Experimental and Kinetic Modeling Study of C_2H_2 Oxidation at High Pressure

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Acetylene (C_2H_2) is an important intermediate in combustion of hydrocarbons as well as in atmospheric chemistry. Previous studies of C_2H_2 oxidation have been conducted in static reactors, jet-stirred reactors, flow reactors, shock tubes, and premixed laminar flames. Comprehensive modeling studies of acetylene oxidation have been published by Tan et al. [1] and by Lindstedt and Skevis [2]. Even though the reported studies cover a fairly wide range of stoichiometries and temperatures, results for the low-to-medium temperature oxidation chemistry of acetylene is limited to the early static reactor work, and except for the jet-stirred reactor experiments of Tan et al. [1] covering pressures up to 10 bar, data obtained at above atmospheric pressure are scarce.

In the present work, a detailed chemical kinetic model for oxidation of C_2H_2 in the intermediate temperature range and high pressure has been developed. Key reactions, including $C_2H_2 + O_2$ and $C_2H_2 + HO_2$, were characterized theoretically. Experiments with C_2H_2/O_2 mixtures diluted in N_2 were carried out in a high pressure flow reactor at 600–900 K and 60 bar, varying the reaction stoichiometry from very lean to fuel-rich conditions.

Figure 1 compares experimental and modeling results for the major species at stoichiometric conditions. Our work indicates that more work is required to explain the high-pressure chemistry of acetylene. Under the investigated conditions the oxidation pathways for C_2H_2 are more complex than those prevailing at higher temperatures and lower pressures. The major differences are the importance of the CHCHOH adduct, formed from addition of OH to C_2H_2. CHCHOH is oxidized through reaction with O_2, forming significant amounts of glyoxal (CHOCHO) and formic acid (HOCHO), which is then converted to HCO and CO.

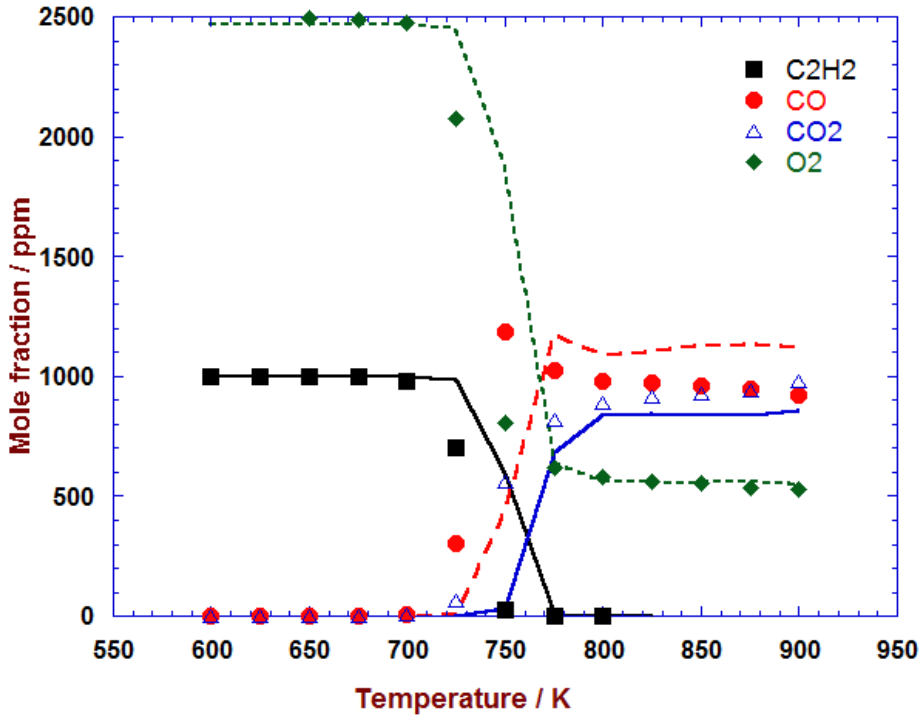

Figure 1: Comparison of experimental and predicted concentration profiles as a function of the reactor temperature for the stoichiometric experiment with C_2H_2/O_2 ($\lambda = 0.99$). The pressure was 59.6 bar and the reactor residence time was 8827/T (s·K). The inlet composition was 1000 ppm C_2H_2, 2470 ppm O_2, and N_2 by difference. The symbols mark experimental data while solid lines denote model predictions obtained at isothermal conditions.

References

[1] Tan, Y.W., Dagaut, P., Cathonnet, M., Boettner, J.C. Combust. Sci. Technol. 102 (1994) 21-55.

[2] Lindstedt, R.P, Skevis, G. Combust. Sci. Technol. 125 (1997) 73-137.