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# Development of g-CN-Pt electrocatalysts for Vis-light activated ethanol electrochemical valorization

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SCAN ME

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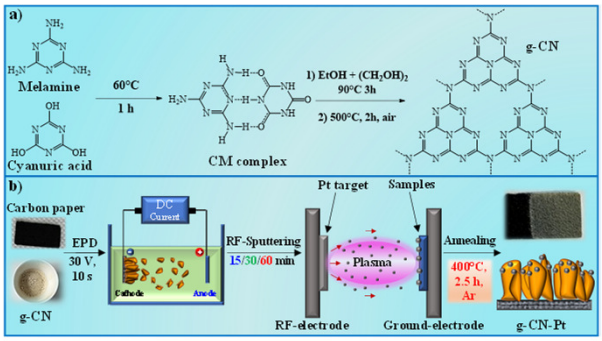
## INTRODUCTION & AIM

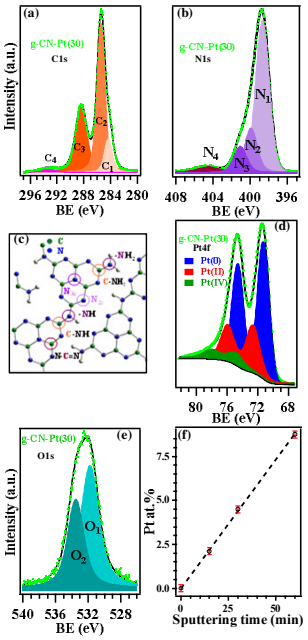
- **Exfoliated graphitic carbon nitride (g-CN)** was immobilized on C paper substrates by electrophoresis, and decorated with **ultra-low amounts** (≈μg/cm²) **of Pt nanoparticles (NPs)** by cold plasma sputtering
- Optimization of processing conditions allowed a **fine tuning of Pt NPs size, loading and distribution** and a *controlled tailoring of g-CN/Pt interfacial interactions*
- The prepared electrodes were used to perform the **Ethanol Oxidation Reaction (EOR)** in alkaline solution → appealing not only for the **valorization of biomass-derived ethanol aimed at $H_2$ production**, but also in **direct ethanol fuel cells (DEFCs) for clean energy generation.**
- The best system yielded a **Tafel slope of 96 mV/dec** and a **current density of 0.58 mA/cm² at 1.55 V *vs.* the reversible hydrogen electrode (RHE).** These results and the good system service life recommend the developed materials as attractive electrocatalysts for ethanol valorization *using electrical energy from renewable sources*

## METHOD




**(a)** Synthesis of g-CN powders; **(b)** deposition, functionalization and thermal treatment to obtain C paper-supported g-CN-Pt specimens. The target specimens are labeled as **gCN-Pt(X)** [X = duration of Pt sputtering process (min)].

## RESULTS & DISCUSSION




### XPS

**(a)** C1s and **(b)** N1s peaks for **g-CN-Pt(30)**. **(c)** Sketch of gCN structure, in which the different non-equivalent C and N sites are marked. **(d)** Pt4f and **(e)** O1s peaks for **g-CN-Pt(30)**. **(f)** Pt atomic percentage (at.%) *vs.* sputtering time.

**g-CN & Pt presence (∝ Sputtering time!)**

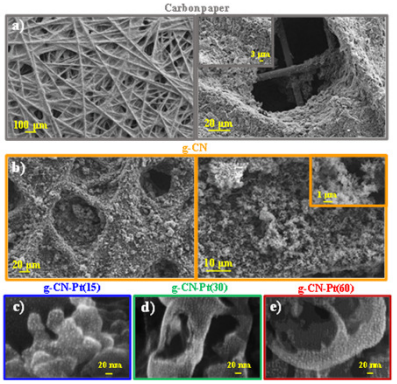
Partially uncovered substrate

**-$NH_x$ occurrence ($N_3$):**

- ⇓ ***e*⁻-*h*⁺ recombination**, ⇑ **photoactivity**
- **C-N=C groups** on g-CN promote **Pt adhesion & dispersion**, also *preventing NPs agglomeration*

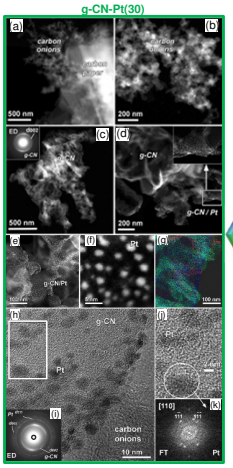
### SEM

**(a)** C paper, **(b)** g-CN, **(c)** **g-CN-Pt(15)**, **(d)** **g-CN-Pt(30)**, and **(e)** **g-CN-Pt(60)**.

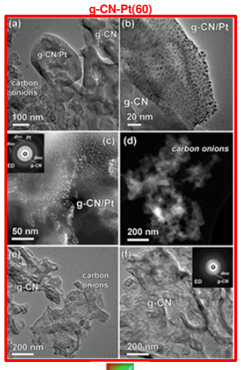



- ✓ g-CN particles (5-10 μm) on C paper fibers
- ✓ **<5 nm Pt NPs** (bright spots)
- ✓ **Pt dispersion**: efficient g-CN/Pt interfacial contact

