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Photocatalysts based on oxidized g-C_3N_4 and their photocatalytic applications

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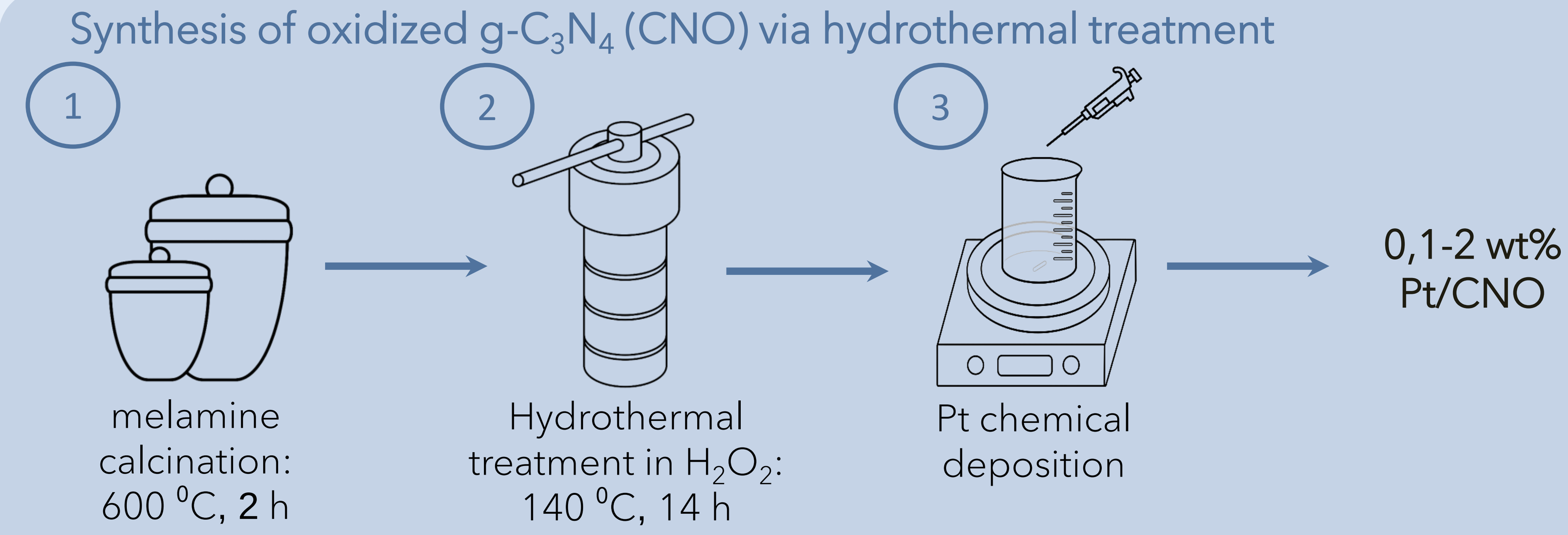
INTRODUCTION

- g-C_3N_4 is a non-metallic semiconductor, which is considered to be one of the most promising materials for photocatalytic applications. Its narrow band gap (~2.7 eV) and negative conduction band position (−1.2 eV vs. SHE) enable visible light absorption and efficient substrate reduction.
- Limitations such as low surface area and poor adsorption can be addressed by oxidative modification, e.g., surface functionalization. The introduction of oxygen-containing groups improves hydrophilicity, surface area, optical properties, electronic conductivity, reactant adsorption, and anchoring of co-catalyst (Pt) particles.
- The effect of hydrothermal treatment on g-C_3N_4 surface properties and photocatalytic activity is considered in this work.

METHODS

Synthesis of oxidized g-C_3N_4 (CNO) via hydrothermal treatment



The photocatalytic performance was investigated in two reactions: H_2 evolution from aqueous organic substrates under LED irradiation (λ_{max}=440 nm) and simulated sunlight (AM 1.5 G), and CO_2 reduction in water vapor atmosphere under LED (λ_{max}=405 nm).

