Nonequilibrium Plasma Kinetics in a Heated Flow Reactor Excited by a Ns Pulse Discharge

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Abstract

A heated plasma flow reactor, with the flow excited by a ns pulse discharge, is used to study kinetics of plasma chemistry and plasma-assisted catalysis at well-characterized conditions. The discharge generates a diffuse volumetric plasma, with ample optical access for laser diagnostics. This approach is employed for several experiments in nitrogen, O_2-Ar, N_2-H_2, and N_2-O_2 mixtures.

Time-resolved, absolute number densities of metastable nitrogen molecules, $N_2(A^3\Sigma_u^+)$, and molecular ions, N_2^+, are measured in nitrogen and N_2-NO mixtures, at temperatures of T=300-1000 K. The measurements are made by Tunable Diode Laser Absorption Spectroscopy (TDLAS) and pulsed UV Cavity Ring Down Spectroscopy (CRDS). The results illustrate the generation and decay of $N_2(A)$, a precursor for metastable excited atoms, during the discharge burst. Time-resolved N_2^+ measurements in the afterglow indicate the effect of associative ionization of excited species accumulated during the discharge.

Kinetics of O_2 vibrational excitation is studied during the O atom recombination in an O_2-Ar mixture partially dissociated by a burst of ns discharge pulses in a flow reactor, at T=400-800 K. Time-resolved vibrational level populations $O_2(v=8\text{-}20)$, are measured by ps Laser Induced Fluorescence (LIF). The O atom number density is measured by ps Two-Photon Absorption LIF (TALIF). The results exhibit a rapid initial decay of $O_2(v)$ populations generated in the discharge, on ~10 μs time scale, mainly due to V-T relaxation by O atoms. A significantly slower decay, on a time scale of ~ 1 ms, indicates the $O_2(v)$ generation by chemical reactions initiated by the O atom recombination.

Kinetics of plasma-catalytic ammonia generation is studied in a "hybrid" ns pulse discharge overlapped with a sub-breakdown RF waveform, used to isolate the effect of vibrational excitation. Time-resolved N and H atom number densities are measured by ns TALIF, and N_2 vibrational temperature measured by ns CARS. The ammonia yield is measured with and without Ni, Ru, and Rh catalysts placed in the reactor. Adding the RF waveform to the ns pulse discharge increases the ammonia yield by up to 25%. Similar effect of RF augmentation is detected on the NO yield in N_2-O_2 mixtures, processed without the catalyst. Both results suggest the enhancement of plasma chemical processes by vibrational excitation of N_2.