

Abstract Submitted
for the GEC18 Meeting of
The American Physical Society

Strong Vibrational Non-Equilibrium in N₂ and CO₂ by Microsecond Pulsed Microwave Plasma GERARD VAN ROOIJ, DIRK VAN DEN BEKEROM, TOM BUTTERWORTH, TEOFIL MINEA, QIN ONG, ALEX VAN DE STEEG, RICHARD VAN DE SANDEN, Dutch Institute for Fundamental Energy Research — Novel means for electrification of chemical processes are essential in order to keep up with the modernizing energy supply. In this context, we investigate microwave plasma to utilize electrical power for activation and conversion of stable molecules, in particular CO₂ and N₂. Preferential vibrational excitation is considered as pathway to their fixation reactions with ultimate efficiency. In this study, the microwave power was pulsed at low duty cycle to limit gas heating and to reveal vibrational excitation dynamics. The typical pulsing scheme was 100 microsec ON-time followed by 25 ms OFF-time. Raman scattering yielded the vibrational and rotational temperature evolution during the pulse. Strong non-equilibrium was observed both in N₂ and CO₂. Specifically, in N₂ the rotational temperature, taken as proxy for the gas temperature, slowly increased to 1200K, after the vibrational temperature already peaked at 3500K. In CO₂, also non-equilibrium between different vibrational degrees of freedom was observed. In fact, the asymmetric stretch immediately heated to over 1000K before equilibrating with the symmetric stretch and bending mode at 1800K. Finally, the vibrational excitation dynamics were correlated to CO yield and efficiency in scans of ON-time.

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Date submitted: 17 Jun 2018

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