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Investigation of bimetallic Pt-Co and Pt-Ni catalysts in the photoreduction of nitrate

Anca Vasile, Florica Papa*, Veronica Bratan, Crina Anastasescu, Irina Atkinson & Ioan Balint*
"Ilie Murgulescu" Institute of Physical-Chemistry, Romanian Academy, 202 Spl. Independentei, 060021 Bucharest, Romania
avasile@icf.ro

INTRODUCTION & AIM

- Photocatalytic nitrate (NO_3^-) reduction represents a novel and transformative technology that has the potential to produce harmless gaseous byproducts.
- The photocatalytic denitrification process is frequently accompanied by the creation of undesirable nitrite (NO_2^-) or ammonium (NH_4^+), resulting in poor selectivity for N_2. The catalytic reduction of NO_3^- ions using bimetallic catalysts requires the presence of a noble metal, alongside a promoting transition metal [1]. The transition metal facilitates the reduction of NO_3^- to NO_2^- ions through a redox mechanism, which subsequently results in its oxidation. Furthermore, the noble metal's function is to maintain the transition metal in its lower oxidation states via hydrogen spillover.
- Among oxide photocatalysts, titanium dioxide (TiO_2) has found significant application in photocatalysis and the cleanup of environmental contaminants.
- This study examined the Pt-Co and Pt-Ni bimetallic nanoparticles supported on TiO_2 in the nitrate photoreduction process.

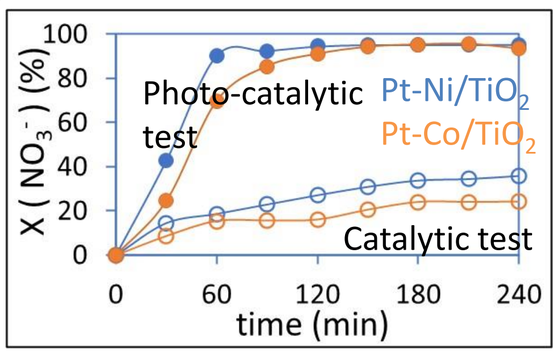
Objectives:

- Comparative catalytic and photocatalytic testing of the prepared catalysts.
- Evaluation of nitrate conversion and selectivity towards reaction products.
- Investigation of hydrogen formation.

METHODS

- **Synthesis**: a successive incipient wetness impregnation method, 2wt.%(Pt-Ni)/TiO_2 and 2wt.%(Pt-Co)/TiO_2, Pt: Ni, Co=1:1 molar ratio; metal precursor salts: $H_2PtCl_6 \cdot 6H_2O$, $Ni(NO_3)_2 \cdot 6H_2O$, and $Co(NO_3)_2 \cdot 6H_2O$.
- **Characterization**: XRD, UV-Vis, H_2-TPR, and PL analyses.
- **Catalytic test**-Reaction conditions: 0.1 g cat.; 25 °C; reactant, NO_3^- (100 mg L^{-1} from $NaNO_3$), 200 mL; H_2.
- **Photo-catalytic test**-Reaction conditions: 0.035 g cat.;18 °C; reactant, NO_3^- (100 mg L^{-1} from Na NO_3), 70 mL; Ar, 20 cm^3 min^{-1}; UV lamp. The GC-TCD monitored the amount of H_2 generated under continuous UV irradiation at 30-minute intervals. The concentrations of NO_3^-, NO_2^-, and NH_4^+ in the solution after tests were analyzed using ion chromatography.
- **Preliminary test for water splitting**: Reaction conditions: 0.035 g cat.;18 °C; H_2O, 70 mL; Ar, 20 cm^3 min^{-1}; UV lamp.

RESULTS & DISCUSSION



NO_3^- conversion as a function of time

- In the photocatalytic reaction, NO_3^- conversion exceeds 93% for both catalysts.
- Additionally, the selectivity for NH_4^+ is significantly reduced compared to that recorded in the catalytic test.
- In the photocatalytic tests, the selectivity toward N_2 was around 68%.

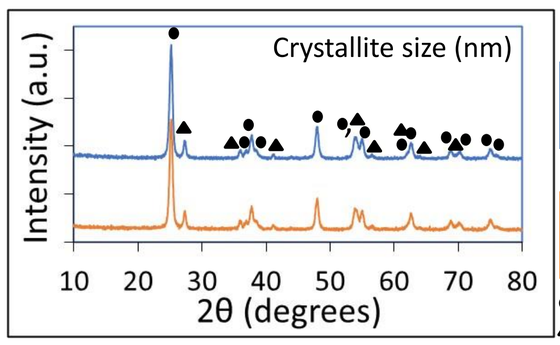
	Pt-Ni/TiO_2		Pt-Co/TiO_2	
Nitrate reduction	Catalytic	Photo-Catalytic	Catalytic	Photo-Catalytic
X(NO_3^-)	35.74	95.31	24.01	93.32
S(NO_2^-)	33.00	30.07	18.40	31.50
S(NH_4^+)	22.90	0.96	31.50	0.60
S(N_2)	44.10	68.34	50.10	67.90

