

INSTITUT DE CHIMIE ANNÉE ACADÉMIQUE 2018-2019



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Managing Reactivity of Hydrides in CO₂ Reduction to Formate

Mardi 30 avril 2019 à 11h Salle D2

Biography:

Louise Berben was born in Sydney, Australia. She received a Bachelor of Science degree with 1st class honors from The University of New South Wales in 2000, and in 2005 was awarded a Ph.D. from the University of California Berkeley for research undertaken with Professor Jeffrey Long. In 2006 Louise began postdoctoral research with Professor Jonas Peters at the California Institute of Technology and in July 2007, moved with the Peters research group to the Massachusetts Institute of Technology. In July 2009, Louise joined the faculty at the University of California Davis where her research program focuses primarily on synthetic and physical inorganic chemistry.

Abstract:

In this talk I will discuss the reduction of CO_2 into C-H bond containing fuels using $[Fe_4N(CO)_{12}]$ - and related small metal carbonyl clusters as electrocatalysts. At pH 6.5 or in MeCN/H₂O (95:5), $[Fe_4N(CO)_{12}]$ - promotes the selective formation of formate from CO_2 saturated solution at -1.2 V vs. SCE which is about 440 mV of overpotential. The intermediate hydride in this reaction is believed to be the iron-iron bridged hydride: $[H-Fe_4N(CO)_{12}]$ -which forms via an electron transfer, proton transfer (EC) process. We have considered both thermochemical and kinetic factors that control selectivity in the reaction of $[H-Fe_4N(CO)_{12}]$ - with substrates so that we can better understand why the Faradaic efficiency for formate is 97%, with very little H2 produced. Results of mechanistic studies will be discussed, along with efforts in synthetic inorganic chemistry, to understand the role of the secondary coordination sphere in substrate transport.