RESULTS & DISCUSSION

- CO_2 reduction reaction (CO_2 RR)

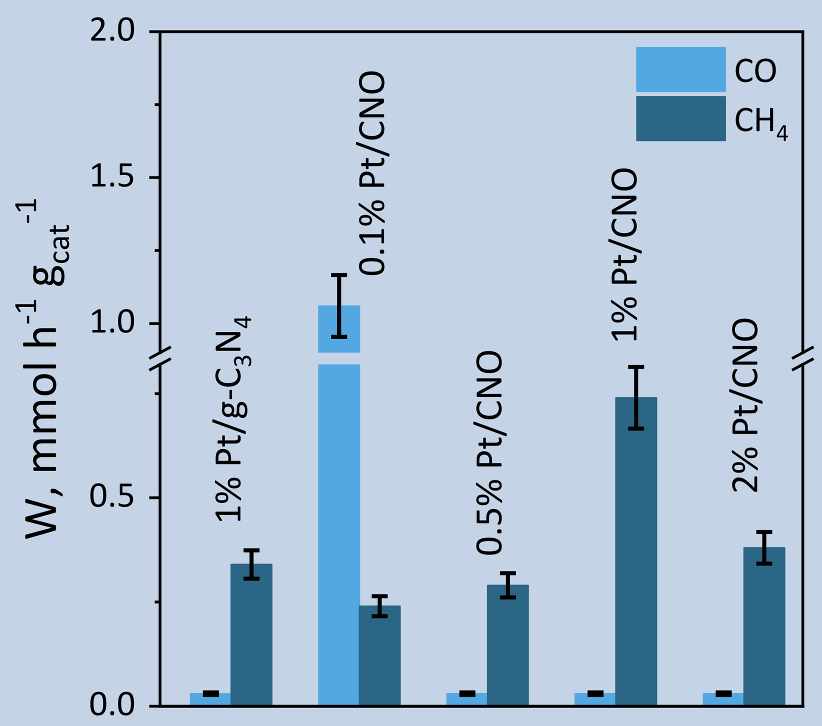
CH_4 is the main product of CO_2 reduction on platinized CNO, except for 0.1% Pt/CNO.

Surface oxidation facilitates electron transfer from CNO to Pt, enabling the 8-electron reduction pathway while suppressing CO formation.

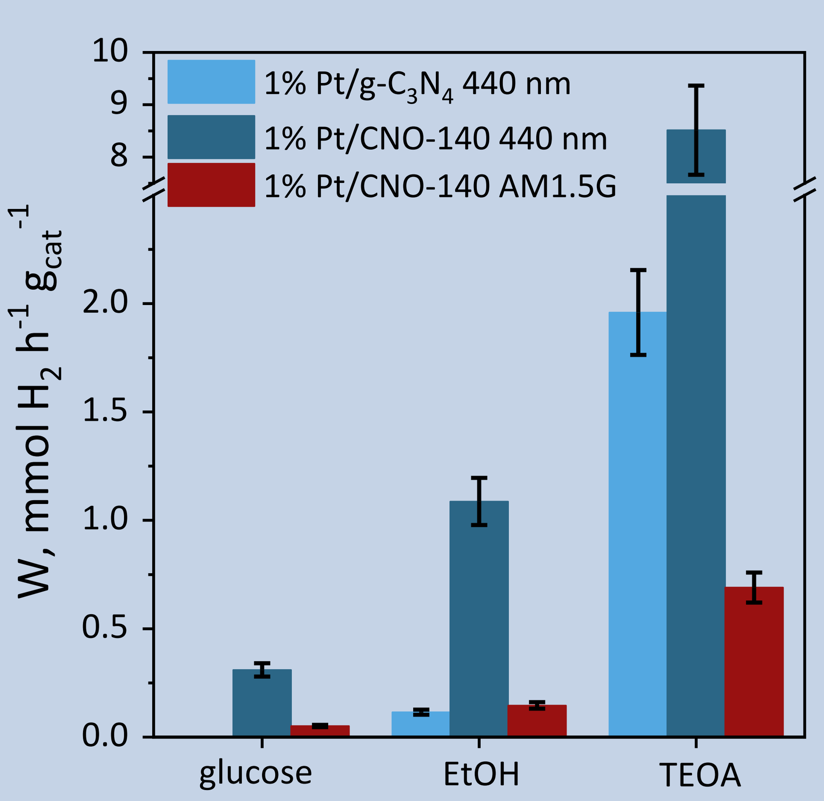
1% Pt/CNO exhibited the highest W_e equal to 5,92 μmol h^{-1} g_{cat}^{-1}.

