HIGHLY DURABLE N-DOPED GRAPHENE/CdS NANOCOMPOSITES WITH ENHANCED PHOTOCATALYTIC HYDROGEN EVOLUTION FROM WATER UNDER VISIBLE LIGHT IRRADIATION

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INTRODUCTION

- Hydrogen is one of most promising green and renewable energy source for future.
- Hydrogen production from photocatalytic water splitting is a challenging and significant research topic today.
- Semiconductor photocatalysis is extensively used for hydrogen evolution.
- CdS (band gap = 2.4 eV) is one of the most studied visible-light photocatalyst for hydrogen production.
- Low efficiency of electron-hole separation and photocorrossion under prolonged irradiation limits the applicability of CdS.
- Conducting polymer films, carbon nanotubes, graphene etc. can be used as efficient electron transport matrices to retard the electron-hole recombination.
- **Graphene can be used as an efficient electron-transport matrix due to high specific surface area, tunable band gap, and very high electron mobility.**

- Incorporation of heteroatoms (N and B) are used to modulate the electronic properties of carbon nanomaterials.

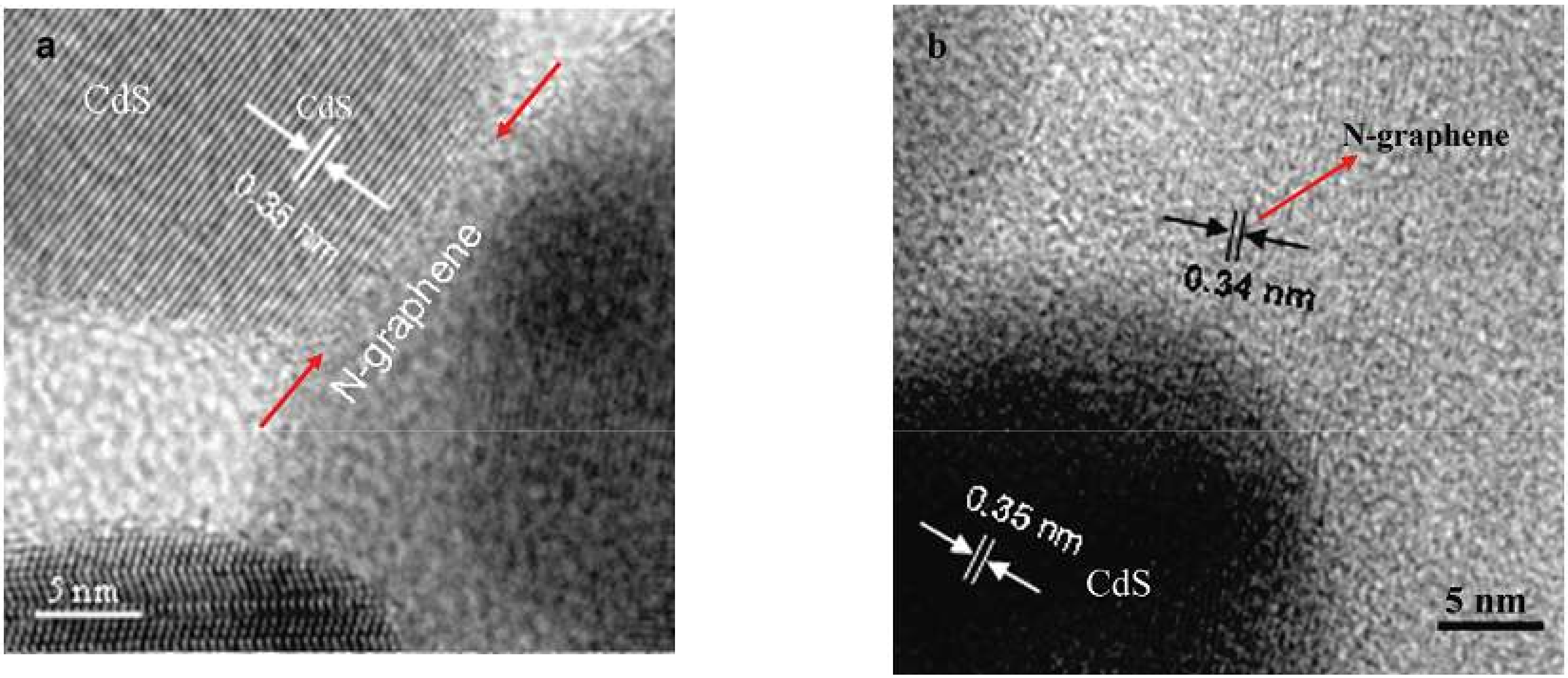
- N-doped carbon nanomaterials are highly promising materials for enhancing efficiency of fuel cells, photovoltaic cells, etc.

