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Kinetic Study of Ammonia Synthesis by Using Non-Equilibrium Plasma and Pulzed Heating

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Plasma-assisted catalysis is one possibility for ammonia synthesis that takes advantage of the expanding increase of renewable electricity from solar and wind power. This approach utilizes excited gaseous molecules or new reactive species formed in a non-equilibrium plasma, along with a catalyst to enable increases in the activity and selectivity for carrying out desirable chemical reactions at low temperature and pressure conditions. We investigated ammonia synthesis in a coaxial dielectric barrier discharge (DBD) plasma reactor with different catalyst particles at atmospheric pressure [1]. Gas-phase species were monitored *in-situ* using an electron impact molecular-beam mass spectrometer (EI-MBMS). Gas-phase species NNH and N_2H_2 were first identified under common conditions of plasma-assisted ammonia synthesis and were present at levels comparable to NH_3 in the plasma discharge. These observations point to the importance of NNH and N_2H_2 in plasma-assisted surface reactions in ammonia synthesis.

Moreover, we recently presented a non-equilibrium, continuous synthesis technique that uses pulsed heating and quenching (PHQ, for example, 0.02 s on, 1.08 s off) using a programmable electric current to rapidly switch the reaction between high (for example, up to 2,400 K) and low temperatures [2]. The rapid quenching ensures high selectivity and good catalyst stability, as well as lowers the average temperature to reduce the energy cost. The ammonia synthesis was also studied by using the PHQ technique and the key synthesis chemistry was discussed in the PHQ-enabled ammonia synthesis processe. It achieves a stable and high synthesis rate of about 6,000 μmol $g_{Fe}^{-1}h^{-1}$ at ambient pressure for >100 h using a non-optimized catalyst. This study establishes a new model towards highly efficient non-equilibrium thermochemical synthesis.

References

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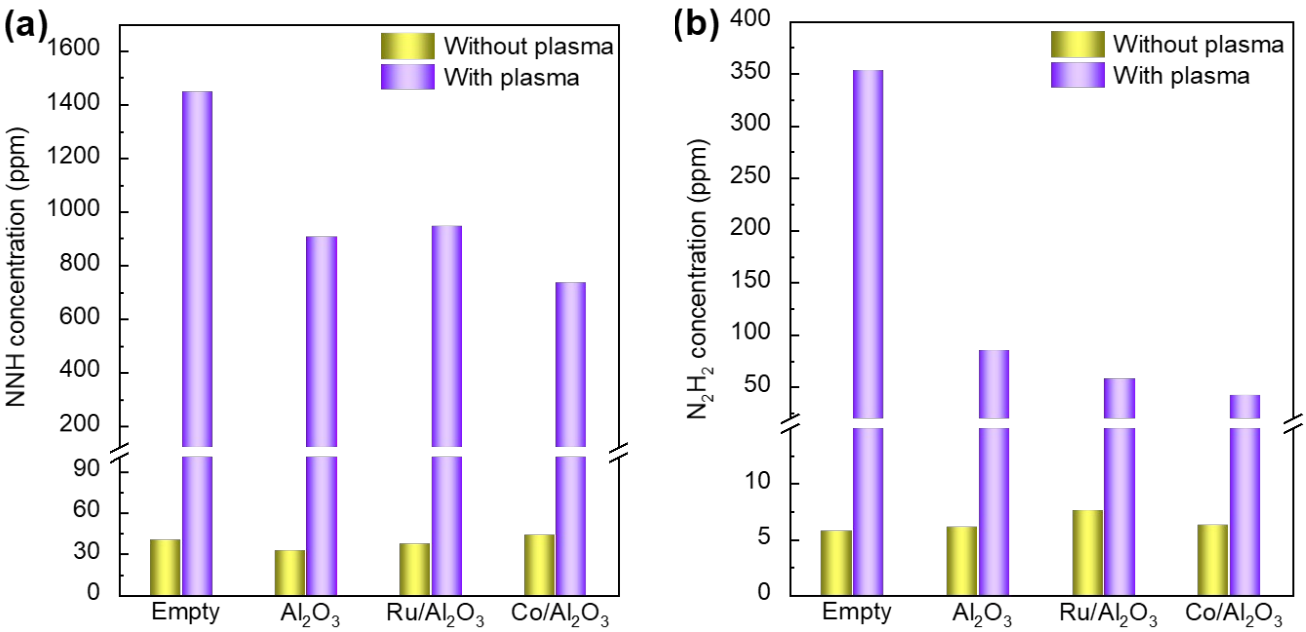

Figure 1 Concentrations of gas-phase (a) NNH and (b) N_2H_2 detected by MBMS during DBD plasma-assisted NH3 synthesis. Data were obtained for an empty reactor and with the reactor loaded with porous γ-Al2O3, 5 wt% Ru/γ-Al2O3, and 5 wt% Co/γ-Al2O3 particles. See Figure 2 caption for the reaction conditions.