On activation of the non-equilibrium vibrational CO_2 dissociation in plasma chemistry

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The plasma-chemical splitting of CO_2 has been intensively studied in the last decade in view of its potential applications for Power-2-X technologies. Dissociation from high vibrational states in conditions of strong vibrational non-equilibrium at relatively low translational-rotational temperatures T was considered in the past as the most efficient mechanism of the CO_2 plasma conversion [1]. This mechanism could not be repeated so far in modern day experiments which rather focus on thermal quenching [2]. The contribution will present a theoretical investigation of the conditions at which the non-equilibrium vibrational mechanism could be triggered. Two approaches are applied. First is a semi-empiric estimate based on the vibrational relaxation rates obtained in shock tube and sound absorption experiments. Second is the state-to-state model of the CO_2 vibrational kinetics and the primary dissociation process $CO_2 + M \rightarrow CO + O + M$ in 2-modes approximation [3]. The governing parameter Q/n_0^2 has been introduced, where Q is the specific volumetric power coupled in plasma and n_0 is the initial number density of CO_2. The state-to-state simulations suggest that the non-equilibrium process can be activated when Q/n_0^2 exceeds some critical value. $\left(Q/n_0^2\right)_{crit}$ around $6{\cdot}10^{-40}$ W·m^3 at T=300 K found by the first approximate approach agrees well with the numerical results. The semi-empiric method is further extended to estimate the potential reduction of $\left(Q/n_0^2\right)_{crit}$ in CO_2/CO mixtures.

References

[1] Legasov V A *et al.* 1978 *Dokl. Akad. Nauk.* **238** 66

[2] den Harder N *et al.* 2017 *Plasma Process Polym.* **14**

[3] Kotov V 2021 *Plasma Sources Sci. Technol* **30** 055003