- **In this work,**

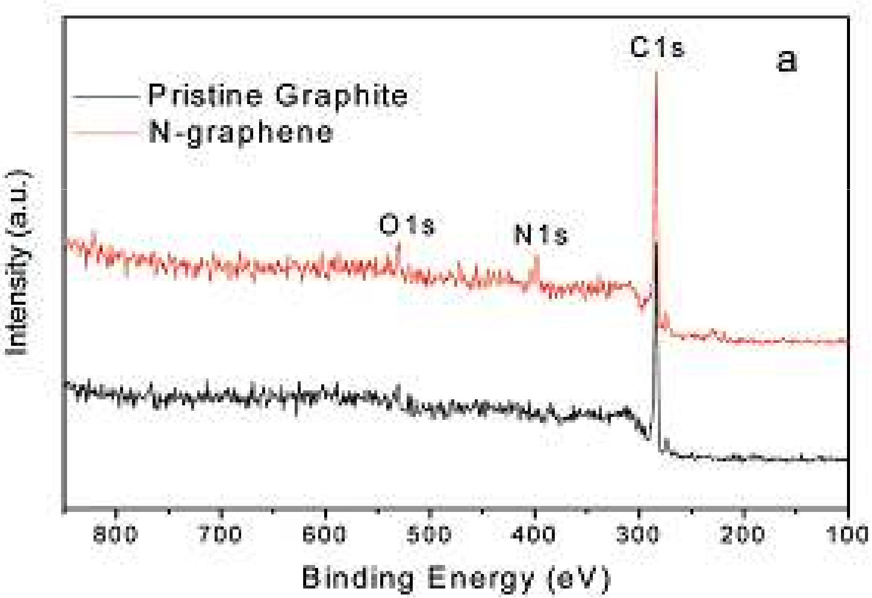
N-doped graphene/CdS photocatalyst was prepared and used for effiecient hydrogen production under visible-light irradiation.

EXPERIMENTAL SECTION

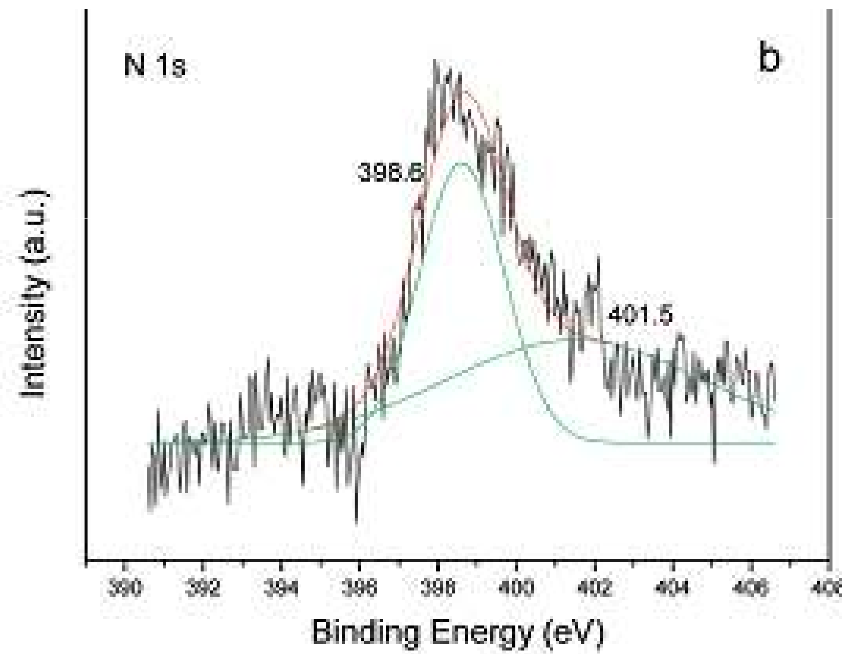
- Graphene oxide was prepared by a modified Hummers' method from natural graphite.
- N-Graphene was obtained by annealing graphene oxide in NH_3 gas.
- N-Graphene/CdS was prepared in two steps from N-Graphene, $CdCl_2$ 2.5 H_2O and Na_2S as starting materials.
- Three electrode system: Ag/AgCl reference elctrode, Pt wire as counter electrode and N-Graphene/CdS coated on ITO-plates as the working electrode and 0.5 M Na_2SO_4 as electrolyte.
- The amount of hydrogen formed was monitored by gas chormatography.


