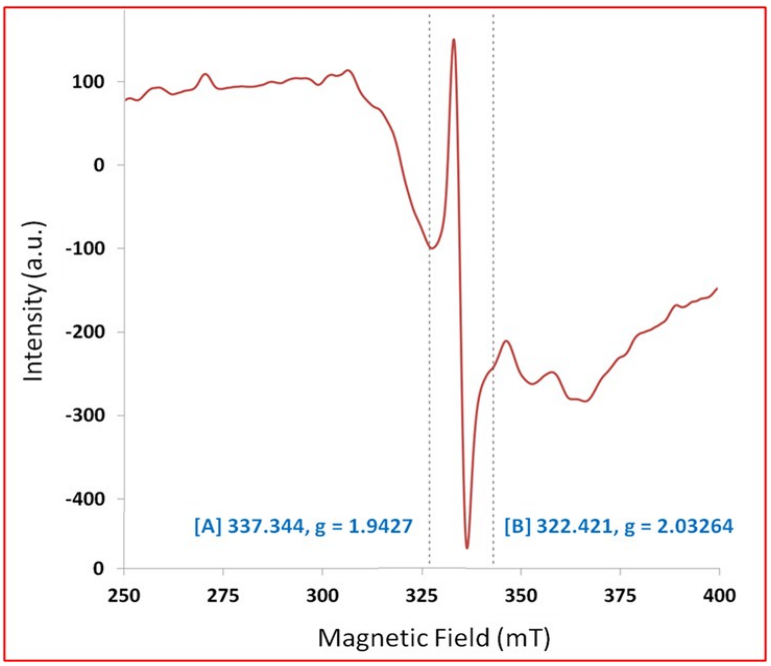


Figure S3. EPR spectrum of IMD-Si/$FeCl_4^-$@GNS catalyst

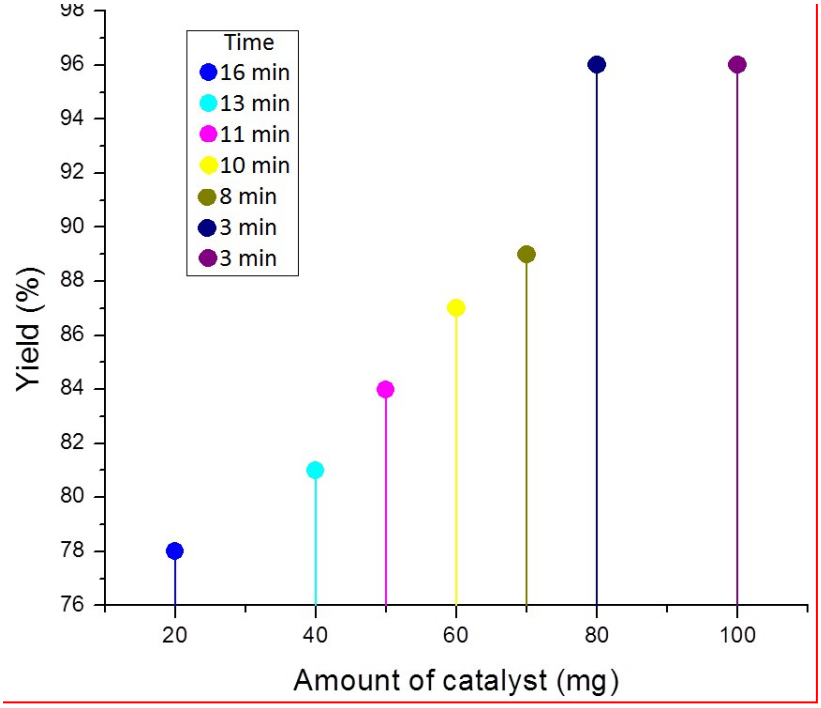


Figure S4: Effect of amount of material on model reaction[a] ([a]*Reaction conditions*: acetyl 1,3-dimethylbarbituric acid **1c** (2 mmol), thiosemicarbazide **2** (2 mmol), IMD-Si/$FeCl_4^-$@GNS, T = 30˚C. [b]Reaction progress monitored by TLC. [c]Isolated yield)

