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Robust Charge Carrier Engineering Via Plasmonic effect and Conjugated Π-Framework on Au loaded ZnCr-LDH/RGO Photocatalyst towards H_2 and H_2O_2 Production

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S1. Calculation of Number of H_2 evolved (theoretical) and apparent conversion efficiency (ACE).

(a) Number of H_2 molecules generated over Au@LDH/RGO composite was calculated by the reported literature of Deka *et al.*:[3]

Number of H_2 produced from Au@LDH/RGO composite:

Volume of H_2 generated during the reaction period = 20.6 ml = 0.0206 L

Form standard gas equation, we have **PV= nRT**

n (no. of H_2 gas evolved) = 0.0206 L x 1 atm / 0.082 L.atm mol^{-1} K^{-1} x 298 K

The corresponding amount of hydrogen in moles/2h = 0.000843 moles/2h

As we know, 1 mole of H_2 gas = 6.023 x 10^{23} molecules of H_2

Therefore, 0.000843 moles = 6.023x 10^{23} x 0.000843 H_2 molecules

H_2 molecule (*per* cm^2 *per s*) = (6.023 x 10^{23} x 0.000843) / (14.13 x 2 h x 60 min x 60 s)

= **4.9903 x 10^{15} $cm^{-2}s^{-1}$**

Number of H_2 molecule (*per s*) = (6.023 * 10^{23} * 0.000843) / (2 h x 60 min x 60 s)

= **7.0513 x 10^{16} s^{-1}**

(b) Apparent conversion efficiency (ACE) of Au@LDH/RGO hybrid for H_2 production (918.76 µmol/2h in methanol solution) under 125W Hg lamp irradiation was calculated by following the below given formula. [2]

$$\Rightarrow \textbf{ACE} = \frac{Stored\ chemical\ energy\ (SCE)}{Incident\ photon\ intensity\ (IPI)}$$

SCE= Number of H_2 generated (moles /sec) * Heat of combustion of H_2 (kJ/mole)

= 0.127 * 10^{-6} mole/sec *285.8 * 10^3J/mole

=0.0362 W

IPI= Intensity of 125 W Hg lamp * Distance between lamp and reaction suspension surface * spherical surface area on which light is irradiated (2πr)

$= 0.027 \times 9 \times 2 \times 3.14 \times (1.5)^2$

= 0.3433W

$$\Rightarrow \textbf{ACE}= \frac{SCE}{ILI}$$

$$\Rightarrow \frac{0.0362W}{0.3433W} = \textbf{10.5\%}$$

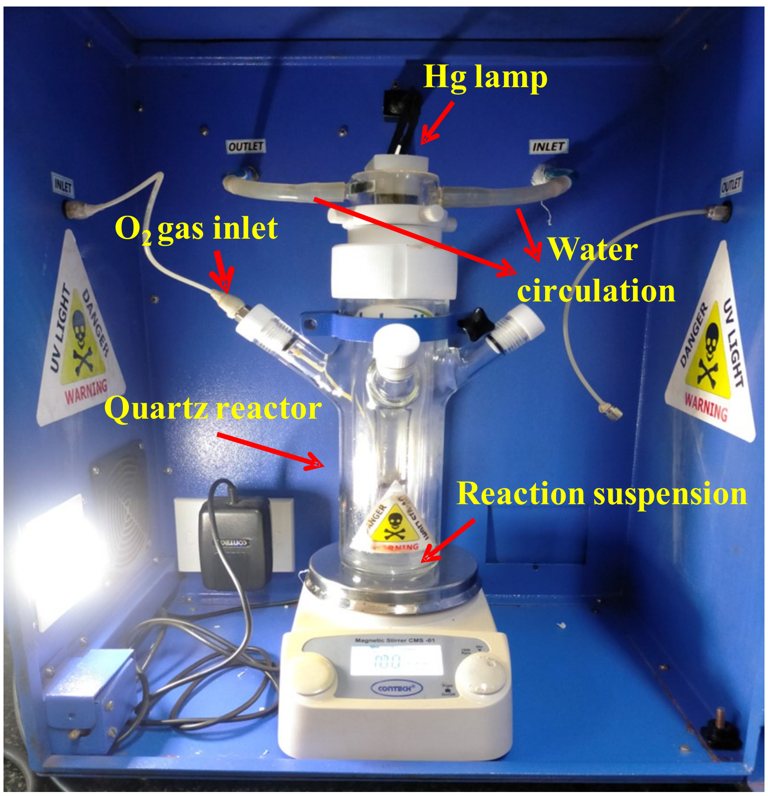

Fig. S2 Picture of photoreactor for H_2O_2 generation.