a
CdS
CdS
0.35 nm
N-graphene
5 nm
b
N-graphene
0.34 nm
0.35 nm
CdS
5 nm

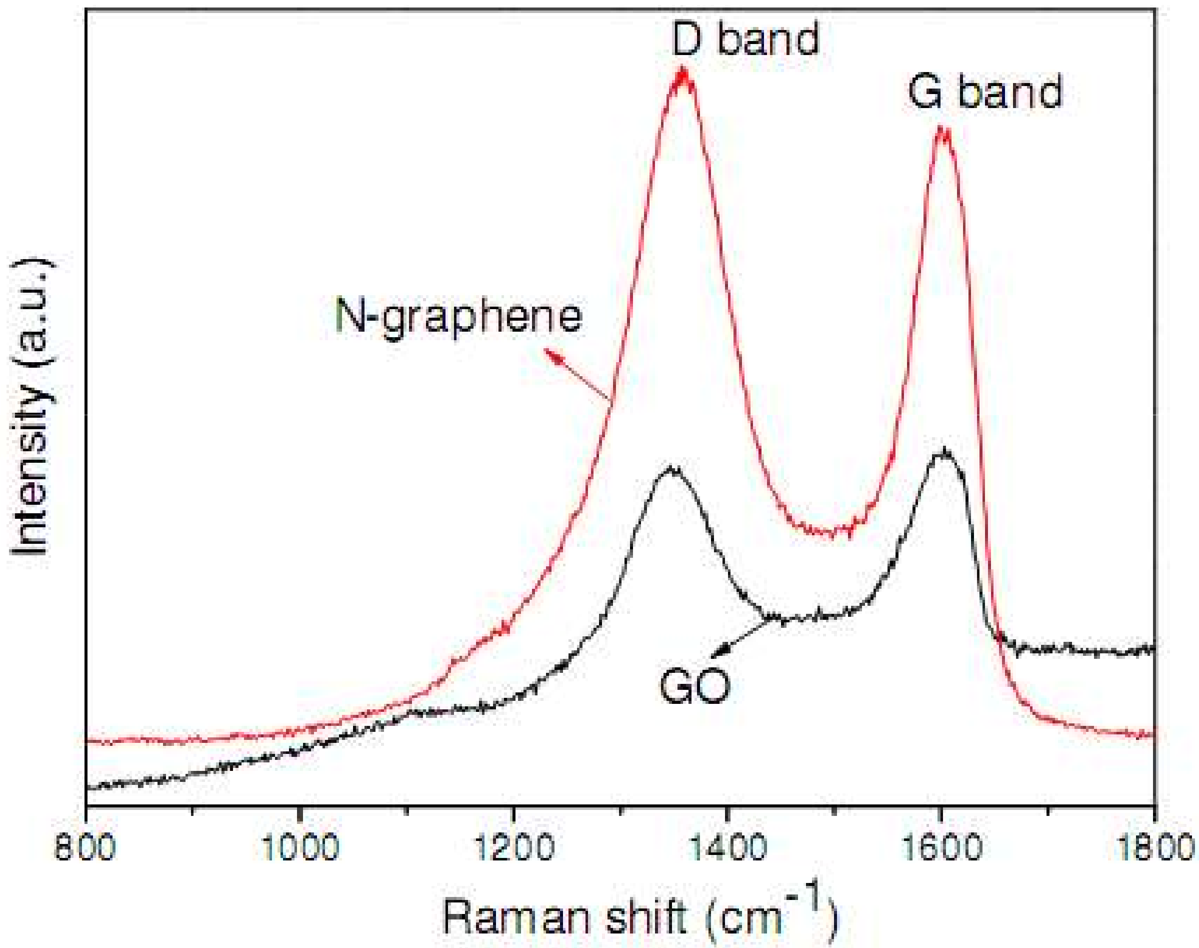
XPS spectra of pristine graphite and the N- doped graphene.

