Isotope labelling: A new technique to analyse reaction mechanisms in plasma-gas processes

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This work is concerning the plasma reforming of methane using labelled D_2O molecules as reactant with the aim of identifying some of the key intermediate species intervening in the reaction mechanisms. The study herein reveals important clues about those intermediate plasma processes running in parallel to the main reaction leading to the formation of CO and hydrogen. In concrete, we have found that a considerable exchange of H(D) by D(H) atoms occurs in the exhaust gases (i.e, hydrogen, methane and water) under different operating conditions, proving that much of the plasma energy is used to produce intermediate species which are inefficient for the formation of final products.

Isotope labelling, a classical method in catalysis to ascertain reactions routes [1, 2], has been scarcely applied in plasma processes [3]. In this work we have used deuterated water as labelling compound to analyse the molecular fragmentation during the plasma wet reforming of methane in a parallel plate packed-bed DBD reactor filled with ferroelectric material [4]. Reaction products were monitored by means of a mass spectrometer. Apart from the expected hydrogen (H_2) and carbon monoxide (CO), deuteromethanes and molecular hydrogen isotopes (D_2, HD, CH_3D, CH_2D_2) appear in the reaction products. The existence of isotope labelled molecules (CH_3D, CH_2D_2) is a clear proof of the occurrence of backwards reactions during the wet reforming of methane. These processes imply a waste of energy and, thus, a decrease in the efficiency of the DBD plasma processes, one of the major drawbacks for its industrial implementation. The influence of different parameters, namely, the gas residence time, the current and the addition of oxygen on the H/D distribution and efficiency of the process is analysed.

References

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Acknowledgements

We acknowledge financial support from Junta de Andalucía through the project P12–2265MO and from the European Regional Development Funds program (EU-FEDER) and the MINECO (project MAT2013-40852.