Expanding the Biocatalytic Toolbox for Sustainable Chemistry: Semi-synthetic and Artificial Metalloenzymes for Energy Conversion

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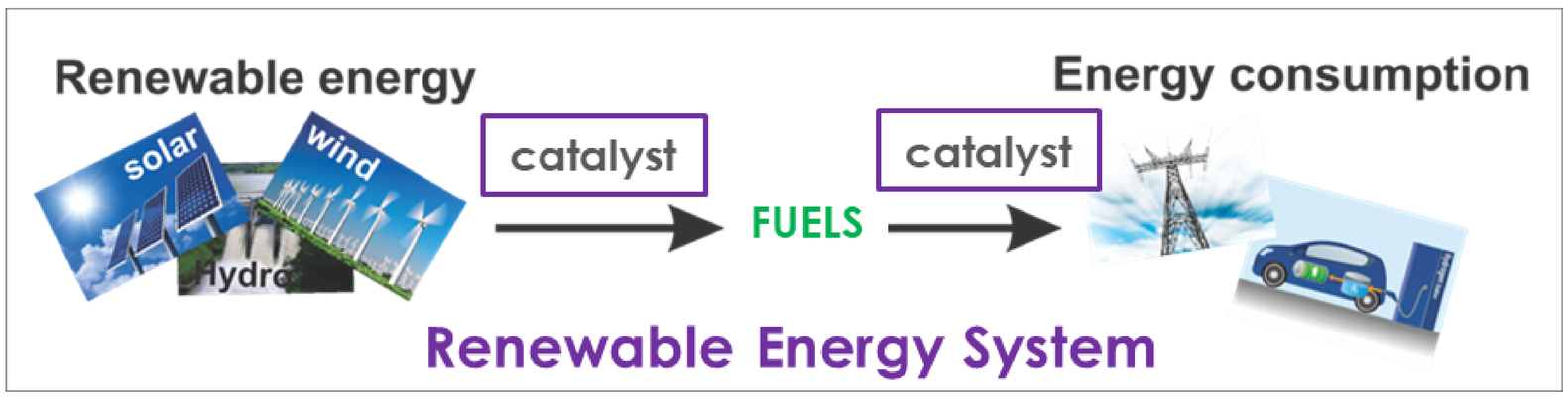
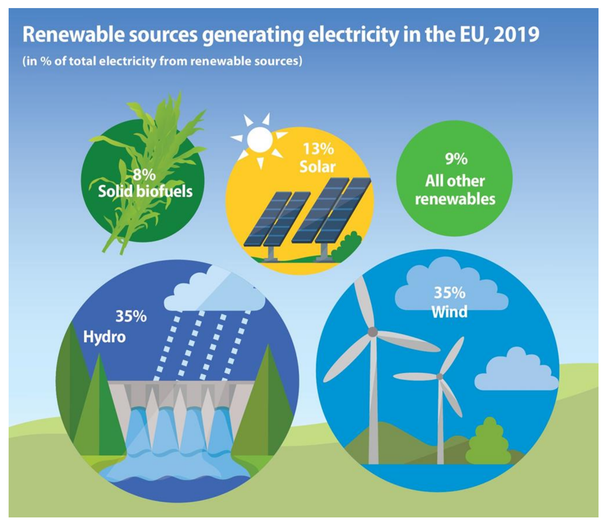
1. Why is this Research Important? Motivation: Energy Sustainability

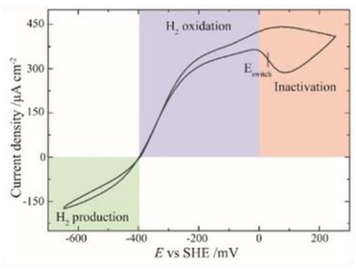




- ❑ Society consumes a lot of energy mostly from non-renewable sources
- ❑ **Energy production** generating 87% of global **greenhouse gas emissions**
- ❑ Non-renewable resources (i.e. fossil fuels) are limited so we need to shift towards a **renewable energy-system**
- ❑ Example of renewable energy-system: get rid of the **carbon dioxide** from the atmosphere and use **hydrogen** as a fuel
- ❑ Big problem: **energy storage** and **transportation**
- ❑ For **energy storage** and **transportation** we need **efficient catalysts** based on **earth-abundant metals (cheap, easy to obtain and produce)**
- ❑ **Catalysts** speed up chemicals reactions
- ❑ **Enzymes are nature's catalysts**

2. Learning from Nature's Enzymes

Electrochemistry



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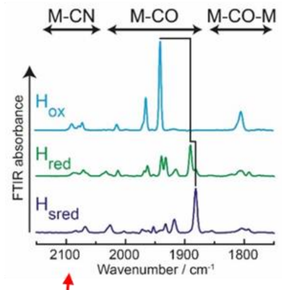
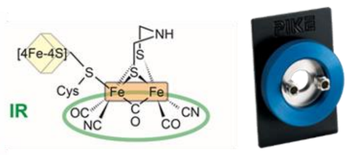
Infrared (IR) Spectroscopy





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Electron Paramagnetic Resonance (EPR) Spectroscopy

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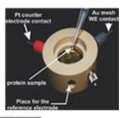
IR Spectro-electrochemistry