S3. Calculation of solar to chemical conversion efficiency (SCC %).

Solar to chemical conversion efficiency (SCC %) of Au@LDH/RGO composite towards H_2O_2 production under 250 W Hg lamp was calculated by following the below mention equation:

$$\text{SCC \%} = \frac{([\Delta G^\circ\ for\ H_2O_2\ production\ (J/mol)]\ \times\ [H_2O_2\ formed\ (mol)])}{([Input\ energy\ (W)]\ \times\ [reaction\ time(s)])} \times 100$$

Input energy = Intensity of used Hg lamp × Distance of lamp from catalyst mixed solution (9 cm) × Surface area of the spherical region on which light is focused (2πr, r =1.5 cm)

$=1.33 \times 9 \times 2 \times 3.14 \times (1.5)^2$

$= 169.13$ W

$$= \frac{117 \times 10^3 \times 24.3 \times 10^{-6}}{169.13 \times 2 \times 3600} \times 100$$

= **0.23%**

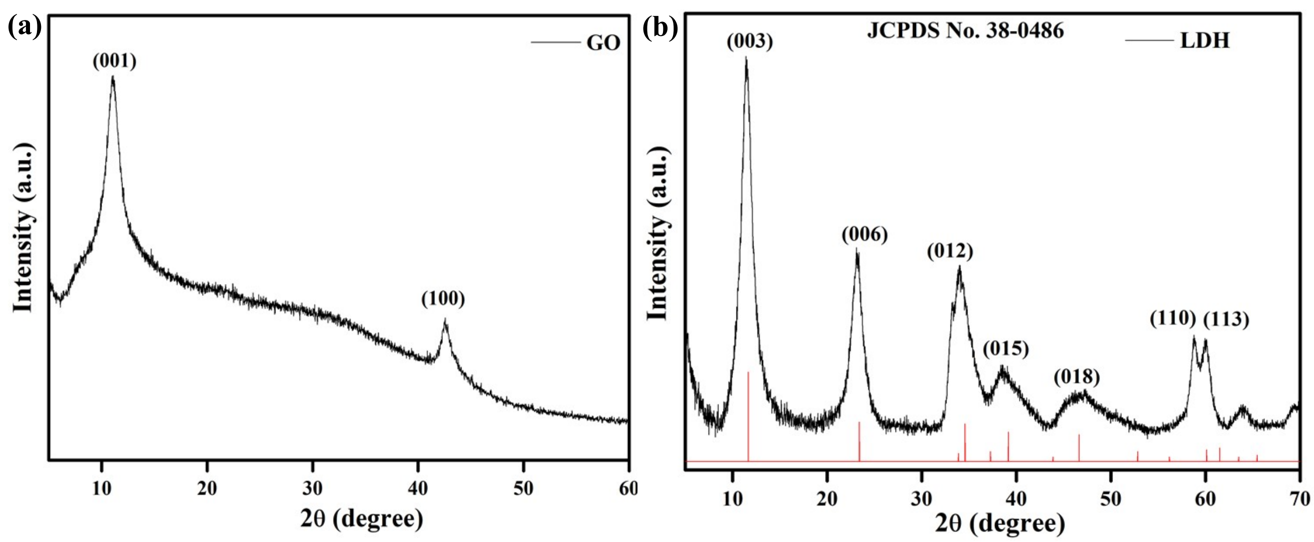

Fig. S4 XRD pattern of (a) GO and (b) LDH.

