Understanding NO formation and destruction by non-thermal effect in the quenching process of microwave air plasma

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Plasma-based nitrogen fixation driven by renewable electricity holds promise to counteract the substantial CO_2 emission and energy consumption associated with the Haber-Bosch (HB) process [1]. Microwave plasma is gaining increasing interest worldwide due to its high operation flexibility, but a better performance in terms of energy efficiency is still required. An important step in this plasma-based gas conversion is the quenching of the dissociated air flow, which, next to the desired product NO, also contains atomic oxygen and nitrogen. Since most of the recombination energy released in the quenching process by these atomic fragments ends up in vibrational modes, the state of the gas could be non-thermal with the vibrational temperatures different from the gas temperature.

To explain the underlying mechanisms, we introduce a time-dependent multi-temperature quenching model coupling chemical and vibrational kinetics at different pressures, which gains insights into the pathways of NO formation and destruction. Relaxations of the vibrational and gas temperatures during the forced cooling trajectory are shown in this work. A new method, different from the theoretical-informational method [2], to calculate chemical reaction rate enhancement by vibrational excitation is proposed. A series of energy transfer channels including vibrational-vibrational (V-V) and vibrational-translational (V-T) energy transitions [3], together with chemical processes and heat conduction are tracked in the quenching process (Fig 1).

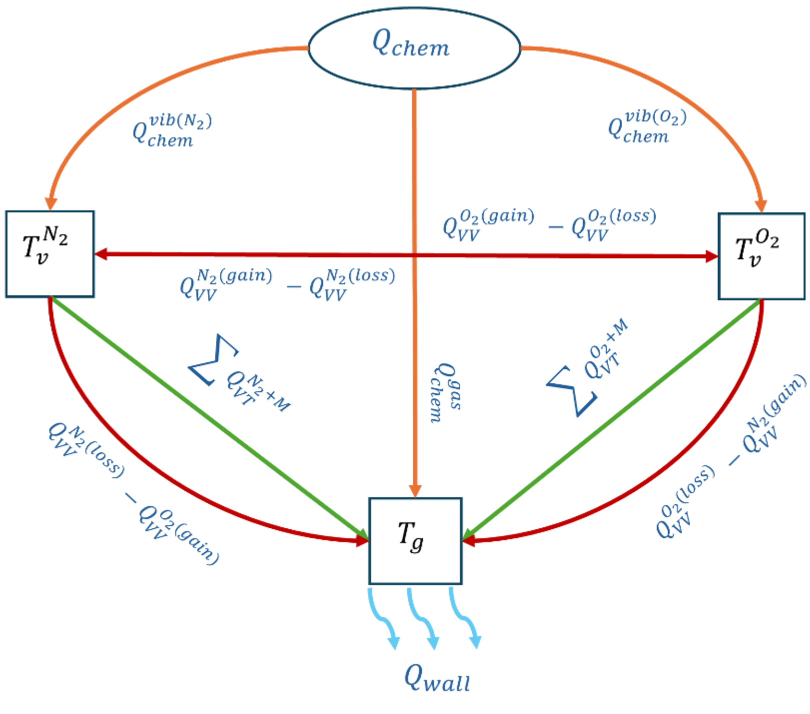

Fig. 1: Overview of the energy transition scheme in the quenching process.

Through manipulation of the cooling rate and the gas temperature in the plasma region, our model elucidates the effect of vibrational kinetics on NO production for different conditions. The predominant factor limiting the vibrational non-equilibrium is the V-T relaxation of N_2 and O_2 molecules due to collisions with oxygen atoms. The model also predicts that accelerating the oxygen atom recombination process in the quenching region is beneficial to preserve NO concentration from the plasma region, especially at high temperatures. This research establishes a foundation for the further advancement and optimisation of plasma reactors for the efficient production of nitrogen oxides.

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