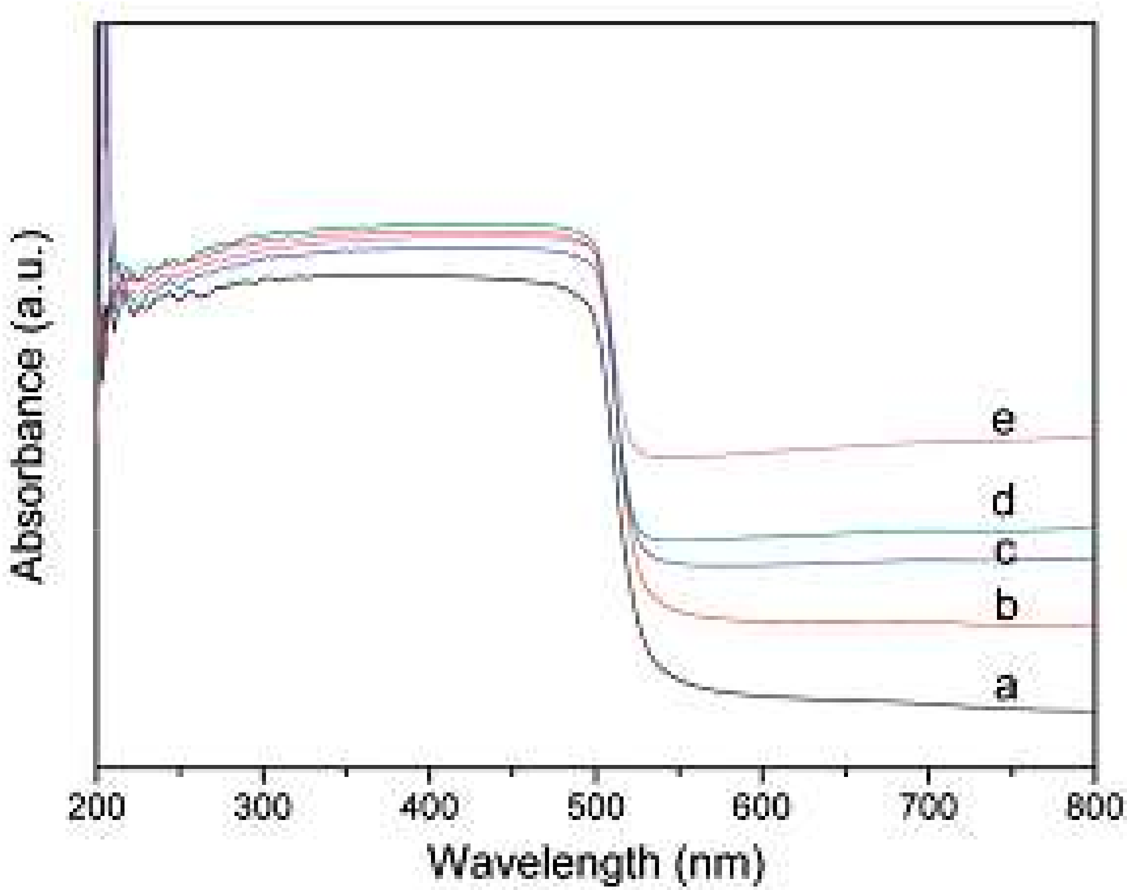


XPS spectra showing the presence of pyridinic and quarternary nitrogens.



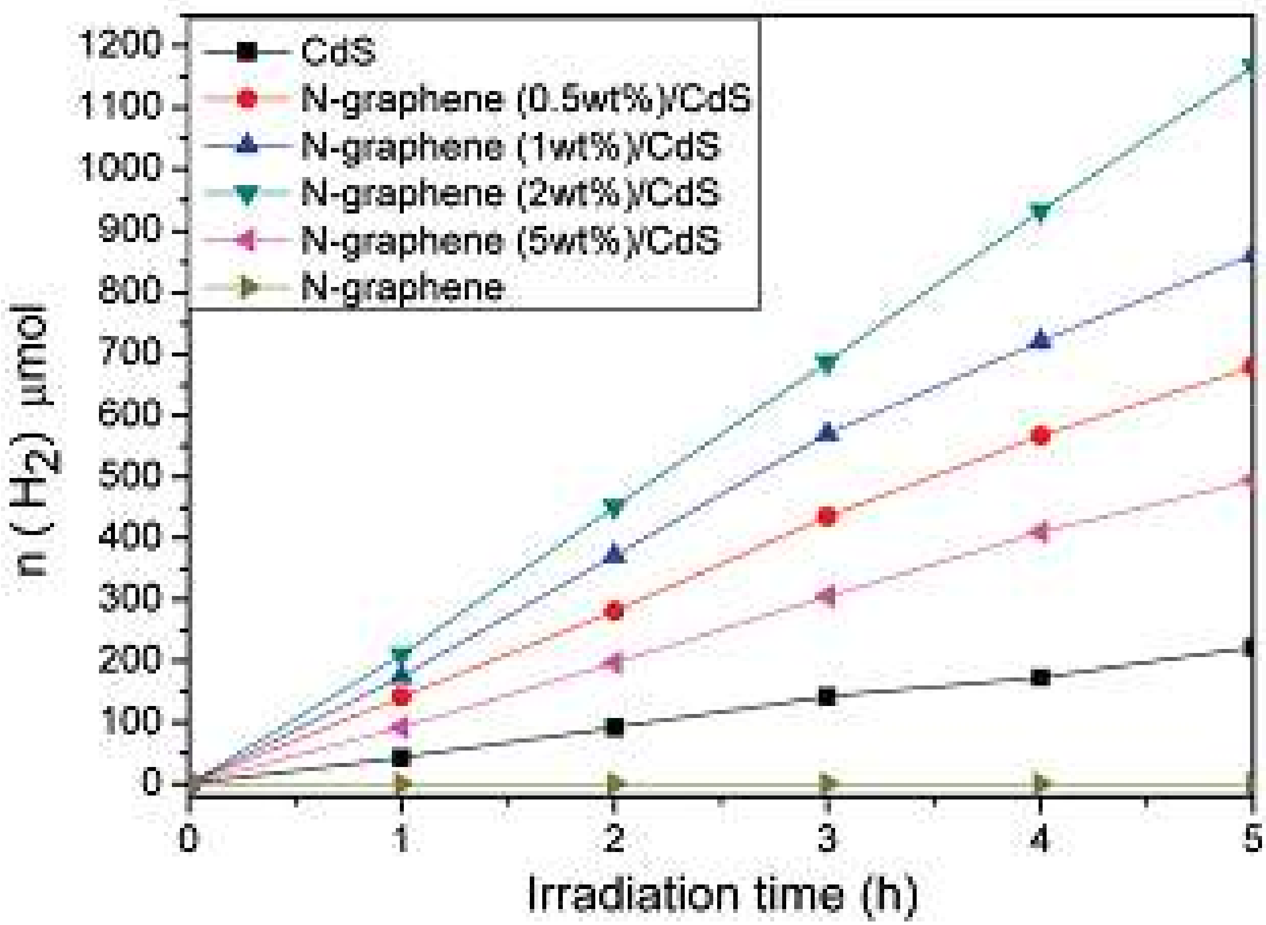
Raman spectra of GO and N-Graphene samples.

