Université Lille Nord de France
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Ecole doctorale régionale Sciences Pour l'Ingénieur Lille Nord-de-France - 072

Titre Thèse	Plasmon-enhanced CO_2 Electrochemical Reduction Reaction to Added Value Chemicals	
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Résumé du sujet :

CO_2 is a potential greenhouse gas and its anthropogenic and natural emissions are expected to create global catastrophe. On the one hand, integrated carbon capture and subsequent sequestration is generally advanced as the most promising option to tackle greenhouse gases in the short to medium term. On the other hand, transformation of CO_2 to added value chemicals or fuels is a promising area of current research interest. Photocatalytic reduction of CO_2 with water for the production of hydrocarbons could be one of the viable solutions to address the issues related to the climate change as well as energy shortage. In natural photosynthesis, CO_2 is converted into glucose *via* a series of intermediate products with the assistance of solar light. Unfortunately, such a process has low conversion efficiency. Processes mimicking natural photosynthesis, so-called artificial photosynthesis, have been developed to increase this conversion efficiency and provide more pathways to produce valuable chemicals using CO_2 as the carbon feedstock.

Semiconductor-based photo- or photoelectrochemical methods are currently being used widely for CO_2 reduction in that the synergistic conjunction of photoelectrocatalysis (namely, electrocatalysis and photocatalysis) facilitates the separation of photo-induced electrons and holes under applied electric fields. Light irradiation-induced band bending compensates the required over potential for CO_2 reduction, causing the CO_2 conversion to occur at relatively positive potentials. However, most of the semiconductors have poor conductivities, low charge mobility, high charge-carrier recombination, and reduced diffusion length. Thus, photoelectrochemical CO_2 reduction still occurs at relatively negative potentials in these systems. Finally, the lack of selectivity encountered during CO_2 reduction under different experimental conditions represents an unmet demand that requires constant and continuous attention from the scientific community.

Plasmonics has recently emerged as an effective approach for overcoming most of the limitations encountered in the photocatalytic processes even though the understanding of the physical mechanisms remains elusive. Although plasmonics have been successfully introduced for various electrochemical processes such as water splitting, the field of plasmon-enhanced CO_2 electroreduction is still in its infancy. Additionally, all of the reported plasmonic interfaces for CO_2 reduction are based on coupling the classical semiconductors with plasmonic nanostructures (Au, Ag). The investigation of the plasmonic properties of Cu-based nanostructures for electrocatalytic processes (CO_2 reduction) has not been yet described in the scientific literature.

The aim of the present proposal is to develop new plasmonic interfaces based on copper nanohole arrays for plasmon-enhanced electrochemical reduction of CO_2 to added value chemicals and gain a better understanding on the reduction mechanism. This project will be performed in close collaboration with IEMN-EPHONI group to simulate the most appropriate architecture and LASIRE-PCS on time-resolved spectroscopy to tackle the plasmon-enhanced reduction mechanism.