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Towards a fundamental understanding of energy-efficient, plasma-based CO_2 conversion

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Plasma-based CO_2 conversion is worldwide gaining increasing interest. The aim of this work is to find potential pathways to improve the energy efficiency of plasma-based CO_2 conversion beyond what is feasible in thermodynamic equilibrium. To do so, we use a combination of modeling and experiments to better understand the underlying mechanisms of CO_2 conversion, ranging from non-thermal to thermal equilibrium conditions. Zero-dimensional (0D) chemical kinetics modelling, describing the detailed plasma chemistry, is developed to explore the vibrational kinetics of CO_2, as the latter is known to play a crucial role in the energy efficient CO_2 conversion [1,2]. The 0D model is successfully validated against pulsed CO_2 glow discharge experiments [3], enabling the reconstruction of the complex dynamics underlying gas heating in a pure CO_2 discharge, paving the way towards the study of gas heating in more complex gas mixtures, such as CO_2 plasmas with high dissociation degrees. Since gas heating has a strong effect on vibrational excitation, this work represents a substantial step forward in the definition of the experimental conditions suitable for vibrationally-enhanced CO_2 splitting in a plasma.

Energy-efficient, plasma-based CO_2 conversion can also be obtained upon the addition of a reactive carbon bed in the post-discharge region [4,5]. The reaction between solid carbon and O_2 to form CO allows to both reduce the separation costs and increase the selectivity towards CO, thus, increasing the energy efficiency of the overall conversion process. In this regard, a novel 0D model to infer the mechanism underlying the performance of the carbon bed over time is developed. The model outcome indicate that gas temperature and oxygen complexes formed at the surface of solid carbon play a fundamental and interdependent role. These findings open the way towards further optimization of the coupling between plasma and carbon bed, which proved very promising.

Experimentally, it has been demonstrated that “warm” plasmas (e.g. microwave or gliding arc plasmas) can yield very high energy efficiency for CO_2 conversion, but typically only at reduced pressure [6]. For industrial application, it will be important to realize such good energy efficiency at atmospheric pressure as well. However, recent experiments illustrate that the microwave plasma at atmospheric pressure is too close to thermal conditions to achieve a high energy efficiency [7–9]. Hence, we use a comprehensive set of advanced diagnostics to characterize the plasma and the reactor performance, focusing on CO_2 and CO_2/CH_4 microwave discharges. In particular, laser scattering is coupled with optical emission imaging to reconstruct the shape of the plasma and link it with the evolution of electron density and temperature, and gas temperature. The results lead to a deeper understanding of the mechanism of power concentration with increasing pressure, typical of plasmas in most gases, which is of great importance for model validation and understanding of reactor performance.

[1] A. Fridman, Plasma Chemistry, Cambridge: Cambridge University Press, 2008.
[2] T. Kozák, A. Bogaerts, Plasma Sources Sci. Technol. 23 (2014) 045004.
[3] B.L.M. Klarenaar, R. Engeln, D.C.M. van den Bekerom, M.C.M. van de Sanden, A.S. Morillo-Candas, O. Guaitella, Plasma Sources Sci. Technol. 26 (2017) 115008.
[4] P. Liu, X. Liu, J. Shen, Y. Yin, T. Yang, Q. Huang, D. Auerbach, A.W. Kleiyn, Plasma Sci. Technol. 21 (2019) 012001.
[5] H. Zhang, Q. Tan, Q. Huang, K. Wang, X. Tu, X. Zhao, C. Wu, J. Yan, X. Li, ACS Sustainable Chem. Eng. 10 (2022) 7712–7725.
[6] R. Snoeckx, A. Bogaerts, Chem. Soc. Rev. 46 (2017) 5805–5863.
[7] G.J. van Rooij, D.C.M. van den Bekerom, N. den Harder, T. Minea, G. Berden, W.A. Bongers, R. Engeln, M.F. Graswinckel, E. Zoethout, M.C.M. van de Sanden, Faraday Discuss. 183 (2015) 233–248.
[8] S. Heijkers, A. Bogaerts, J. Phys. Chem. C 121 (2017) 22644–22655.
[9] D.C.M. van den Bekerom, J.M.P. Linares, T. Verreycken, E.M. van Veldhuizen, S. Nijdam, G. Berden, W.A. Bongers, M.C.M. van de Sanden, G.J. van Rooij, Plasma Sources Sci. Technol. 28 (2019) 055015.