Plasma-Catalytic Ammonia Dissociation for Hydrogen Release

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Abstract: In this contribution, we examine the thermal and plasma effects on NH_3 dissociation rates in a Dielectric Barrier Discharge (DBD), packed-bed reactor, using Fourier Transform Infrared Spectroscopy (FTIR). Preliminary findings indicate that the plasma significantly enhances the degree of dissociation, achieving near 100% dissociation at reduced temperature compared to the thermal case.

1.Introduction

Hydrogen, a clean and versatile fuel, can revolutionize energy systems by offering a zero-emission alternative to fossil fuels. However, its physical and chemical properties make storage and handling challenging. Conversely, ammonia, with 17.6% H_2 content by weight, is an excellent hydrogen carrier and remains liquid at room temperature under moderate pressure [1].

While conventional ammonia dissociation utilizes metal catalysts at relatively high temperatures, recent studies show plasma-based electrical decomposition achieves high dissociation rates (~87%) [2]. In this study, we use a DBD plasma in combination with a furnace to examine the effects of temperature and non-thermal plasma on ammonia dissociation rates. Since ammonia dissociation is mildly endothermic, the furnace aids in pre-activating the reaction.

2. Methods

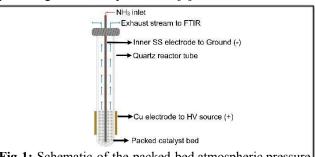
A Dielectric Barrier Discharge (DBD) plasma is employed to dissociate NH₃ within a thermally operated packed bed cylindrical reactor [Figure 1]. Metal catalysts (Cu, Ni, Fe, Ru, Pt) are pre-treated by baking in hydrogen for two hours before the experiments. FTIR spectra of exhaust gases are measured at various temperatures with the plasma on and off to assess thermal and plasma effects on NH₃ dissociation. The AC peak-to-peak voltage and frequency supplied to the reactor are kept constant for fair comparison. XPS analysis of the catalyst at different stages (fresh, after baking, and after plasma exposure) is conducted to determine the chemical composition and understand surface adsorption mechanics.

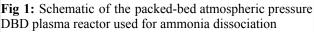
3. Results and Discussion

Figure 2 shows the change in ammonia dissociation with temperature and plasma at constant AC voltage and frequency for a 5% (by weight) copper catalyst on alumina support. Thermal dissociation is negligible at lower temperatures but increases sharply after 500°C. The synergistic effect of plasma at high temperatures significantly surpasses the thermal effect due to plasma species lowering the catalyst's activation energy. The presence of DBD plasma also enhances ammonia dissociation at lower temperatures (<500°C), which improves energy efficiency and reduces catalyst sintering by allowing dissociation to occur at lower temperatures.

4. Conclusion and Future Work

While we conclusively show that plasma-enhanced ammonia dissociation works markedly better than purely thermal dissociation, the reaction pathways are not fully understood. Future work will involve mass spectroscopy to study the reaction mechanics and the contribution of the plasma in improving dissociation rates. XPS on the catalysts at various stages (thermal vs. thermal+plasma) will be conducted to confirm that plasma reduces nitrogen poisoning on the catalyst surface [3].





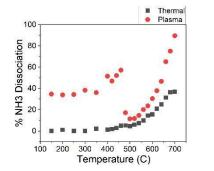


Fig 2: Change in ammonia dissociation with temperature and plasma at constant AC voltage (13kV) and frequency (25kV), using Cu/Al_2O_3 catalyst

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References

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