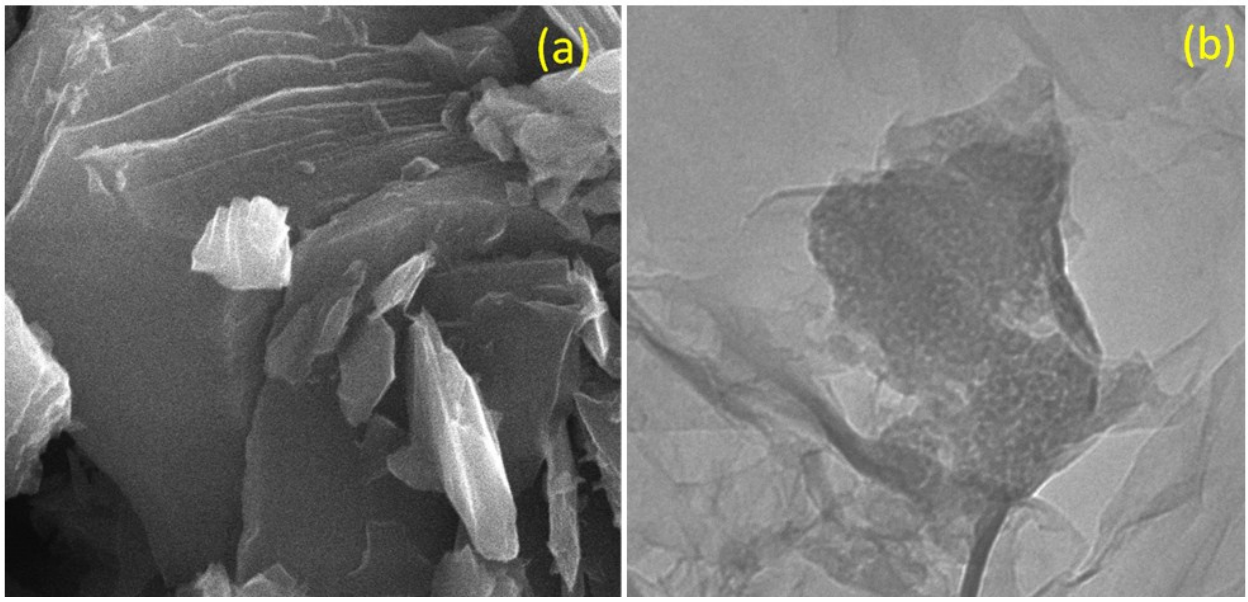
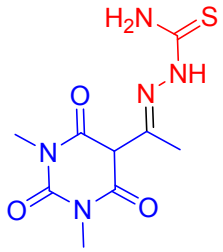
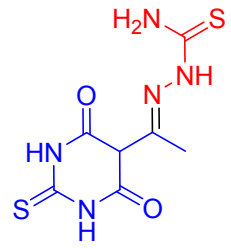
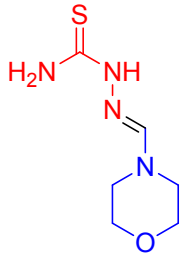
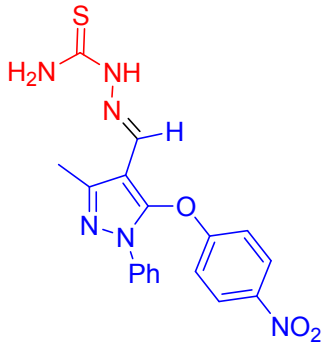


