Diagnostics and modeling of plasma assisted combustion kinetics

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Recent experimental studies of repetitive nanosecond pulse discharges demonstrate their significant potential for plasma assisted ignition and combustion. The main advantage of using these discharges for ignition is efficient generation of electronically excited and radical species, such as O and H atoms, as well as OH. In recent experiments, time-resolved temperature, N\textsubscript{2} vibrational level populations, absolute O, H, and OH number densities, and ignition delay time are measured in premixed hydrocarbon-air, hydrogen-air, and hydrogen-oxygen-argon flows excited by repetitive nanosecond pulse discharges in plane-to-plane and point-to-point geometries. Time-resolved temperature and OH number density in lean H\textsubscript{2}-air, CH\textsubscript{4}-air, C\textsubscript{2}H\textsubscript{4}-air, and C\textsubscript{3}H\textsubscript{8}-air mixtures are measured by picosecond, broadband Coherent Anti-Stokes Raman Spectroscopy (CARS) and by OH Laser-Induced Fluorescence (LIF). Time-resolved, spatially resolved temperature and absolute number densities of OH and H in Ar-O\textsubscript{2}-H\textsubscript{2} mixtures are measured by UV Rayleigh scattering, LIF, and Two-Photon Absorption LIF (TALIF), respectively. The results demonstrate that ignition occurs due to efficient generation of radical species in the discharge, and provide insight into the kinetic mechanism of low-temperature plasma assisted ignition. The results are compared with kinetic modeling calculations, showing the need for development of an accurate, predictive low-temperature plasma / fuel chemistry model applicable to fuels C\textsubscript{3} and higher.

The principal challenges in development of a predictive kinetic model include (i) lack of “conventional” chemical kinetics mechanisms validated at low temperatures, and (ii) lack of data on rates and products of reactions of excited species generated in the plasma, and their coupling with fuel-air plasma chemistry. “Conventional” combustion chemistry mechanisms have been developed for relatively high temperature conditions, and their applicability below ignition temperature, common in plasma assisted combustion environments, needs to be critically evaluated. This requires time-resolved measurements of radical species concentrations during low-temperature fuel oxidation, when an initial pool of primary radicals (O, H, and OH) is generated in the plasma, such as in the late afterglow of an electric discharge. This approach allows isolating relatively slow “conventional” low-temperature fuel oxidation reactions triggered by the radicals from the reactions of excited species generated in the discharge, which decay relatively rapidly. A complementary approach is to focus on kinetics of “rapid” reactions of excited species in the electric discharge and their effect on production of radicals in the early afterglow. These experiments would provide key data on coupling of molecular energy transfer processes in the plasma with “conventional” chemical reactions. Time-resolved measurements of temperature, excited species, and radical species concentrations are critical for characterization of the nonequilibrium reacting mixture at these conditions. Kinetic modeling of recent experiments in nanosecond pulse electric discharges in air suggest that the role of electronically excited N\textsubscript{2} \textsuperscript{*} molecules on chemical reactions in the afterglow, such as NO generation reactions, has been significantly underestimated in the past. Further experiments in fuel-air mixtures are expected to provide additional data on the role of these excited species on low-temperature fuel-air chemistry.