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Screening of supported metal oxides for dielectric barrier discharge plasma-catalytic ammonia production

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PURPOSE OF THE ABSTRACT

Ammonia, primarily utilized in agriculture as the main feedstock for fertilizer production, is gaining attention as a potential energy carrier across various sectors. Achieving this transition, however, calls for the development of an environmentally sustainable alternative to the energy-intensive Haber-Bosch process. Plasma-catalytic systems have emerged as a promising solution, offering compatibility with intermittent renewable energy sources and sustainability for decentralized, small-scale production. Despite extensive research effort, these systems face significant challenges, most notably their low energy efficiency, which currently still falls short of the benchmark set by the Haber-Bosch process.

In recent years, there has been growing interest in utilizing of porous materials for the design of supported metal catalysts in plasma-catalytic ammonia production. Micro- and mesoporous supports have demonstrated significant benefits, particularly by enhancing the diffusion of plasma-activated species to active sites1. In addition, packing of the plasma reactor with a porous material can further improve the efficiency of ammonia production through the shielding effect2. This phenomenon occurs when the produced ammonia molecules diffuse into the pores of the material and are therefore protected from dissociation in plasma, which is one of the main challenges in plasma-catalytic system optimization. By limiting of ammonia decomposition, porous supports could contribute to higher yields and improved overall efficiency. To date, various supported metal catalysts, primarily containing Co, Ni, Ru, and Fe, have been explored for plasma-catalytic ammonia synthesis. While these transition metals exhibit promising catalytic activity, most of them are prone to partial oxidation in air due to the high surface energy of their nanoparticles3. This motivated us to investigate supported metal oxides as alternative catalysts for dielectric barrier discharge (DBD) plasma-catalytic ammonia production. To harness the beneficial properties of porous materials, USY zeolite was selected as a support due to its high specific surface area, stability, and well-defined pore

structure. While wetness impregnation is the most commonly used method for synthesizing supported metal catalysts, the adapted precipitation method was employed in this research. To systematically evaluate the effect of the synthesis method on catalytic performance, the best performing catalyst was synthesized using both wetness impregnation and precipitation method. Characterization via powder X-ray diffraction and scanning electron microscopy analysis revealed that metal oxide particles of the catalyst prepared by precipitation method are smaller, which correlated with higher catalytic activity compared to the catalyst prepared via wetness impregnation. In terms of activity, cobalt and nickel oxide exhibited the highest performance, in line with previously observed activity trends for transition metal catalysts.

FIGURES

FIGURE 1

FIGURE 2

KEYWORDS

green ammonia | plasma catalysis | dielectric barrier discharge | nitrogen fixation

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