Figure S5: (a) SEM and (b) TEM images of recycled catalyst after five runs

Spectral data of newly synthesized compounds

(E)-2-(1-(1,3-dimethyl-2,4,6-trioxohexahydropyrimidin-5-yl)ethylidene)hydrazinecarbothioamide ***3c***	
	White solid: Mp: 240-242°C, IR (KBr cm^{-1}) 3315 (NH_2, asym), 3154 (NH), 2949 (C-H), 1705 (C=O), 1623 (C=N), 1519 (C=C, str.), 1161 (C=S); ^{1}HNMR (DMSO-d_6, 400 MHz): δ 1.90 (s, 3H, CH_3), 3.16 (s, 6H, 2xCH_3), 3.37 (s, 1H, CH), 7.86, 8.27 (s, 2H, NH_2), 13.20 (s, 1H, NH). ^{13}C NMR (DMSO-d_6, 100 MHz): δ 14.33, 54.00, 138.73, 141.12, 167.15, 176.93 Anal.Calcd ($C_{10}H_9N_3$): C, 39.84; H, 4.83; N, 25.81. Anal. Found ($C_{10}H_9N_3$): C, 39.16; H, 4.81; N, 25.90, ESI-MS *m/z*: 271.102 (M^+ + 1).
(E)-2-(1-(4,6-dioxo-2-thioxohexahydropyrimidin-5-yl)ethylidene)hydrazinecarbothioamide ***3d***	
	White solid: Mp: 212-215°C, IR (KBr cm^{-1}) 3437 (NH_2, sym), 3281 (NH_2, asym), 3165 (NH), 2993 (C-H), 1696 (C=O), 1601 (C=N), 1525 (C=C, str.), 1106 (C=S); ^{1}HNMR (DMSO-d_6, 400 MHz): δ 2.13 (s, 3H, CH_3), 3.30 (s, 1H, CH), 7.15, 7.51 (s, 2H, NH_2), 10.31, 10.59 (s, 2H, NH), 13.15 (s, 1H, NH). ^{13}C NMR (DMSO-d_6, 100 MHz): δ 14.33, 54.00, 138.73, 141.12, 167.15, 176.93 Anal.Calcd

	($C_{10}H_9N_3$): C, 32.43; H, 3.50; N, 27.10. Anal. Found ($C_{10}H_9N_3$): C, 32.27; H, 3.51; N, 27.24, ESI-MS *m/z*: 259.083 (M^+ + 1).
(E)-2-(morpholinomethylene)hydrazinecarbothioamide ***3e***	
	White solid: Mp: 223-225°C, IR (KBr cm^{-1}) 3437 (NH_2, sym), 3270 (NH_2, asym), 3156 (NH), 3030 (C-H, Ar.), 1599 (C=N), 1554 (C=C, str.), 1111 (C=S); ^{1}HNMR ($CDCl_3$, 400 MHz): δ 2.30 (t, 4H, 2xCH_2), 2.89 (t, 4H, 2xCH_2), 8.22, 8.35 (s, 2H, NH_2), 13.09 (s, 1H, NH). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 14.33, 54.00, 138.73, 141.12, 167.15, 176.93 Anal.Calcd ($C_{10}H_9N_3$): C, 38.31; H, 6.45; N, 29.74. Anal. Found ($C_{10}H_9N_3$): C, 38.43; H, 6.40; N, 29.68, ESI-MS *m/z*: 188.101 (M^+ + 1).
(E)-2-((3-methyl-5-(4-nitrophenoxy)-1-phenyl-1H-pyrazol-4-yl)methylene)hydrazinecarbothioamide ***3g***	
	White solid: Mp: 245-248°C, IR (KBr cm^{-1}) 3383 (NH_2, sym), 3256 (NH_2, asym), 3179 (NH), 3079 (C-H, Ar.), 1599 (C=N), 1538 (C=C, str.), 1155 (C=S); ^{1}HNMR ($CDCl_3$, 400 MHz): δ 2.71 (s, 3H, CH_3), 5.30 (s, 1H, CH), 7.54-8.50 (m, 11H, Ar-H+NH_2), 12.75 (s, 1H, NH). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 14.33, 54.00, 138.73, 141.12, 167.15, 176.93 Anal.Calcd ($C_{10}H_9N_3$): C, 54.47; H, 4.10; N, 21.32. Anal. Found ($C_{10}H_9N_3$): C, 54.42; H, 4.17; N, 21.45, ESI-MS *m/z*: 396.19 (M^+ + 1).