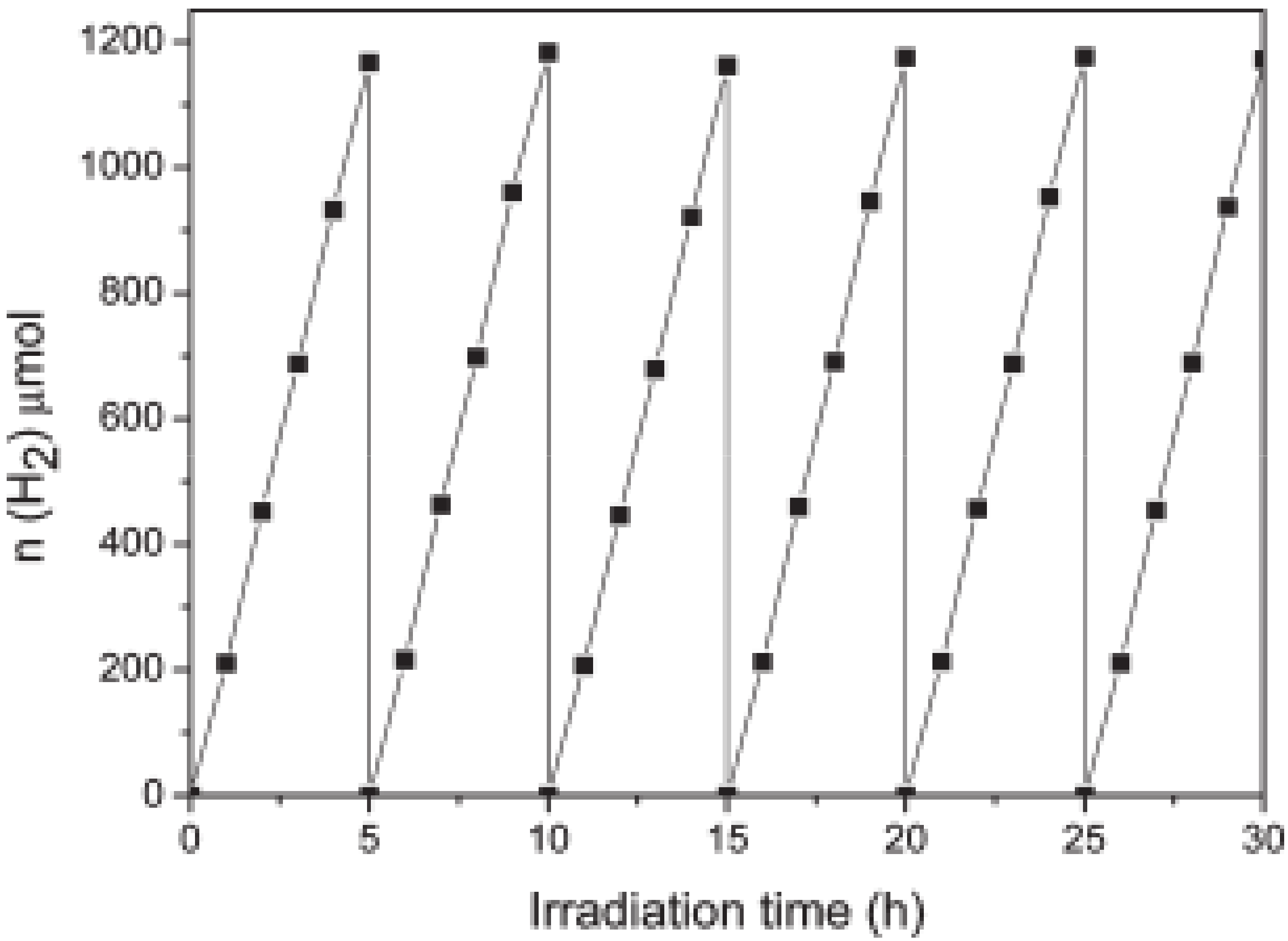




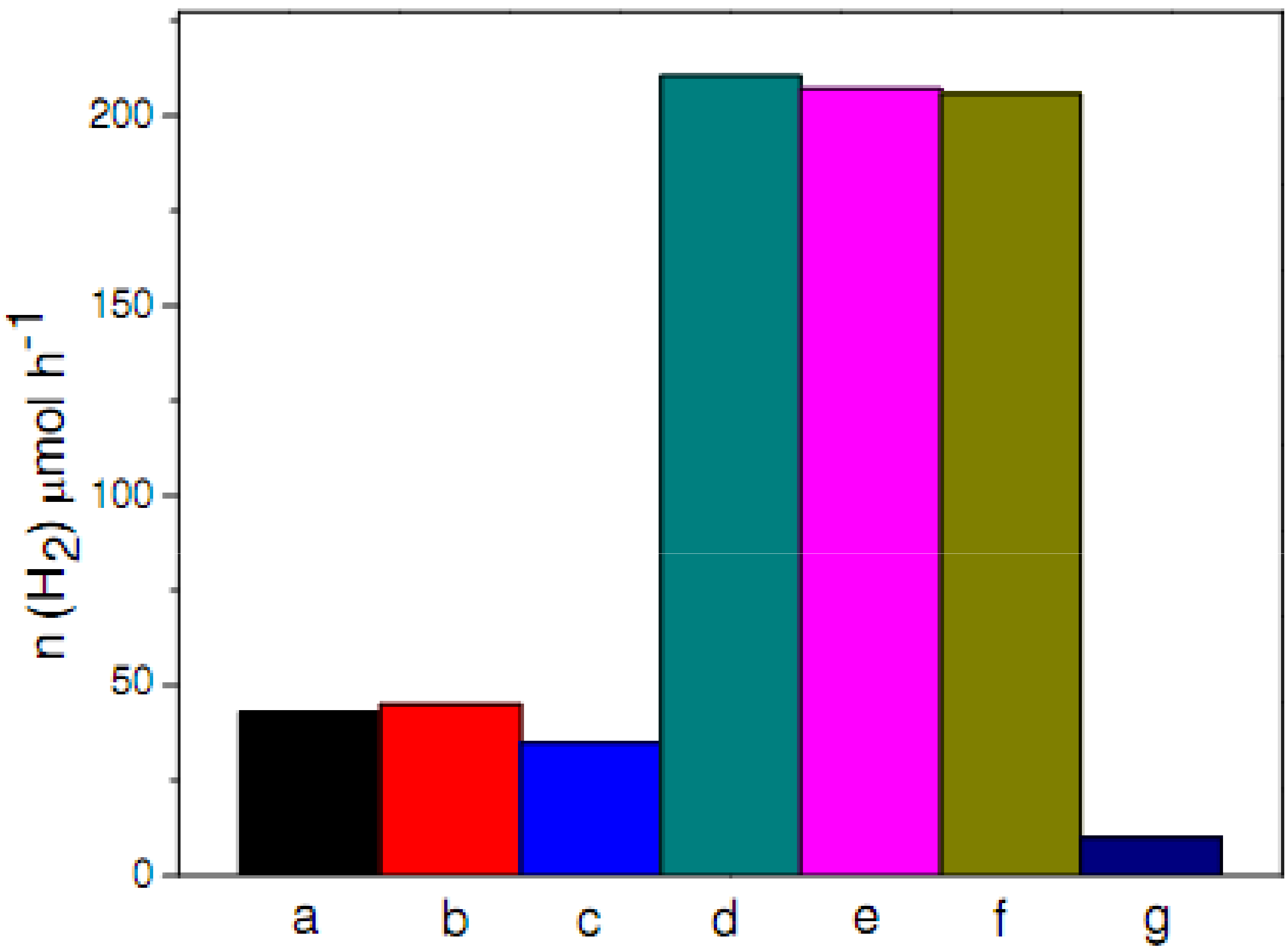
UV-vis diffuse reflectance spectra of pure CdS (a) and the prepared composite samples with different loading of N-graphene: 0.5 wt % (b), 1 wt % (c), 2 wt % (d), and 5 wt % (e).

