INVESTIGATING THE ONSET OF NUCLEATION: LOW TEMPERATURE REACTION KINETICS OF HET-ERODIMER FORMATION

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Nucleation is a key process in cloud and dust particle formation in different systems including planetary atmospheres and circumstellar shells. It consists of a succession of unions of small gas-phase molecules leading to the formation and growth of droplets and particles. Under the appropriate conditions, low temperatures and/or high degrees of supersaturation, nucleation can become a barrierless process, and dimerization, the complexation of the two smallest molecular species involved, becomes its rate-limiting step. The kinetics of formation of homodimers have been investigated in a few previous studies, using the CRESU (reaction kinetics in uniform supersonic flow) technique mainly coupled with mass spectrometric detection. Here we have employed a completely new detection scheme, chirped-pulse Fourier transform mm-wave spectroscopy, to study for the first time the kinetics of formation of heterodimers. This innovative technique combines the ability to generate continuous cold uniform supersonic flows with the high selectivity and general applicability of rotational spectroscopy, allowing us to follow both reactant and product concentrations simultaneously. Furthermore, the high sensitivity achieved has allowed us to employ pseudo-first-order conditions to obtain absolute rate constants. We will report the first measurements of rate constants for the formation of a set of heterodimers, including formic acid, CO2 and water among others, at temperatures between 35 and 150 K.