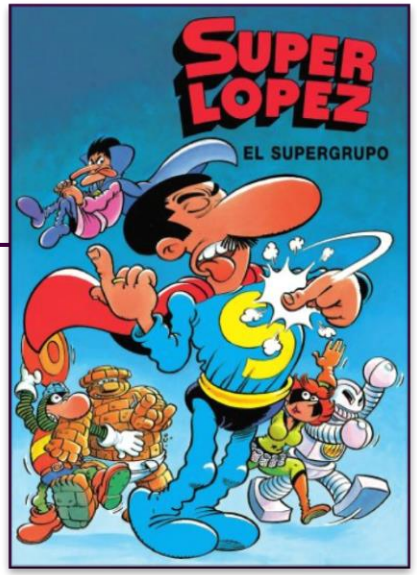
THEORETICAL HETEROGENEOUS CATALYSIS

PROF. NÚRIA LÓPEZ GROUP



@TheoHetCatICIQ

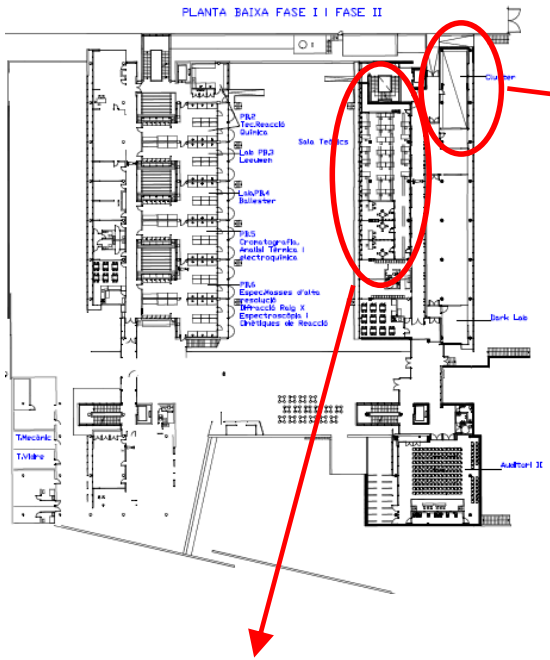
Franziska Simone Hegner

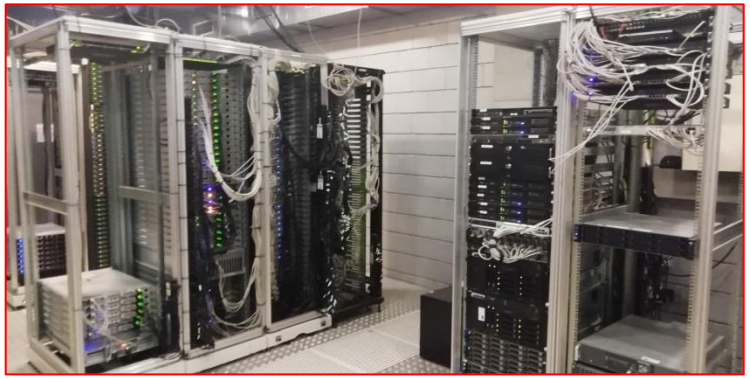
Summer Fellows Meeting Day

2nd of August 2019

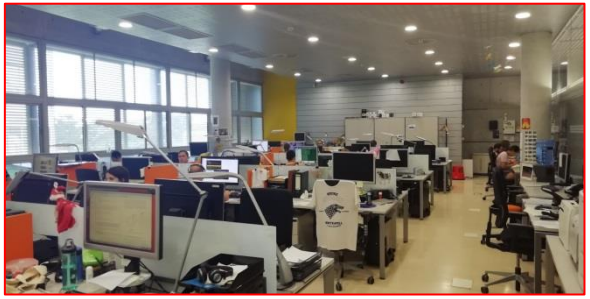
ICIQ

Where are we?



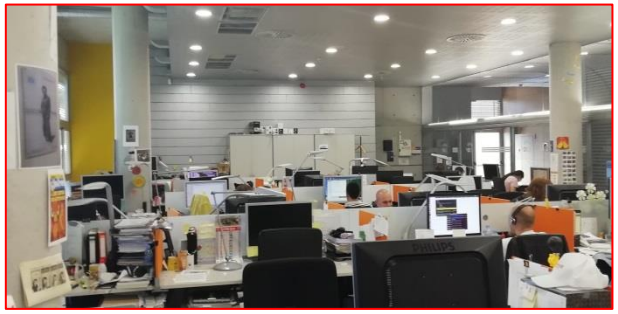


Be careful!