DEPENDENCE ON AMOUNT OF N-GRAPHENE



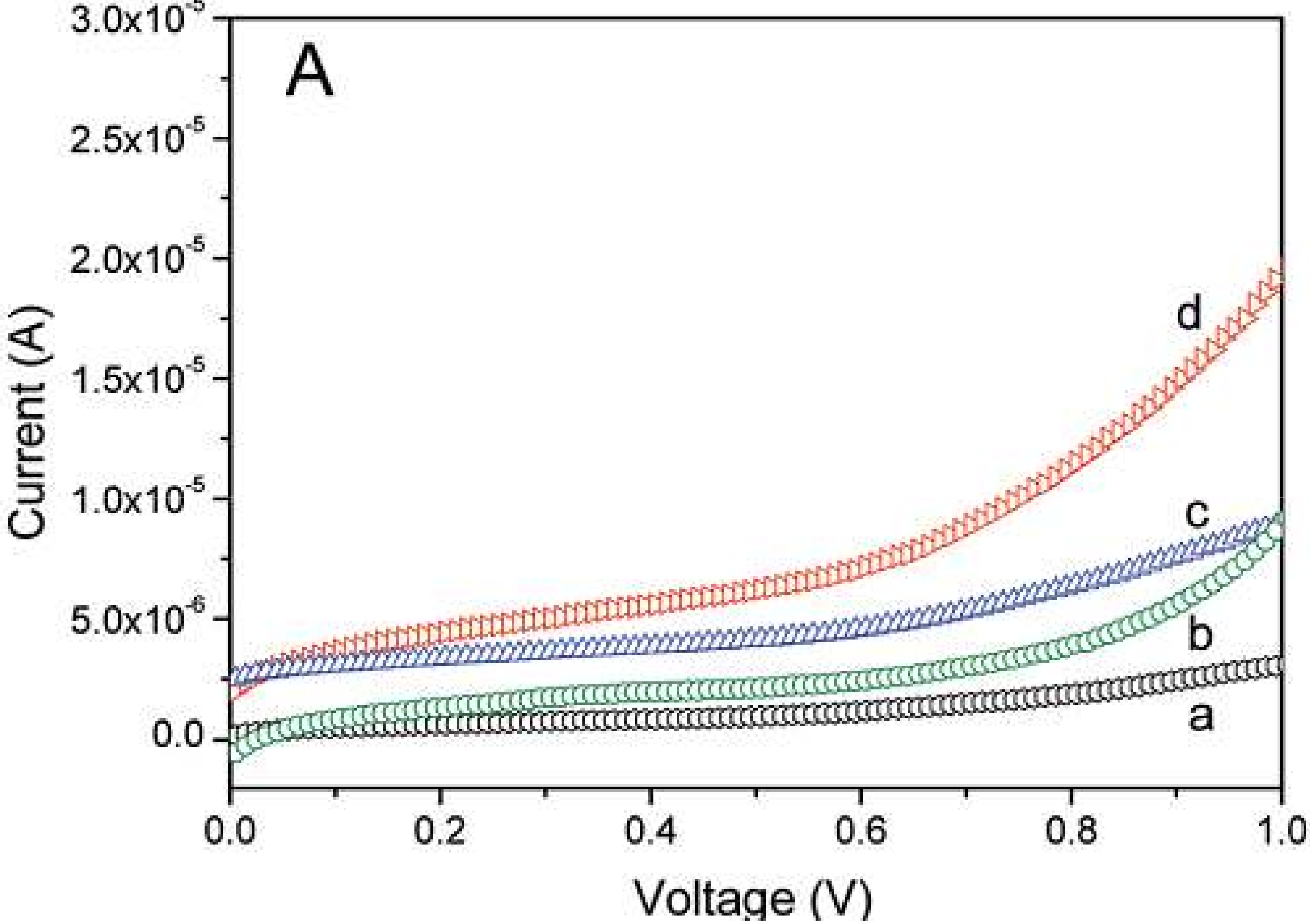


1200
1000
800
600
400
200
0
0
5
10
15
20
25
30
n (H2) µmol
Irradiation time (h)

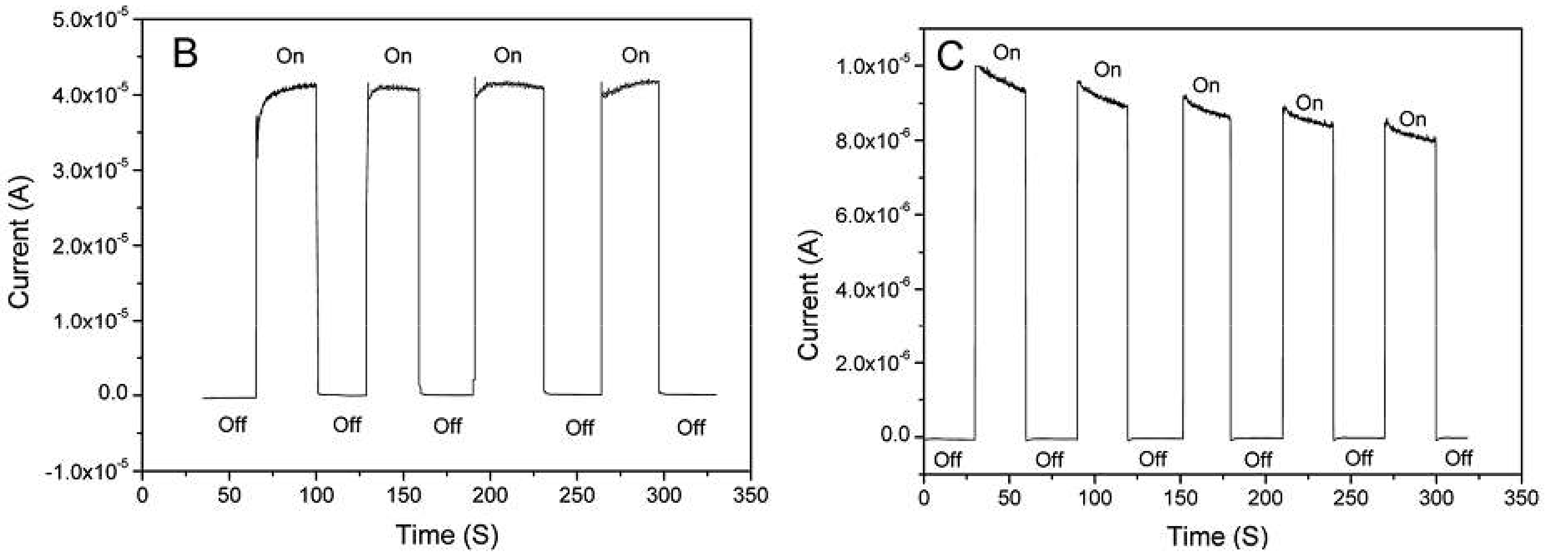


n (H2) µmol h-1
0
50
100
150
200
a
b
c
d
e
f
g

TRANSIENT PHOTOCURRENTS OF N-GRAPHENE/CdS CATALYSTS AND CdS WITH AND WITHOUT IRRADIATION (λ > 420 nm)