$$W_e = W(CH_4) * 8 + W(CO) * 2$$

W_e – overall rate of electron consumption



p(CO_2)=1 atm, V(H_2O)=2 mL, LED λ_{max}=405 nm, m_{cat}=30 mg



C_0(glucose) =0.1 M, C_0(EtOH/TEOA) =10 vol%, C_{cat} =0.5 g/L, LED λ_{max}=440 nm

- H_2 evolution reaction (HER)[1]

1%Pt/CNO exhibits the HER rate of 310 μmol h^{-1} g_{cat}^{-1} in glucose solutions, unlike inactive 1% Pt/g-C_3N_4.

Using TEOA and EtOH, the 1%Pt/CNO activity increases by factors of 4.3 and 9.5, respectively, compared to 1%Pt/g-C_3N_4.

Under simulated solar light, HER rate on 1%Pt/CNO reaches 690 μmol h^{-1} g_{cat}^{-1} in TEOA solution.

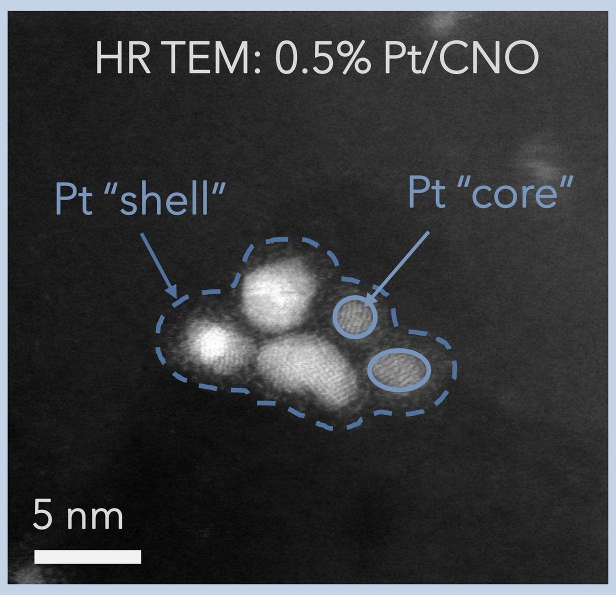
- Photocatalyst characterization[1]

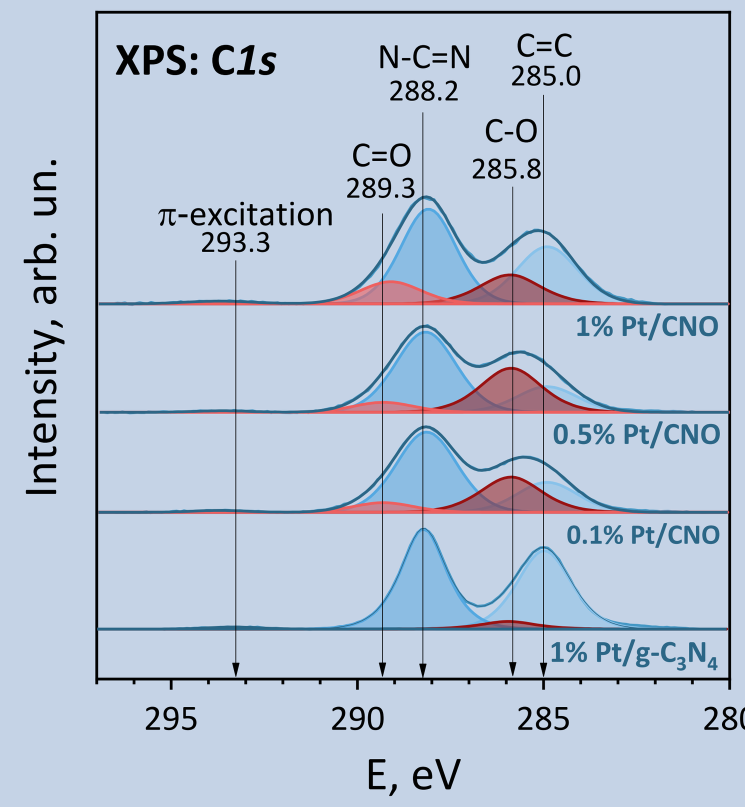
Hydrothermal treatment leads to the partial decomposition of g-C_3N_4 structure, followed by surface functionalization (C=O/COOH, C-OH).

