Cs-enhanced Ru-based catalysts for low-temperature NH_3 decomposition

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Introduction

In this study, ruthenium-based cesium-promoted catalysts were synthesized using different ruthenium loading and Ru/Cs molar ratios. The obtained materials were characterized via X-Ray Diffractometry and X-ray Photoelectron Spectroscopy. Then, the catalytic activity of these catalysts was tested for ammonia decomposition in a fixed bed reactor.

Catalysts synthesis

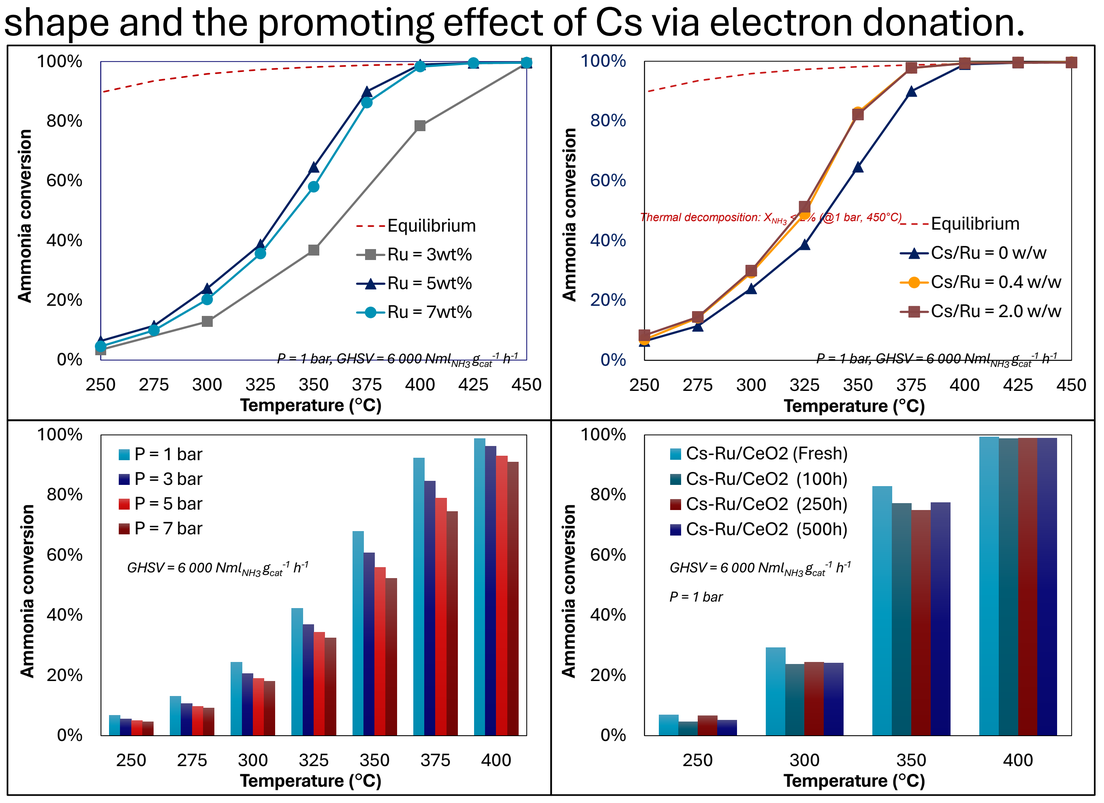
The catalysts employed in this work were prepared as follows:

- ❖ Metal precursors dissolution in ethylene glycol;
- ❖ Heating up to 110°C and reaction for 2 hours;
- ❖ Particles cleaning and separation;
- ❖ Drying (12h, 120°C) and calcination (4h, 550°C).

