**Low-temperature Gas-Phase Formation of Methanimine (CH2NH; X1A') – the Simplest Imine – under Single-Collision Conditions**

Zhenghai Yang, Iakov A. Medvedkov, Shane J. Goettl, Ralf I. Kaiser\*

*Department of Chemistry, University of Hawai’i at Manoa, Honolulu, Hawaii 96822, USA*

\*Email: [ralfk@hawaii.edu](mailto:ralfk@hawaii.edu);

**EXPERIMENTAL**

**EXPERIMENTAL:** The reaction of the D1-methylidyne radical (CD; X2Π) with ammonia (NH3; X1A1) was investigated utilizing a crossed molecular beams machine.[1](#_ENREF_1),[2](#_ENREF_2) The D1-methylidyne beam was produced through photodissociation (COMPex 110; 248 nm) of D1-bromoform (CDBr3; Sigma Aldrich; ≥99.5%) held in a stainless-steel bubbler at a stagnation temperature of 283 K. Helium acted as carrier gas with backing pressure of 2.2 atm and the mixture was released via a pulsed piezoelectric valve at 60 Hz repetition rate.[3](#_ENREF_3) The 248 nm laser output operated at 30 Hz and intersected molecular beam 1 mm downstream of the nozzle with output energy of 160 mJ pulse-1. The selection of 60 Hz of the pulse valve and 30 Hz of the excimer laser allowed a background subtraction during the experiment data collection. The primary CD molecular beam was skimmed and then velocity-selected via a four-slot chopper wheel rotating at 120 Hz mounted between the skimmer and the interaction region and controlled by a high stable motion system (Faulhaber; 2057024B). The selected well-defined section of CD beam possesses a peak velocity (vp) of 1762 ± 22 m s-1 along with a speed ratio (S) of 9.2 ± 1.1 and intersected perpendicularly with the crossing segment of the secondary pure ammonia (NH3; Matheson; 99.99%) released at repetition rate of 30 Hz and backing pressure of 550 Torr with the peak velocity *v*p of 1143 ± 34 m s-1. The collision in the interaction region results in a CM angle of 38.2 ± 0.4° and a collision energy of 16.9 ± 0.3 kJ mol-1. The molecular beam parameters are compiled in Table S1.

Products of reactive scattering were ionized by an electron impact ionizer operated at 80 eV and 2 mA. A quadrupole mass spectrometer (QMS) was exploited at time-of-flight (TOF) mode to filter the produced ions according to distinct mass-to-charge ratios (m/z). The designated ions first hit to a stainless-steel target coated with aluminum and initiated a cascade-of-electron pulse which then fly to an organic scintillator generating a photon pulse. The signal was finally detected by a Burle photomultiplier tube and collected via a multichannel scaler after filtered by a discriminator operating at 1.6 mV.[4](#_ENREF_4" \o "Doddipatla, 2021 #1368) The detector assembly is rotatable and operated under ultrahigh-vacuum conditions of 7 × 10-12 Torr. To obtain reaction dynamics information, a forward-convolution routine is employed, and the data is transferred from the laboratory frame to the center-of-mass (CM) frame resulting in best-fits and the corresponding CM functions of *P*(*E*T), *T*(*θ*) and the contour flux map, *I*(*u*, *θ*) ~ *P*(*u*) × *T*(*θ*) with the CM scattering angle *θ* and product velocity *u*.[2](#_ENREF_2),[5](#_ENREF_5) The flux contour map depicts the flux of the reactive-scattering products and contains dynamics information of the scattering reaction.

**Table S1.** Peak velocities (vp) and speed ratios (S) of the D1-methylidyne radical (CD), and ammonia (NH3) beams along with the corresponding collision energy (EC) and center-of-mass angle ().

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| --- | --- | --- | --- | --- |
| Beam | vp (m s-1) | S | Ec (kJ mol-1) | (deg) |
| CD | 1762 ± 22 | 9.2 ± 1.1 |  |  |
| NH3 | 1143 ± 34 | 10.3 ± 1.4 | 16.9 ± 0.3 | 38.2 ± 0.4 |

**References**

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