H_2 production via light-assisted ammonia decomposition over xNi-CeO_2 catalyst

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Highlights

- Visible light accelerates ammonia decomposition reaction at lower temperatures
- Catalyst with 3wt.% of Ni deposited is optimal
- Illumination lowers the apparent activation energy from 100 to 30 kJ/mol
- Illumination with 500 nm photons is the most efficient

1. Introduction

Ammonia decomposition offers a CO_x-free pathway for hydrogen production. Photo-thermal catalysis can enhance NH_3 conversion by synergistically coupling light and heat to accelerate reaction kinetics. [1] Ni-based catalysts represent a cost-efficient alternative to Ru, demonstrating high activity, stability and resistance to sintering.[2] Supporting Ni nanoparticles on high-surface area CeO_2 nanorods leads to a highly active catalyst with strong metal-support interactions that improve charge transfer, facilitate N-H bond activation or N_2 desorption.[3] This research analyzes the light-driven reaction pathways through catalytic experimentation, kinetic analysis, structural characterization and application of *in-situ* spectroscopy to unravel new insights into the low energy barrier ammonia decomposition.

2. Methods

CeO_2 nanorods were synthesized from nitrate precursor $Ce(NO_3)_2$*$6xH_2O$, followed by different amounts of Ni (1, 3 and 15 wt.%) deposition via deposition precipitation method. Comparison of thermal and photo-thermal (using only visible light) performance of the catalysts was tested using 10% NH_3 in Ar. The analyzed temperature varied depending on the process but was kept between 300 and 450 °C and ambient pressure. Data for catalytic activity of the analyzed catalysts was obtained by gas chromatography. Other detailed characterization will include XRD, SEM-EDX, in-situ UV-Vis as well as DRIFT spectroscopy.

3. Results and discussion

XRD and SEM analyses showed that nickel was effectively deposited and dispersed on ceria nanorod support in the form of small nanoparticles.
Initial catalytic tests showed that all three catalysts exhibited similar H_2 rates under thermal-only conditions, with 3Ni-CeO_2 showing the highest activity, Figure 1A. In contrast, light-assisted operation enabled substantial activity at just 350 °C (100 °C lower compared to dark conditions).

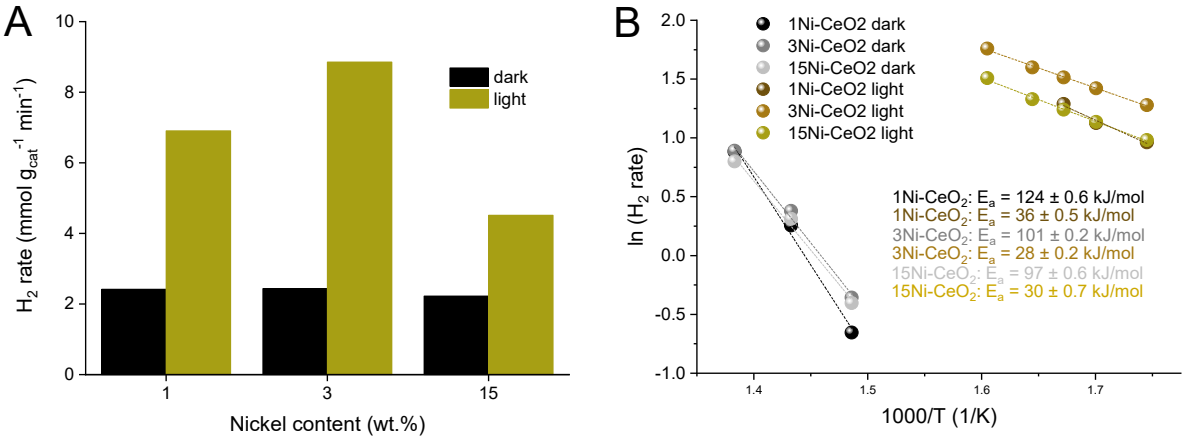

Figure 1. Figure 1: A) H_2 rate comparison under thermocatalytic (450 °C) and light-assisted (350 °C) conditions for different nickel loadings deposited on CeO_2 nanorods. B) Apparent activation energy values (E_a) under dark and light-assisted conditions.

All catalysts outperformed their dark-condition counterparts, with 3Ni-CeO_2 achieving almost a forth fold increase in H_2 production. Additionally, Arrhenius plots (Figure 1B) show a substantial decrease in apparent activation energy upon illumination (from about 100 to 30 kJ/mol), indicating a change in the reaction mechanism. Wavelength dependent photocatalytic experiments showed that illumination with 500 nm photons is the most efficient for ammonia decomposition.

4. Conclusions

xNi-CeO_2 catalysts can effectively harness visible light to accelerated ammonia decomposition, demonstrating a clear photo-thermal synergy beyond thermal-only operation. By combining photo-driven effects with catalytic heat-driven pathways at the Ni-CeO_2 interface, this material can enhance the reaction rate under otherwise identical reactor conditions, effectively reducing the temperature (or energy input) required to achieve a given H_2 productivity. As a result, photo-thermal ammonia decomposition emerges as a promising alternative to conventional thermal cracking used industrially, with potential to lower operational costs through more moderate operating conditions. Overall, these findings support integration of visible light utilization for NH_3 conversion as a part of scalable, zero-carbon hydrogen production pathways.

References

The reference format is provided below [1 – 3]. [Times New Roman 10].

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Keywords
Photo-thermal catalysis, activation energy, visible-light, xNi-CeO_2 catalyst