



Monday, November 27, 2023

10h30-12h

Room 174 Sciences II

Studying and using Hydrogenases

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Hydrogenases are metalloenzymes that use a dinuclear inorganic active site (based on either iron or nickel and iron) to catalyze H_2 oxidation and formation. They are studied in a variety of contexts, ranging from bioenergetics and fundamental inorganic chemistry to the design of (photo)electrochemical fuel cells. Within each family of hydrogenases, the immediate environment of the active sites is very much conserved, but the enzymes differ by their protein sequences, cofactor composition and quaternary structures. There is recent evidence that their reactivity is determined by residues and structural features that are very remote from the active site. Nature therefore provides a quasi-infinite playground for investigating the very important and general concept of "outer sphere" control of the catalytic activity. Studying the effect of inhibitors (such as CO and O_2) is a common approach for learning about the reactivity of enzymes' active sites; in the case of hydrogenases, understanding these reactions is particularly important because they may negatively impact the use of these enzymes. We will show that combining site-directed mutagenesis and electrochemical kinetics is a powerful approach for understanding at a molecular level the reactivity of hydrogenases and to improve their resistance to inhibitors. It is also possible to embed and protect these enzymes in redox polymer films; this strategy affords new perspectives for the design of electrocatalysts of hydrogen oxidation and production.