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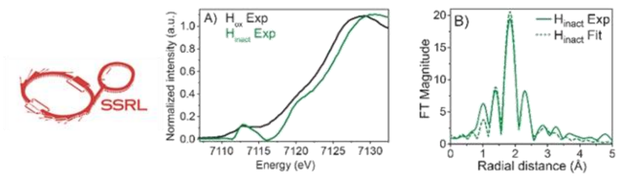
X-ray Spectroscopy and NRVS



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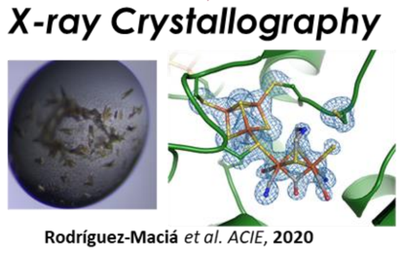
X-ray Crystallography



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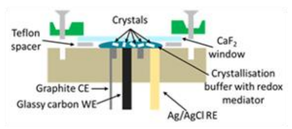
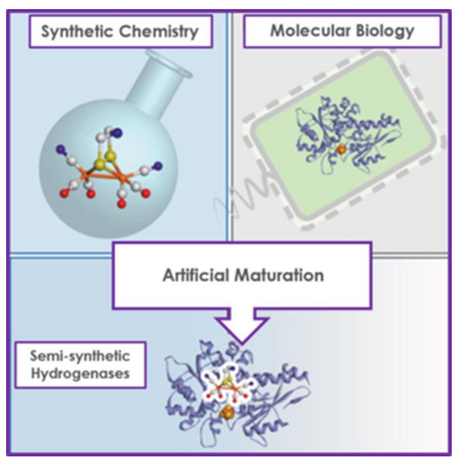
Single Crystal Electrochemical IR Microspectroscopy



- ❑ We use a wide combination of techniques to investigate enzymes
- ❑ Because they are very fast, we often cannot investigate them by conventional techniques and have to develop new methods
- ❑ With the knowledge gained we develop design principles to build catalysts and artificial enzymes for energy-conversion reactions
- ❑ We aim to understand what is the minimal subunit we need to 'reproduce' for catalytic activity

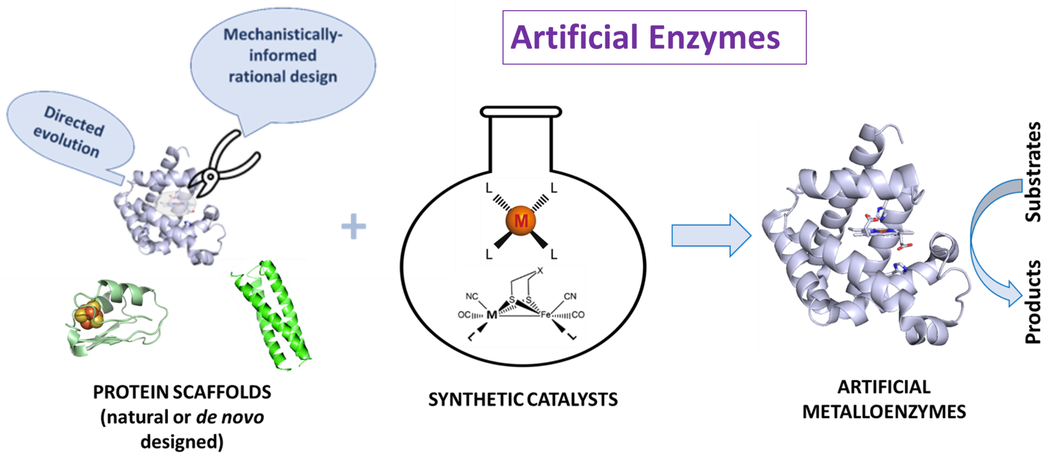
3. Approaches to Generate Semi-synthetic and Artificial Metalloenzymes

Semi-synthetic Enzymes



- ❑ 'Artificial maturation' allows incorporation of synthetic cofactors into natural protein-scaffolds
- ❑ Can determine important elements for catalysis and reactivity
- ❑ Allow preparation of semi-synthetic enzymes with novel properties

Artificial Enzymes



- ❑ Artificial enzymes result from incorporating synthetic catalysts into natural or *de novo* protein-scaffold
- ❑ They combine transition-metal catalysis with the unique benefits of enzymes
- ❑ They have applications in many fields:
  - Green energy
  - Pharmacology
  - Biosensing

4. Conclusions

- ➢ Can acquire **inspiration from nature** to help **solve the climate crisis**
- ➢ E**nzymes found in nature catalyse** reactions that are key to **energy sustainability**
- ➢ Need to **understand natural enzymes** to **learn how to develop better catalysts** for biotechnological applications
- ➢ **Semi-synthetic and artificial enzymes** have important **applications** in **energy sustainability**

References:
(1) Yue et al., *Renewable Sustainable Energy Rev.* **2021**, *146*, 111180
(2) Lubitz *et al.*, *Chem rev.* **2014**, *114*, 4081–4148
(3) Birrell *et al.*, *Coord. Chem. Rev.* **2021**, *449*, 214191
(4) Berggren *et al.*, *Nature* **2013**, *499*, 66–69

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