Electrochemical CO_2 Splitting into CO and O_2 in Neutral Water Using Earth-Abundant Materials

From Molecular Catalysts to a Whole Electrolyzer

Thesis defense presented by Arnaud TATIN,
on Tuesday November, 29th at 2:00 PM in room #774 (Bât. Lavoisier)

Abstract

Electrical energy can be stored in chemical bonds thanks to an appropriate electrolyzer. Typical reactions include hydrogen generation and, more recently, the production of hydrocarbons and oxygenates from CO_2. Besides electricity storage in the chemical bonds of high-energy-content molecules, such advances allow the production of various chemical feedstocks to manufacture higher value compounds. Thus, electrofuels synthesis supplements the integration of renewable energy sources in the electrical production mix; these fuels are compatible with the current industrial infrastructure and supply chain while they can be stored easily.

The present thesis focuses at first on the development of new molecular catalysts for the selective CO_2-to-CO conversion in water using only earth-abundant materials, namely iron-based porphyrin derivatives. Miscellaneous attempts to derive new catalysts with diverse substituents are reviewed. Once an active, water-soluble catalyst is identified, a performance, selectivity, and stability assessments are completed using electrochemical techniques such as cyclic voltammetry investigations and bulk electrolysis. Then, the immobilization of said catalyst onto the electrode surface is discussed until a robust integration in the catalytic film is secured.

Eventually, the thesis reviews diverse considerations in the design of the whole electrolysis cell that lead to the selection of a proton-exchange membrane device where the CO_2-reduction catalyst (cathode) is coupled to a heterogeneous water-oxidation catalyst (anode). In the assembly of the reported electrolyzer, a cobalt-based thin film is picked for the anode. Finally, economic perspectives provide a clear, rational basis for future optimization of the device in a genuine industrial environment.

Composition of the PhD Committee:

Prof. Jean-Michel SAVÉANT	DR, CNRS Paris	Chairman
Dr. Elsje Alessandra QUADRELLI	DR, CNRS Lyon	Rapporteur
Prof. Marc T.M. KOPER	PR, Universiteit Leiden	Rapporteur
Dr. Régis RÉAU	Scientific Director, Air Liquide	Examiner
Dr. Fabien AUPRÊTRE	CTO, AREVA H_2Gen	Examiner
Dr. Claire FONGY	Project Manager, SATT Idf Innov	Examiner
Prof. Marc ROBERT	PR, Université Paris 7	Supervisor