Model analysis and experimental determination of noble gas metastable for plasma-assisted catalysis of ammonia

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A zero-dimensional (0D) kinetic model has been developed for N_2-H_2-noble gas (He or Ar) atmospheric pressure plasmas in our dielectric barrier discharge (DBD) reactors, based on the ZDPlasKin plasma kinetics solver [1]. The goal of this kinetic modeling is to understand both the volumetric and surface (Ru or Fe on γ-Al_2O_3) reactions related to plasma-assisted catalysis as a potentially lower energy alternative to the standard Haber-Bosch process. Results have shown that the dominant metastables, Ar_{meta} (1s5 and 1s3) and He(2^3S), formed via electron-impact excitation, can support N atom and ion formation through the Penning excitation and ionization reactions $N_2 + Ar_{meta} \rightarrow 2N + Ar$ and $N_2 + He(2^3S) \rightarrow N + N^+ + He + e$. These N species play an important role in NH_3 formation via Eley-Rideal reactions [2].

We have performed optical emission spectroscopy to measure He spectra from He DBD plasmas for near atmospheric pressures (APs) up to 200 Torr. Two reactor configurations, coaxial cylinder and parallel plates, were examined for an applied AC voltage of 1-5 kV at 20 kHz. The obtained He line intensity ratios were investigated using a He collisional-radiative (CR) model to determine the plasma parameters T_e and n_e, and also He(2^1S) and He(2^3S) densities. The He CR model, originally developed by Goto [3], was integrated with the radiation trapping effect [4] and AP reactions such as heavy-particle collisions between He atoms and molecules [5]. Our model analysis showed that He excited-state populations for AP plasma are strongly affected by the population density of metastables He(2^1S) and He(2^3S). The results suggest the possibility of He CR modeling as a method of estimating the He(2^3S) density in order to understand the ammonia formation process.

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