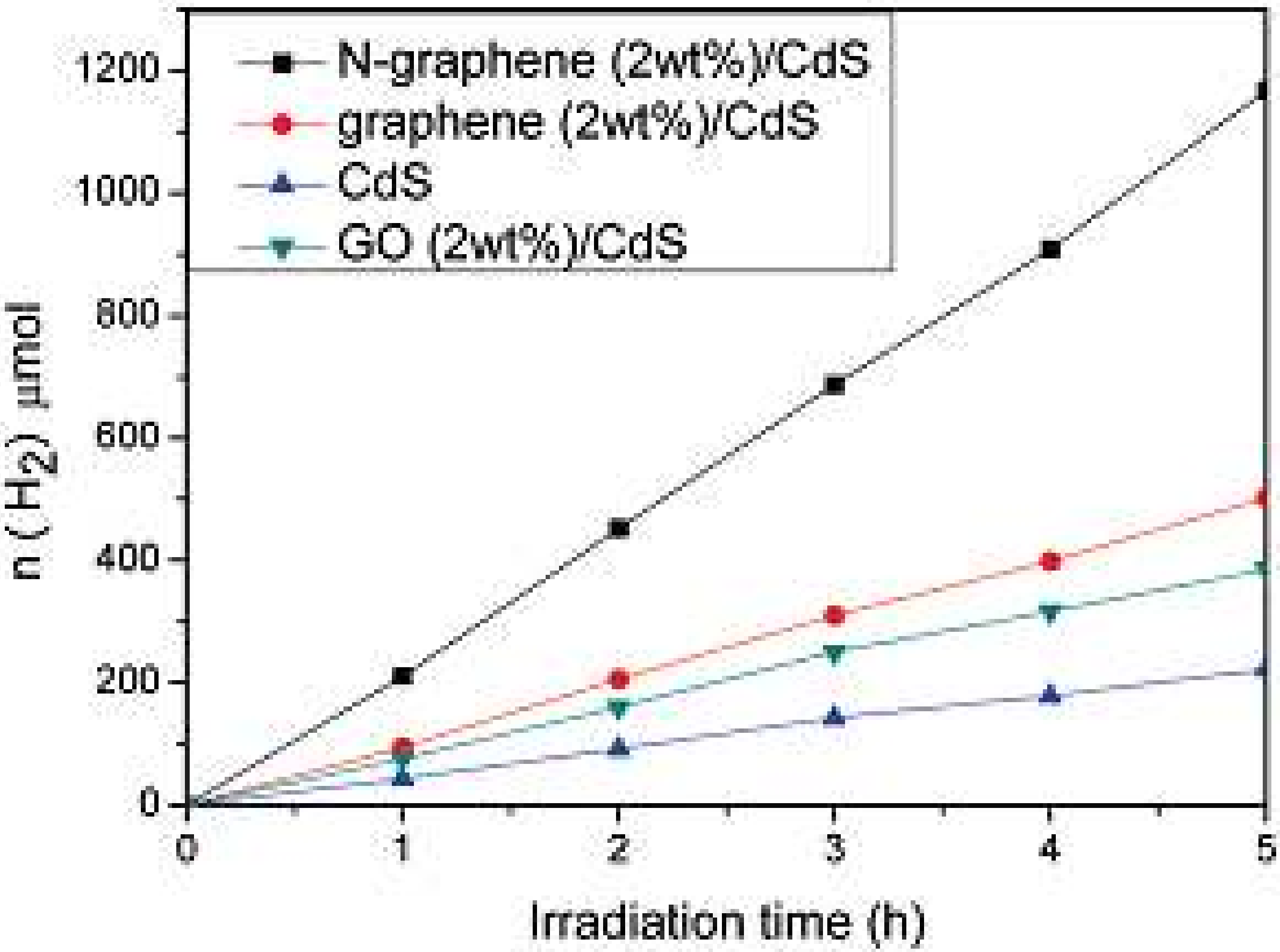


(a) CdS (light off), (b) N-graphene/CdS (light off), (c) CdS (light on), and (d) N-graphene/CdS (light on)



Current vs time curves for N-Graphene/CdS (B) and for CdS (C), under chopped irradiation with an electrode potential of 0 V vs Ag/AgCl

COMPARISON WITH UNDOPED GRAPHENE SYSTEMS



REASONS FOR ENHANCED PHOTOCATLYTIC ACTIVITY OF N-GRAPHENE-CdS COMPOSITE

- The intimate contact between N-Graphene and CdS favors the formation of junctions between the two materials, favoring the charge separation.

- BET surface area of samples annealed atd ifferent teperatyres ar almost equal, which suggest that surface area is not the determining factor.

- Due to the band structure modification, N-Graphene can absorb the visible light, and cause a slight shift in the absorption edge to the visible range.

- Enhancement of the catalytic activity could be attributed to predominent electrical conductivity of N-Graphene.

- It was suggested that carbon atoms of graphene sheets are accessible to protons that can readily tranform to hydrogen by accepting photogenerated electrons.

- Teng et al. reported that graphene oxide works as a metal-free photocatalyst to give hydrogen from water-methanol mixture, indicatin the hydrogen evolution on graphene surfaces.

CONCLUSIONS

- N-Graphene/CdS photocatalyst was successfully prepared for hydrgen production under visible light irradiation.
- N-Graphene/CdS can be used for continuous and stable hydrogen production from water.
- Optimal catalytic activity was found for 2 wt % loading of N-Graphene in the composites.
- This study shows that the new catalysts can substitute noble metal as cocatalysts.
- N-Graphene as a cocatalyst prevents CdS from photocorrosion.
- Further study is needed to establish the exact mechanism for enhanced catalytic activity.

THANKS