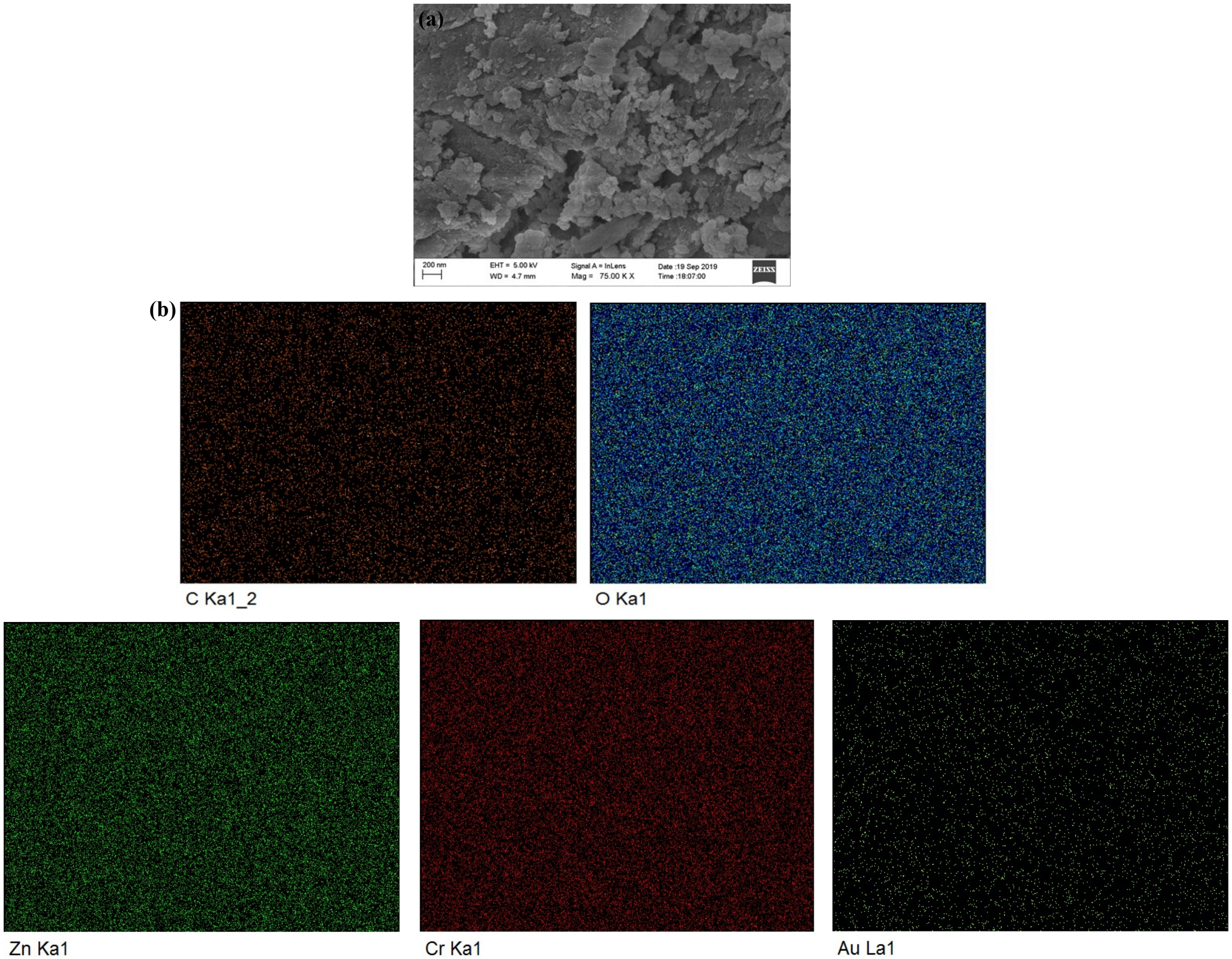

Fig. S5 (a) FESEM image and (b) colour elemental mapping image of Au@LDH/RGO.