NO_3^- conversion (X%) and NO_2^-, NH_4^+, and N_2 selectivity (S%) after 4 h

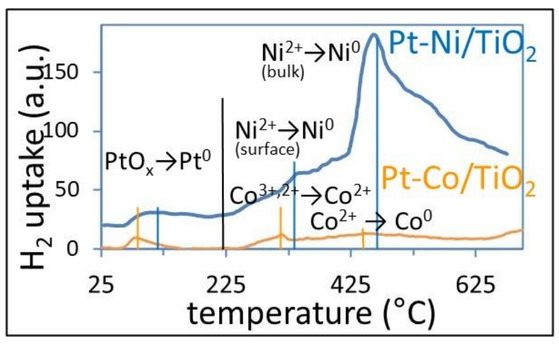
- The N_2 amounts were calculated using a mole balance, assuming that only NO_2^- and NH_4^+ are formed as side products.

Acknowledgements:
Romanian Academy – Vietnam Academy of Science and Technology Joint Research Project 2026-2027, An innovative Approach to Nitrogen Compound Removal via Reduction and Ammonia Nitrogen Ozonation Using Bimetallic Nanoparticles-Based Heterostructures

- Pt, Co, or Ni do not modify the essential crystalline phase of TiO_2.



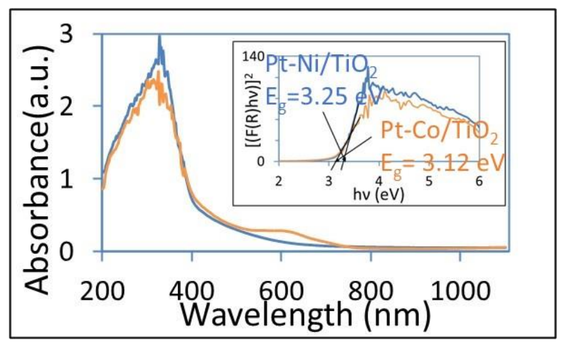
X-ray diffraction (XRD) pattern



H_2-TPR profiles

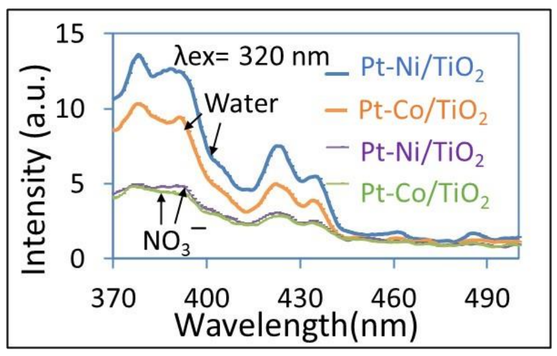
- Pt species are reduced at a lower temperature for Pt-Co/TiO_2 (80 °C) compared to Pt-Ni/TiO_2 (100 °C).
- Presence of different Ni and Co species on the TiO_2 support (different reduction peaks: Ni→340 and 463 °C; Co→315 and 450 °C.

- The absorption bands within the observed spectrum of 550–620 nm can be attributed to metal charge transfers (O^{2-} – Co^{2+}) occurring on Co_3O_4 nanoparticles.
- The addition of metals to TiO_2 results in an increased capacity for visible light absorption due to a reduction in the band gap (E_g).

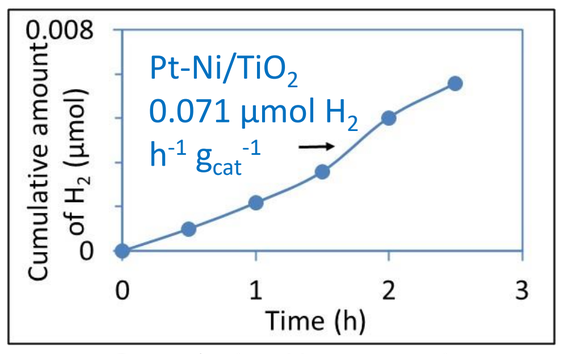


Kubelka-Munk light absorption curves (Inset: Reflectance [F(R)] spectra)

- The photogenerated charges are being used in a chemical reaction induced by the presence of NO_3^- at the catalyst surface.
- Pt-Ni/TiO_2 is capable of producing H_2 via the photocatalytic water splitting process.



Comparative PL spectra of the catalysts in water and NO_3^- aqueous solution



Cumulative H_2 generation versus time

CONCLUSION

- The conversion of NO_3^- exceeds 93% and 95% in photocatalytic experiments, respectively.
- Selectivity for benign N_2 is significantly improved following photocatalytic nitrate reduction, while selectivity for NH_4^+ is diminished.
- The formation of hydrogen was observed; the hypothesis of its use in the reduction reaction is proposed.

FUTURE WORK / REFERENCES

- Evaluation of photocatalytic activity under visible light irradiation.

[1] Vasile, A., Papa, F., Bratan, V., Munteanu, C., Teodorescu, M., Atkinson, I., ... & Balint, I. (2022). Water denitration over titania-supported Pt and Cu by combined photocatalytic and catalytic processes: Implications for hydrogen generation properties in a photocatalytic system. J. Environ. Chem. Eng., 10(2), 107129.