### TEM



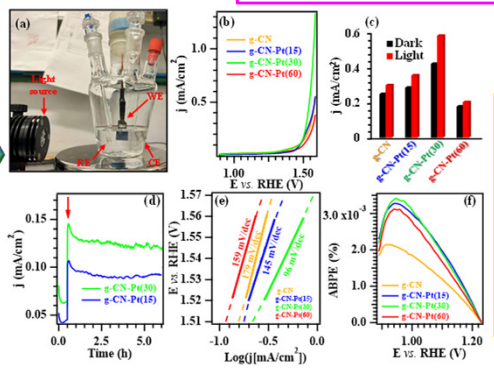

HAADF-STEM) images of **g-CN-Pt(30)**, showing: **(a)** C paper/C onions interface; **(b)** C onions; **(c)** flake-like g-CN & ED pattern; **(d)** g-CN & g-CN/Pt interface. **(e)** HAADF-STEM overview and **(f)** magnified image of g-CN/Pt nanostructures of g-CN-Pt(30). **(g)** EDXS-STEM overlaid image (C, blue; N, green; Pt, red). **(h)** BF HR-TEM overview of g-CN/Pt intermixed region. **(i)** ED pattern from g-CN/Pt (superposition of g-CN & Pt contributions). **(j)** Magnified image of the region highlighted by the white box in **(h)**. **(k)** FT pattern of the region evidenced by the white circle in **(j)**.



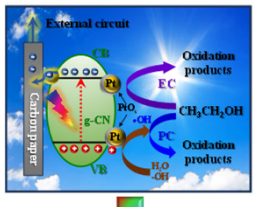

**(a,b)** BF-TEM images of **g-CN-Pt(60)**. **(c)** HAADF-STEM image of g-CN/Pt and ED pattern (superposition of g-CN and Pt contributions). **(d)** HAADF-STEM image of C onions. **(e)** BF-TEM image of g-CN/C onions interface. **(f)** BF-TEM image of g-CN and ED pattern, dominated by the sole g-CN (002) ring.

### Functional characterization

**(a)** Photograph of the used cell. **(b)** LSV curves in KOH 0.5 M+EtOH 1.3 M under irradiation. **(c)** Current density at 1.55 V *vs.* RHE during EOR tests. **(d)** Chronoamperometry for **g-CN-Pt(15)** and **g-CN-Pt(30)** at 1.55 V *vs.* RHE under illumination. *The arrow marks EtOH introduction.* **(e)** Tafel plots under irradiation, corresponding to LSV curves in **(b)**. **(f)** ABPE (%) curves.




ABPE(%) = ([|*j*×(1.23−$E_{RHE}$)|]/*P*)×100




Sketch of the synergic electrocatalytic (EC) and photocatalytic (PC) contributions to EOR process for g-CN-Pt systems. Pt NPs are represented with a Pt(0) core (yellow) and a partially oxidized $PtO_x$ shell (grey).

- **(b, c)** catalytic activities: **g-CN-Pt(60)** < **g-CN** < **g-CN-Pt(15)** < **g-CN-Pt(30)** (beneficial g-CN/Pt NPs interactions)
- **(d)** CA in bare KOH solution (30') and KOH + EtOH aqueous solutions. **Upon addition of EtOH, ⇑ j:** ❶ activity towards EOR than OER. Subsequent j ❶: EtOH consumption down to **≈ constant j**: diffusion-limited process
- **(e)** Tafel slopes: **g-CN** > **g-CN-Pt(60)** > **g-CN-Pt(15)** > **g-CN-Pt(30)** ⇒ **g-CN-Pt(30)** was the best system ⇒ ABPE curves in KOH + EtOH solution **(f)**
- **g-CN-Pt(30):** performances compare favorably with similar (photo)electrocatalysts, and even with various Pt/C systems,

## CONCLUSION

- Fabrication of heterocomposites based on Pt NPs and g-CN through: i) electrophoresis of exfoliated g-CN on C paper; ii) functionalization with **ultra-low amounts of Pt NPs** *via* RF-sputtering from Ar plasmas.
- Key outcomes include: 1) **prevention of Pt NP agglomeration,** g-CN matrix maintaining **high Pt NPs dispersion; 2) improved charge separation** upon Vis light irradiation, leading to better reactivity and stability against poisoning.
- ➲ **effective EOR performances can be achieved with minimal Pt content**, crucial to develop **advanced electrocatalysts for clean energy production**. This research could pave the way for **photo-functional systems in chemical and solar energy conversion**.

## REFERENCES

[1] M. Brugia, D. Barreca *et al.*, *ChemSusChem*, *17*, e202401041 (2024).
[2] M. Brugia, D. Barreca *et al.*, *Surf. Sci. Spectra*, *31*, 024002 (2024).