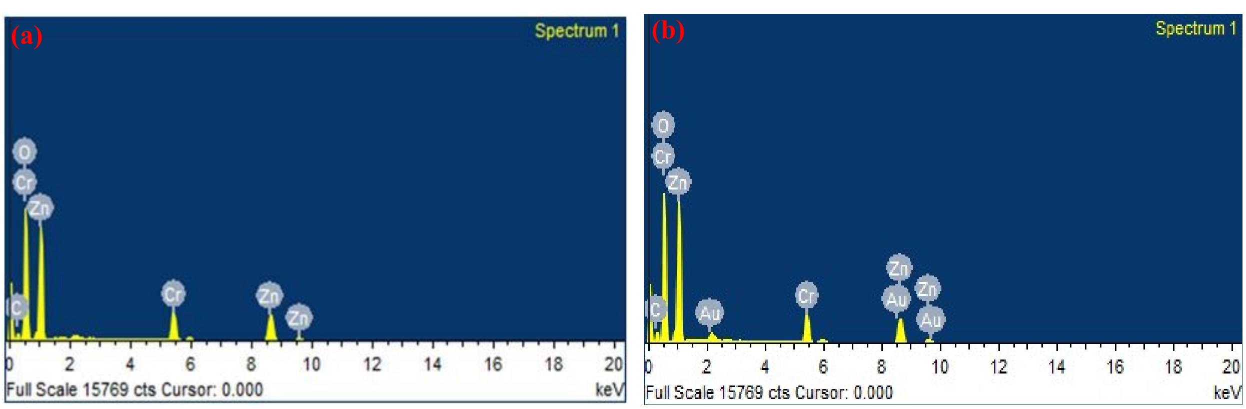

Fig. S6 EDAX of (a) LDH/RGO and (b) Au@LDH/RGO.

