Assessment of low PGM catalyst for Ammonia cracking

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Significance and Relevance

This study highlights the potential of advanced screening techniques to identify cost-effective alternatives to high-cost catalysts for low-temperature ammonia decomposition. By incorporating yttrium (Y) and zirconium (Zr), the ruthenium (Ru) content was successfully halved while maintaining high catalytic activity with minimal performance loss. Detailed investigations revealed that substituting Ru with Y or Zr did not impact the apparent activation energy of the catalysts, demonstrating their viability as economical and efficient options for ammonia cracking applications.

Preferred and 2nd choice for the topic: H2 storage and transportation, green H2 production, hydrogen vectors; Catalysis to electrify the chemical production
Preferred presentation: (Oral only / Oral preferred or Short Oral / Poster): Oral/Short Oral

Introduction and Motivations

Hydrogen (H_2) is a sustainable and long-term energy carrier with the capacity to meet growing global energy demands.[1,2] It can be applied across sectors such as transportation, industry, and heating systems. However, the volatile and flammable nature of hydrogen poses significant challenges, particularly in distribution via pipelines due to the risk of leakage.[3] Ammonia (NH_3) offers a promising alternative as a hydrogen carrier, with a high hydrogen content (17.6 wt%), carbon-free properties, and ease of storage at low pressure (~7.5 atm at 300 K).[4,5] The endothermic NH_3 cracking process can reliably generate hydrogen for clean energy technologies like fuel cells.[6] Despite this potential, the cost and low energy efficiency of current NH_3 cracking catalysts hinder their large-scale adoption.[5,7,8] Traditional catalysts, predominantly based on expensive late-transition metals like ruthenium, are unsuitable for widespread use.[9] Furthermore, current experimental studies lack sufficient depth to identify cost-effective alternatives to ruthenium.[10]

Materials and Methods

The samples were synthesized through a wet impregnation method, beginning from the precursors: ruthenium (III) chloride trihydrate, yttrium (III) nitrate hexahydrate, zirconium (IV) oxynitrate hydrate and strontium nitrate, supported on γ- Al_2O_3. Following dissolution in deionized water and vigorous stirring of the mixture and heating at 80°C for 6 hours, the impregnated supports were dried at 110°C for 12h, then calcined in air for 2h at 200°C and 3h at 550°C. Subsequently, the catalysts were reduced in hydrogen at 550°C for 3h. The characterization of the catalysts was conducted through the utilization of XRD, TEM, N_2-TPR, and NH_3-TPR techniques. The catalytic activity measurements for the decomposition of ammonia were evaluated in a quartz tubular reactor operating under atmospheric pressure and fed with a helium-ammonia mixture (2000 ppm NH_3) with a constant flow rate (100 mL/min). All experiments were conducted within the temperature range of 250 to 500°C, utilizing a catalyst loading of 100 mg.

Results and Discussion

The Ru content was halved and partially replaced with secondary elements to evaluate their impact on catalytic performance. To isolate the effect of the substitutions, the total metal loading was maintained at 4% across all samples. When Ru content was reduced to 2%, catalysts incorporating Y and Zr demonstrated the highest activity, achieving nearly 100% NH_3 conversion at 425°C. In contrast, Sr substitution led to a decline in catalytic efficiency, as active Ru sites were replaced by less effective transition metal sites. Structural and electronic properties of Ru sites were unaffected, as indicated by the Arrhenius plots, which showed no change in apparent activation energy across the catalysts. The activation energy values (17.4–20.6 kcal/mol) align closely with those reported for Ru/Al_2O_3 catalysts in similar studies.[11]

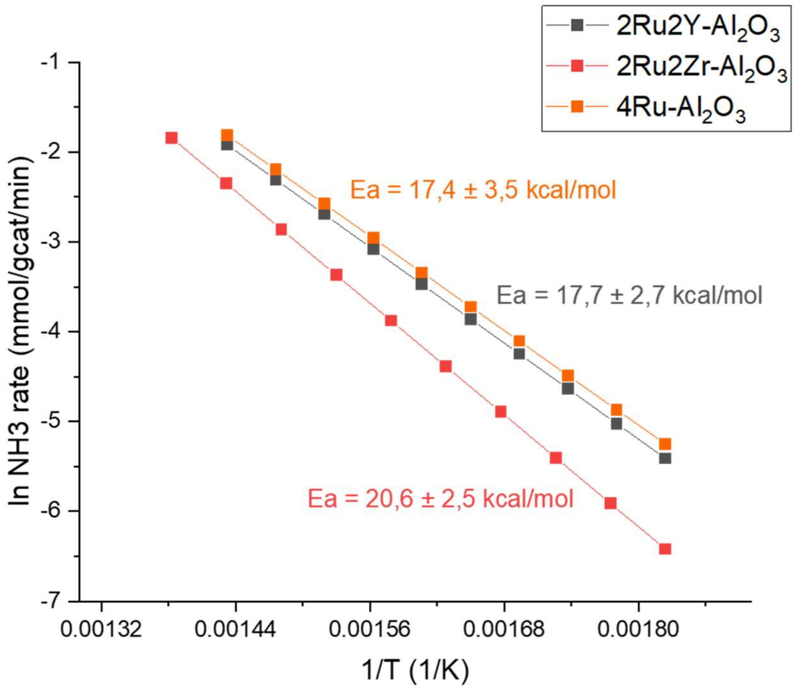

Figure 1. Arrhenius plot for RuY, RuZr and 4Ru. Experimental conditions: p=1 bar, NH3 concentration = 2000 ppm, total flow rate = 100 mL/min, catalyst amount = 100 mg.

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