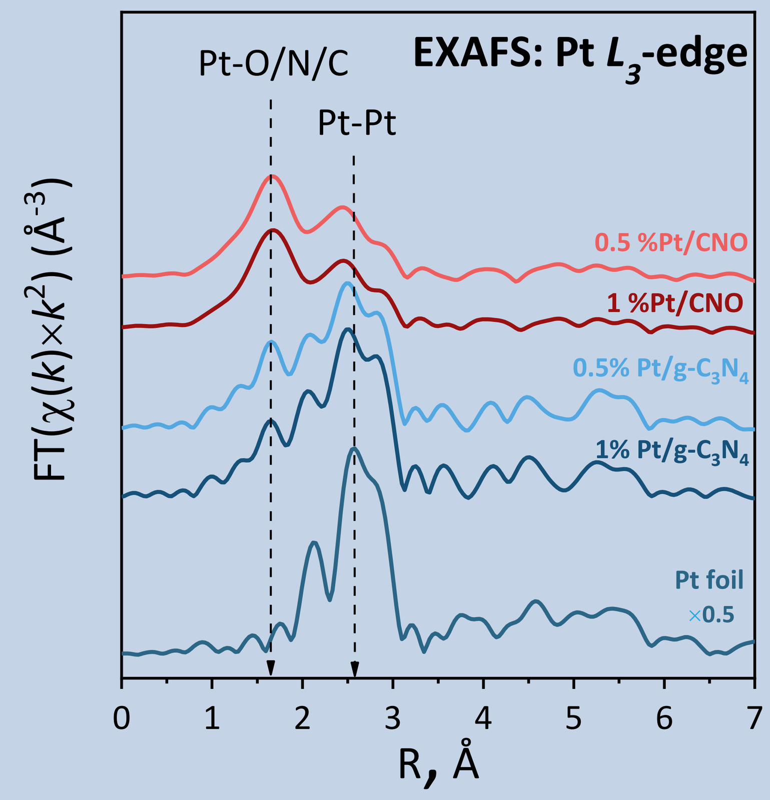
Structural decomposition cause 2,6-fold increase in specific surface area and formation of smaller particulates.

Surface functionalization is responsible for improved adsorption properties and cocatalyst anchoring (strong metal-support interaction, SMSI).

Pt forms core-shell structures on the CNO, where shell – atomically distributed $Pt^{\delta+}$ (δ=0-2), core – Pt^0 nanoparticles.







CONCLUSION

Oxidative hydrothermal treatment has been shown to be a powerful approach for improving the photocatalytic performance of pristine g-C_3N_4. Considering the structural adjustments mentioned, enhanced activity of Pt/CNO is due to hydrophilicity, increased specific surface area, adsorption properties, and faster charge migration between cocatalyst and CNO. Moreover, the SMSI effect observed prevents Pt aggregation during photocatalytic processes and decreased photocatalytic activity.

REFERENCES

[1] Kharina, S.; Kurenkova, A.; Aydakov, E.; Mishchenko, D.; Gerasimov, E.; Saraev, A.; Zhurenok, A.; Lomakina, V.; Kozlova, E. Activation of g-C_3N_4 by Oxidative Treatment for Enhanced Photocatalytic H_2 Evolution. *Appl. Surf. Sci.* **2025**, *698*, 163074, doi:10.1016/j.apsusc.2025.163074.