Thesis: Plasma-Assisted Catalysis for Ammonia Synthesis
by Stephane Sartzetakis
Dept of Chemical and Biological Engineering
Princeton University

Abstract

Ammonia (NH_3) is one of the most popular industrial products in the world, and yet the global production of NH_3 relies on the energy- and carbon-intensive Haber-Bosch process. With the world beginning to set ambitious targets for reducing carbon emissions, and the demand for NH_3 only rising, there is a clear need for environmentally friendly alternatives to the NH_3 production process. Non-thermal plasma-assisted catalysis is being investigated as a possibility because it can operate at ambient temperatures and pressures and is potentially compatible with intermittent renewable energy sources. The current major drawback, however, is that the current conversion efficiencies of N_2 to NH_3 observed thus far are not high enough to be commercially viable. Since plasma-assisted catalysis is significantly different than thermal catalysis due to the altered operating conditions and the presence of the plasma environment, research is required to determine effective catalysts. Existing literature has shown that traditionally inert catalyst supports are capable of producing NH_3 through the process of plasma-assisted catalysis. Additionally, there are studies indicating that high surface area and ordered pore structure are impactful characteristics in a catalyst. Based on these observations, we synthesized and tested a 10 wt. % Al coated SBA-15 composite and compared its performance against a set of commercial catalyst supports (alumina, silica, SBA-15, zeolites, and quartz wool). The performance metrics utilized herein were Lissajous analysis of plasma electron density and mass spectrometry analysis of N_2 conversion. Despite producing a plasma environment with a lower electron density, the synthesized 10 wt. % Al composite had the highest N_2 conversion values observed. Since there was no clear correlation between the plasma electron density and the N_2 conversion for all the catalysts tested, it was concluded that plasma-assisted catalysis is a synergistic process, meaning that the plasma and catalyst surface have respective properties that may combine to be beneficial beyond their typical effects. The successful performance of the 10 wt. % Al composite over the commercial catalysts supports the future study of synthesizing tunable, highly ordered porous materials with high surface areas. Additional experiments were performed on zeolites and quartz wool. The observed N_2 conversions for both the zeolites and the quartz wool were lower than the reactor with the empty catalyst bed.