Front

Back



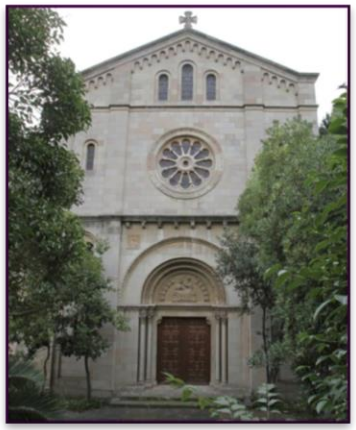
Data centres

MareNostrum @ Barcelona Supercomputing Center
Centro Nacional de Supercomputación

- Most beautiful data centres in the world

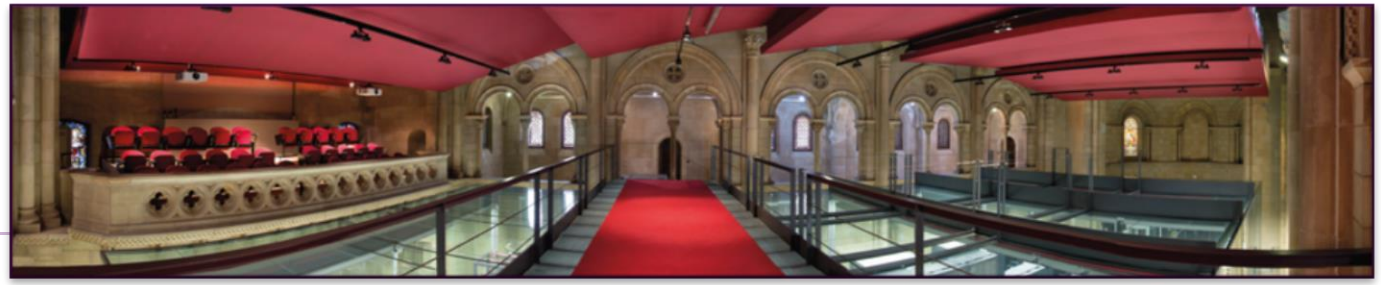
Beauty competition for computers ???

YES !!!

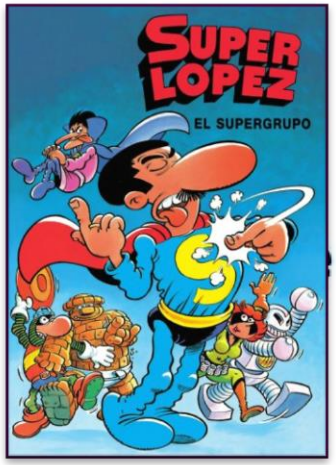


DCD Awards
'Oscars' of the data centre industry.

https://www.dcdawards.global/beautifuldatacenter



Who are we?



$$\Big| \quad \Big\rangle = c_1 \Big| \quad \Big\rangle + c_2 \Big| \quad \Big\rangle + c_3 \Big| \quad \Big\rangle + c_4 \Big| \quad \Big\rangle$$

Materials Researchers & Chemists

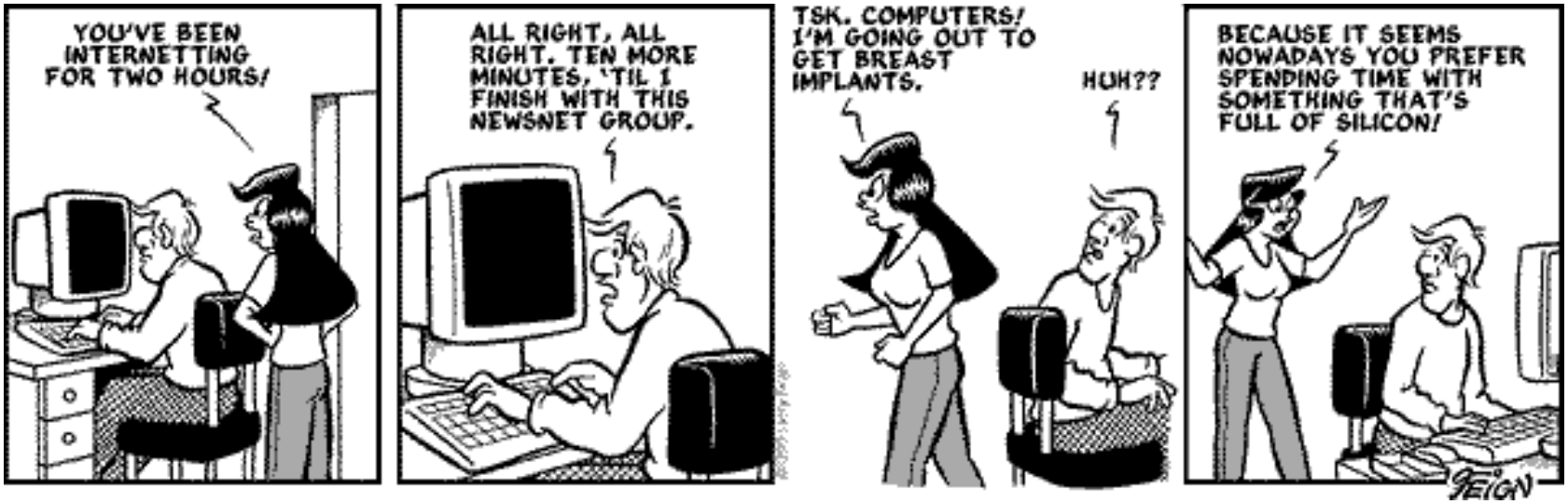
Engineers

Physicists

Mathematicians & Computer Scientists

What are we doing?