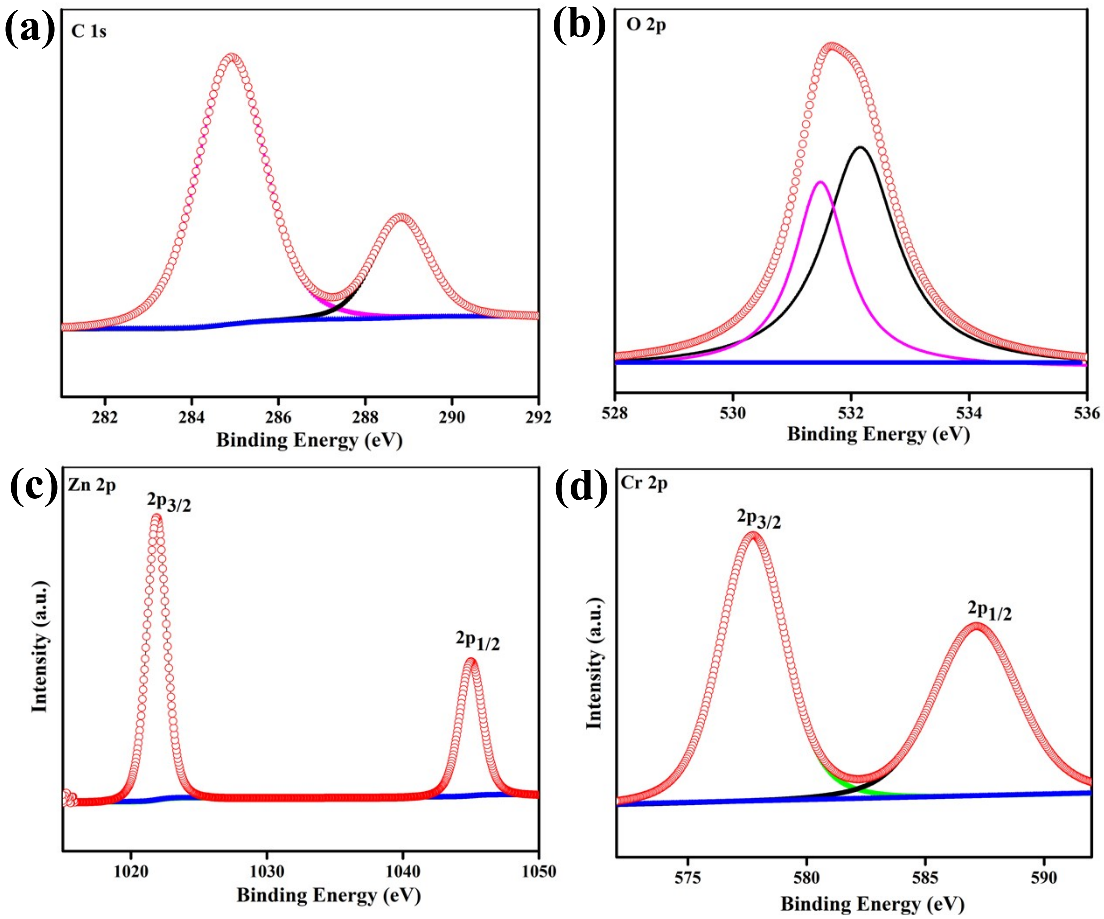

Fig. S7 XPS plot of LDH (a) C1s, (b) O 1s, (c) Zn 2p and (d) Cr 2p.

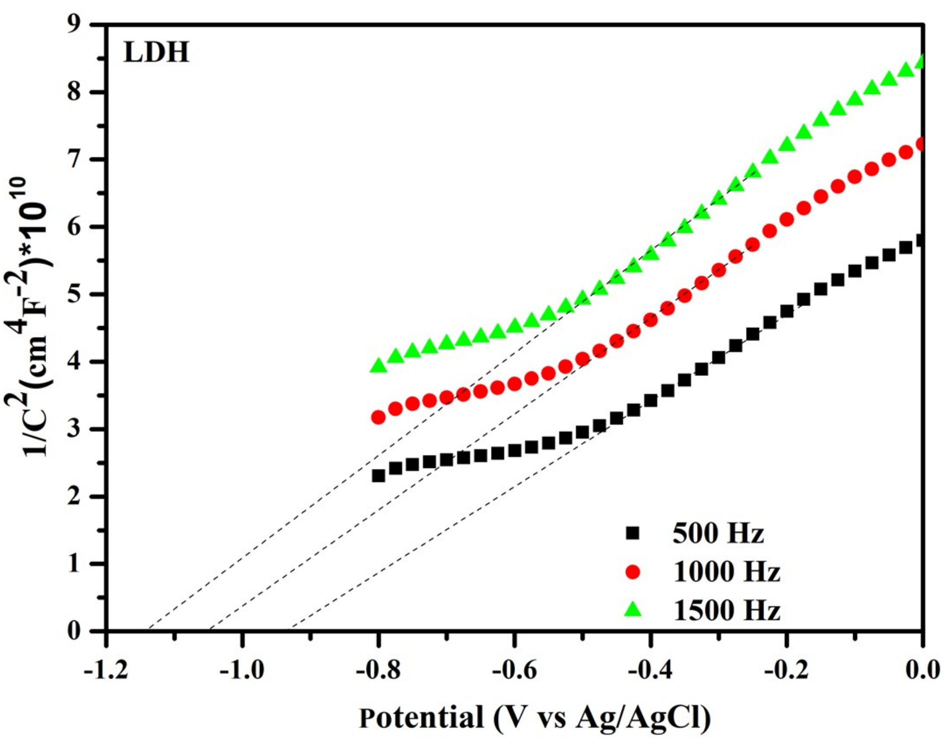

Fig. S8 Mott-Schottky graph of LDH at different frequency.

Table S9. Table represents the comparison study for photocatalytic H_2 evolution over present ternary heterostructure with the reported LDH, RGO and Au based system.

