THESE PPRIME

Joint numerical/experimental analyses of transfers and chemical reactions at interfaces in a catalytic reactor for the transformation of biogas by non-equilibrium plasma

CONTEXT

The European Green Deal, launched in December 2019, aims to make Europe the first climate neutral continent. The set target is to stop net greenhouse gas emissions by 2050. The challenge lies in the efficient use of resources and in the development of new environmentally friendly technologies. In this context, the production of biogas by anaerobic fermentation of agricultural waste, which is widespread in Europe, appears as a key industrial sector. Currently, this biogas consisting mainly of methane but also carbon dioxide in relatively high concentration (from 30 to more than 40% vol) is used to locally supply heat and/or electricity delivered to the network. However, when energy needs are lower it becomes important to offer other outlets for biogas, for example by storing energy. One solution is the production of chemical compounds like methanol to supply bio-sourced molecules to the local chemical industry. It is also possible to produce a liquid fuel from biogas and using "clean" electricity generated on site or the network surplus. These solutions validate the concept of chemical energy storage. The synthesis of methanol from biogas is a promising alternative to the conventional process using fossil carbon to produce syngas and then methanol over Cu/ZnO/Al2O3 catalysts at 493-573K and 5-10 MPa. However, the cost of this conventional process does not allow its use for the onsite methanol production. Accordingly, new breakthrough processes are required. Among them, non-thermal plasma (NTP) technologies have the advantage of being small in size and operating at atmospheric pressure and low temperature. Then, these processes feature the necessary characteristic for biogas valorization and can therefore be perfectly combined with anaerobic digestion. Nevertheless, the transition from a lab concept to an industrial innovation needs first significant scientific advances[1].

THESIS SUBJECT

Accordingly, the work thesis proposed here is a part of a larger project which aims at acquiring new insights into the interaction plasma/catalysis in ceramic geopolymer foams possessing macro-porosity thought to favor the transformation of biogas into methanol. To reach such a target, simultaneous research efforts are planned (i) to synthetize new shaped catalytic materials, (ii) to set up experiments and diagnostics to evaluate catalytic performances, and (iii) to thoroughly analyze the basic physical mechanisms involved at the plasma/catalyst interface. This thesis will focus on these two latter points and will require the development of new numerical tools specifically design to simulate the plasma reactor available at IC2MP[2]. Despite the growing interest of scientist in plasma-catalysis, the interaction between active species generated in gas phase and the surface of the catalyst is not fully understood. Significant advances to figure out how these molecules interact will only be obtained combining experimental and simulation results. Progress has been made in the microkinetic modeling of plasma/catalysis mechanisms[1] when applied to 0D/1D reactors. However, the association of such models with 2D/3D fluid dynamics modeling strategies remains a challenge. Although, providing a 3D computational tool will be necessary to assist engineers to design future industrial facilities and to optimize plasma-catalyst reactors. For research work, such a tool will provide both extrapolated experimental data and numerical results of canonical flows. It will help to understand the smallest scale mechanisms, i.e molecular transport, occurring at the catalyst surface and to develop relevant numerical modeling strategies with a reasonable computational cost.

Eventually, the complexity of interactions between plasma, chemical reactions and surface transfers of species and electrons requires the coupling of different numerical tools and the development of an hybrid modeling strategy. The first step of this work will be the coupling of a Boltzmann solver (Bolos library[3]) with a chemical kinetic solver (Cantera library[4]) to describe the surface chemistry and transfers in the non-equilibrium plasma. From this simple tool (0D, 1D), thermochemical data tables will be created and then reduced by training an artificial neural network. These data will feed the models used in the Navier-stokes solver (Open-Foam library) needed for the 2D and 3D numerical simulations of the reactor. In a second step, experiments will be realized using the existing reactor to validate the developed numerical tools. Numerical results will also provide detailed information on the fluxes at walls completing experimental data.

(1) Bogaerts, A.; Centi, G. Plasma Technology for CO2 Conversion: A Personal Perspective on Prospects and Gaps. Front. Energy Res. 2020, 8, 111.
(2) Bouchoul, N.; Fourré, E.; Tatibouët, J.-M.; Duarte, A.; Tanchoux, N.; Batiot-Dupeyrat, C. Structural Modifications of Calcium Based Catalysts by Non-Thermal Plasma in the CO2 Reforming of CH4 and the Influence of Water. J. CO2 Util. 2020, 35, 79–89. https://doi.org/10.1016/j.jcou.2019.09.006
(3) Hagelaar G. J. M.; Pitchford L. C., Solving the Boltzmann equation to obtain electron transport coefficients and rate coefficients for fluid models, Plasma Sources Sci. Technol., 2005, 14, 722–733.
(4) Goodwin D. G.; Speth R. L.; Moffat H. K.; Weber B. W., Cantera: An Object-oriented Software Toolkit for Chemical Kinetics, Thermodynamics, and Transport Processes. 2009, https://doi:10.5281/zenodo.1174508

CONTACTS

Vincent ROBIN vincent.Robin@isae-ensma.fr

Catherine BATIOT-DUPEYRAT catherine.batiot.dupeyrat@univ-poitiers.fr

ABSTRACT

The development of a new process for biogas valorization by non-equilibrium plasma and a catalyst made of a ceramic geopolymer foam requires to analyze, understand and model the interaction between the gas phase, the chemical reactions and the wall fluxes of species and electrons. The association of experimental measurements in an available catalytic-plasma reactor and 2D/3D numerical simulations of the internal flow will lead to a large database facilitating analyses. A new numerical tool must be developed and validated using new experimental data.