CRIEGEE INTERMEDIATE CH_2OO IN THE OXIDATION OF ETHANE

NATHAN A. SEIFERT, *Department of Chemistry, University of New Haven, West Haven, CT, USA*; RAGHU SIVARAMAKRISHNAN, KIRILL PROZUMENT, *Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, IL, USA*.

The family of Criegee intermediates, commonly designated as QOO, where Q is CH_2, CH_3CH, and so on, has been predicted by Rudolf Criegee in 1949 and discovered recently in laboratory studies. These highly unstable and reactive species are important in the atmosphere where they are formed by ozonolysis of alkenes. To investigate its chemistry, CH_2OO has been produced in various laboratory settings by either ozonolysis of CH_2CH_2, photodissociation of CH_2I_2 followed by oxidation, or oxidation of CH_4 under the discharge conditions. At Argonne, we observe CH_2OO resulting from the oxidation of CH_3CH_3 in a continuous-flow SiC microreactor heated to 1700 K. Cold ($T_{rot} = 7$ K) Criegee intermediate is detected in the supersonic molecular beam emerging from the hot microreactor and using the chirped-pulse Fourier transform millimeter-wave spectrometer, which operates in the 60–90 GHz region and is equipped with a fast narrowband digitizer for averaging 10^7 free induction decay traces in 5 minutes. The branching ratios of CH_2OO to the main oxidation products HO_2 and CH_2O are measured as a function of reactor temperature. We discuss the possible chemical pathways and the thermodynamic conditions within the reactor and outside of it that may lead to the formation and retention of the "fragile" CH_2OO intermediate in this experiment.