43GERSEQ
Ciudad Real 2023

XLIII Reunión del Grupo Especializado de Electroquímica de la RSEQ (43 GERSEQ 2022)

Ciudad Real, 3-5 Julio 2023

Hierarchical porous catalysts based on carbon materials for the production of green fuels: valorization of CO_2 and NH_3 production

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Abstract
In this doctoral thesis I will explore the use of catalysts based on nanoporous carbonaceous materials with controlled porosity for the electrochemical reduction of CO_2 under solar irradiation (and its valorization into fuels and chemical consumables of added value) and the synthesis of NH_3 from various N-sources (e.g., N_2, NO_3^-) using (photo)electrochemical processes. This approach makes it possible to respond to two major concerns: i) the management of CO_2 as a greenhouse gas and the development of technologies based on solar radiation, and ii) opening the use of NH_3 as a carbon-free energy carrier for "long-term" hydrogen storage.This thesis prepares to synthesize hybrid catalysts with hierarchical porosity based on carbonaceous materials and their coupling with semiconductors, and to show that these hybrid catalysts are effective in CO_2 conversion and NH_3 production reactions. In this respect, the use of carbonaceous materials as (photo)electrocatalysts is very interesting because recent studies have proven high performance in energy conversion processes, in particular in the photoreduction of CO_2 and the production of H_2/O_2 under solar irradiation, and the activation of N_2 by nanoconfinement effect in the pores [1-3].
On the one hand, I will study the photoelectroreduction of CO_2 thanks to the control of the nanopore/CO_2 interfaces, the electronic conductivity of carbon materials and photon penetration. We will examine the selectivity for the production of solar fuels and chemical consumables such as methanol, ethanol, or formic acid and the correlation between the performance and the characteristics of the carbon catalysts. On the other hand, we will study the photo(electro)synthesis of NH_3 under moderate reaction conditions.To do this, I will explore the importance of pore size and surface chemistry of the carbon catalysts for improving the interfacial interactions with nitrogen and other N-sources (typically very weak). This work will explore the immobilization of additives (e.g., ionic liquids, DES) in the porosity of the materials to enhance nitrogen solubility in the nanoconfined pores, the functionalization of Lewis sites on the activation of N-compounds and the suppression of competing reactions (e.g., hydrogen evolution reaction) with ammonia production. For the most efficient materials, a (photo)electrochemical reactor will be set up, with the optimization of the operating conditions (e.g., morphology of the electrodes, electrochemical reactor, and gaseous or liquid route).

Agradecimientos

A. Fayad thanks Université d'Orléans for a PhD fellowship.

Referencias

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