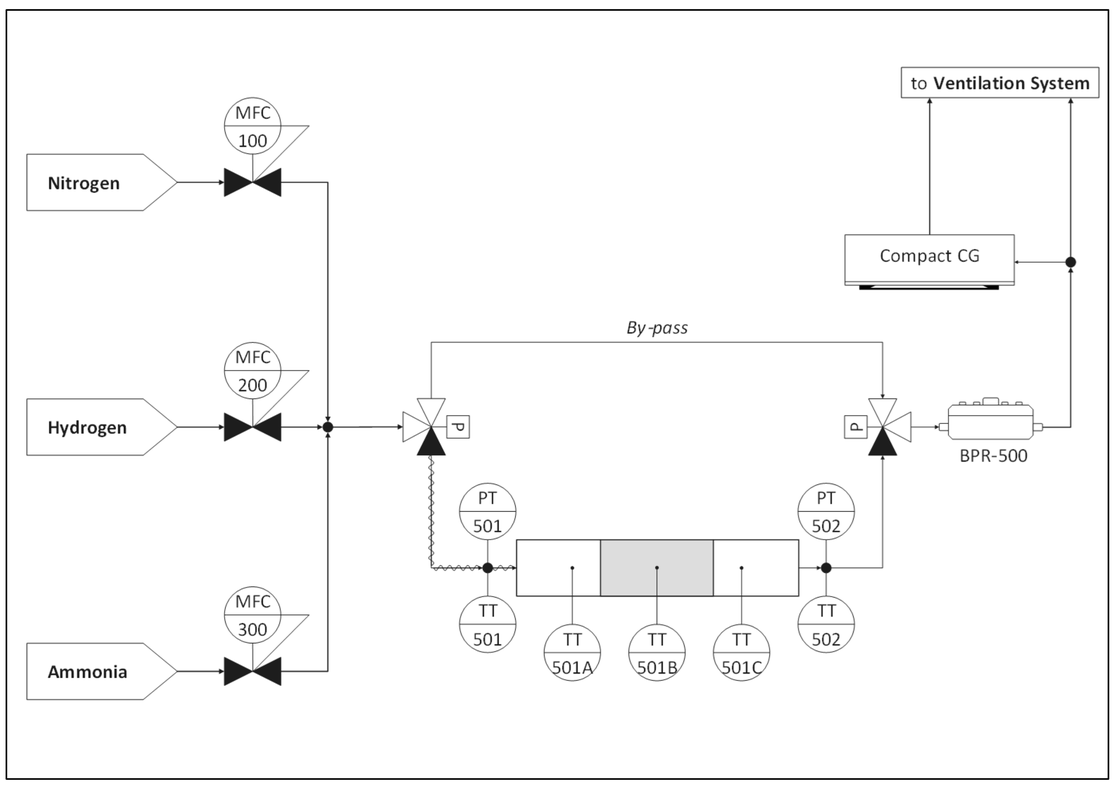
Catalyst	Code	Ruthenium (wt%)	Cesium (wt%)
3wt% Ru/CeO_2	3Ru	3.0	-
5wt% Ru/CeO_2	5Ru	5.0	-
7wt% Ru/CeO_2	7Ru	7.0	-
2wt% Cs – 5wt% Ru/CeO_2	2Cs5Ru	5.0	2
10wt% Cs – 5wt% Ru/CeO_2	10Cs5Ru	5.0	10

Results

The results showed the higher catalytic activity when ruthenium loading is about 5 wt% and Ru/Cs molar ratio is about 0.4 wt%. This can be ascribed to both the Ru cluster shape and the promoting effect of Cs via electron donation.



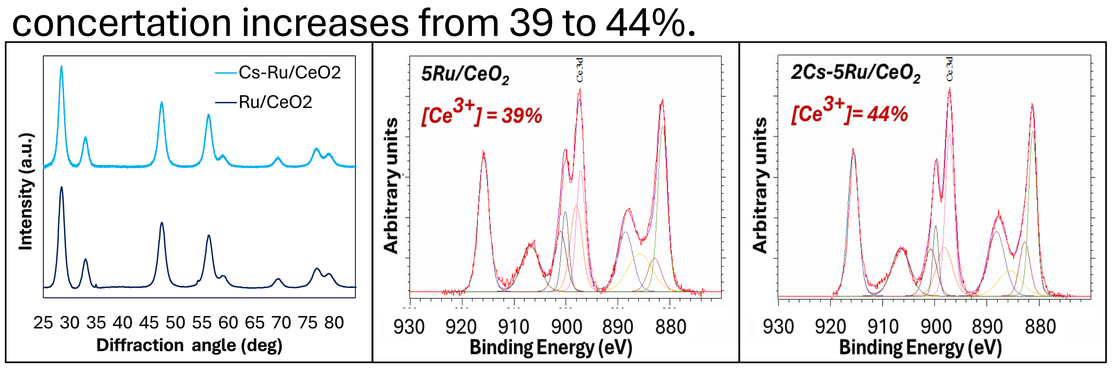
Experimental setup



Characterizations

The XRD patterns for both promoted and non-promoted catalysts revealed the characteristic peaks of cerium oxide cubic lattice and no additional peaks assignable to ruthenium or cesium have been detected.

The cerium 3d XPS spectra were deconvoluted in ten peaks, of which six can be assigned to Ce^{4+} and four can be assigned to Ce^{3+}. The concentration of Ce^{3+} atoms is related to the oxygen vacancies formation and, with the addition of cesium, this concertation increases from 39 to 44%.



Conclusions

- ➢ Ru-based-Ce-supported catalysts where successfully synthesized via polyol reduction method.
- ➢ $5Ru/CeO_2$ allowed an ammonia conversion reaching the equilibrium already between 375 and 400°C (1 bar, 6 000 $Nml_{NH_3}\ g_{cat}^{-1}\ h^{-1}$).
- ➢ The addition of cesium to the formulation resulted in an increase of NH_3 conversion by 33% (350°C, 1 bar, 6 000 $Nml_{NH_3}\ g_{cat}^{-1}\ h^{-1}$).
- ➢ NH_3 conversion decreased less then 1% over 500 hours of test (400°C, 1 bar), proving high stability of the catalyst over time.

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