Photocatalyst	Light irradiation and sacrificial agents	H_2 evolution ($\mu molg^{-1}h^{-1}$)	Ref
rGO/$La_2Ti_2O_7$/NiFe-LDH	simulated solar irradiation, AM 1.5, TEOA	532.2	1
NiAl-LDH/g-C_3N_4/Ag_3PO_4	250 W quartz tungsten halogen lamp (λ≥420), CH_3OH	268	2
$CdIn_2S_4$/$In(OH)_3$/NiCr-LDH	300 W Xe lamp (λ≥400), Na_2S and Na_2SO_3.	1093	3

Au−Pd/rGO/TiO_2	300 W Xe lamp (λ≥420), CH_3OH	21500	4
$ZnIn_2S_4$-rGO-$CuInS_2$	150W Xe lamp, (λ≥420), Na_2S/Na_2SO_3	510	5
rGO/$CuFe_2O_4$-TiO_2	250 W Xe lamp, (λ≥420), glycerol-water mixture	35981	6
$InVO_4$-g-C_3N_4/rGO	simulated solar irradiation, AM 1.5, TEOA	7449	7
TiO_2-Ag-rGO	280 W Xe lamp, (λ≥420), CH_3OH	593.56	8
Au@LDH/RGO	125 W Xe lamp, (λ≥420), CH_3OH	22950	Present work

Table S10. Table represents the comparison study for photocatalytic H_2O_2 evolution over present ternary heterostructure with the reported RGO and Au based system.

Photocatalyst	Light irradiation and sacrificial agents	H_2O_2 production	Ref
CN/rGO@black phosphorus quantum dot	300 W arc Xe lamp (420< λ<780 nm)	181.69 µmol/L, 3h	9
CoPi/rGO/TiO_2	300 W Xe arc lamp (λ≥320 nm), 2-propanol	850 µmol, 3h	10
TiO_2/rGO/Carbon dots	simulated solar irradiation AM 1.5, 2-propanol	350 µmol, 1h	11

TiO_2/WO_3/rGO	simulated solar irradiation AM 1.5, 2-propanol	270 µmol, 1h	12
Au/SnO_2-TiO_2	UV light, alcohol	15000 µmol, 3h	13
Au@LDH/RGO	125 W Xe lamp, (λ≥420), CH_3OH	24.3 µmol, 2h	Present Work

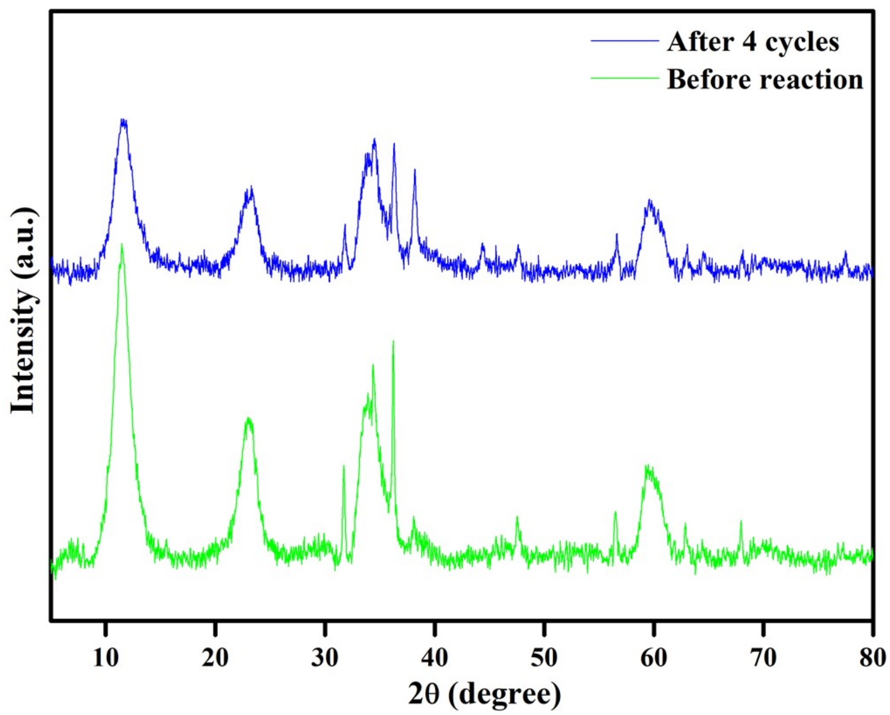

Fig. S11 XRD plot of Au@LDH/RGO sample after and before use.

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