- Sitting in front of computers !!!

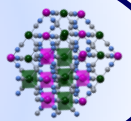


Why ?

characterize
materials structure

elucidate
reaction mechanisms

predict
new catalysts



Theroy vs. Experiment

- **Easy** for theory: provide atomistic information that experimental techniques cannot access
- **Cheap** for theory: test materials for any particular application prior to synthesis or purchase
- **Fast** for theory: screen large libraries of materials *in-silico*

However:

Computers do not know about chemistry

You feed any *input, they provide output*

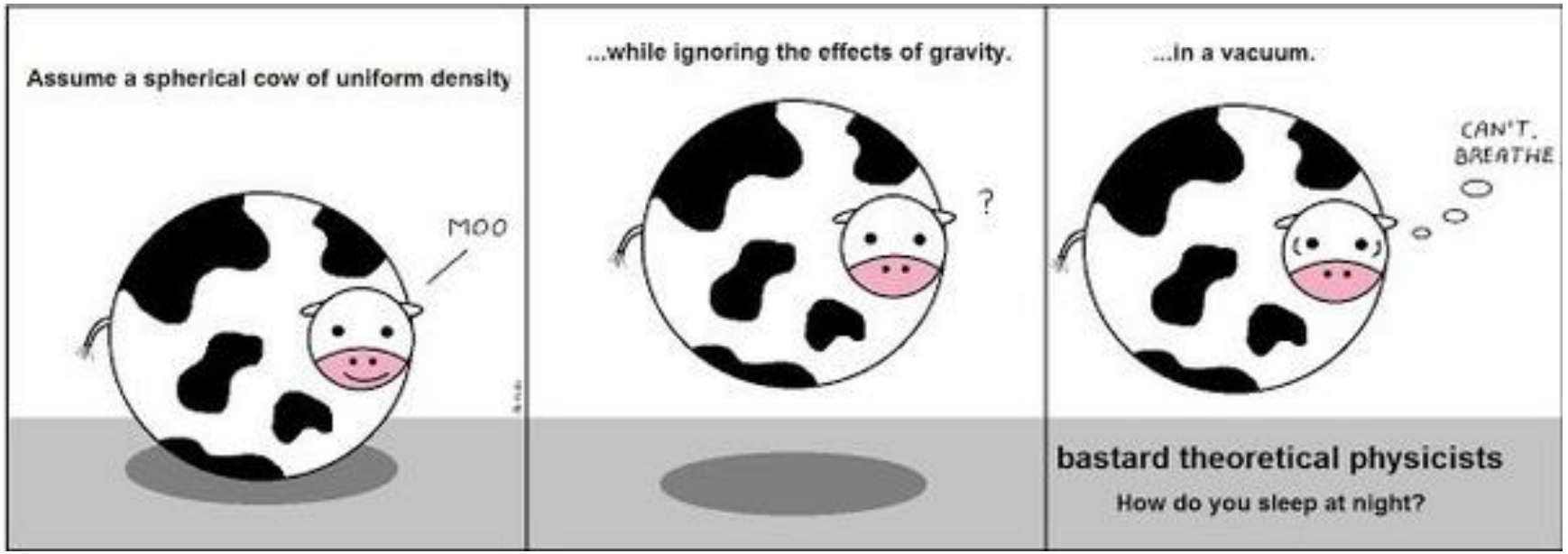
Human factor. Make sure that:

1-We ask the correct question

2-We interpret the result accordingly

3-Use adequate models

Models



We rarely simulate **“reality”**; instead, we use models (as accurate as we can afford) to prove/disprove hypothesis

OVERVIEW

"Classical" Heterogeneous Catalysis (Gas Phase)

(Photo-) Electrocatalysis

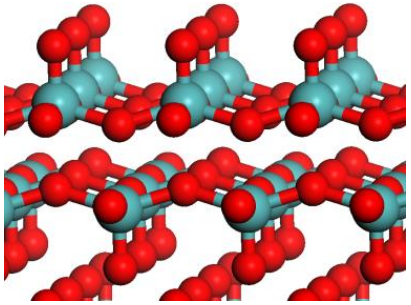
Code Development & Implementation

Catalytic Materials Discovery

New Functionalised Catalysts

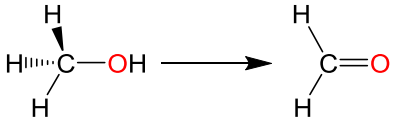
Reactivity on MoO_3

Transformation of hydrocarbons:

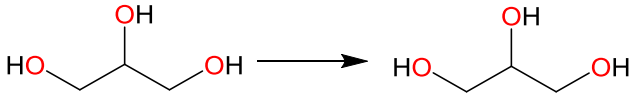


MoO_3

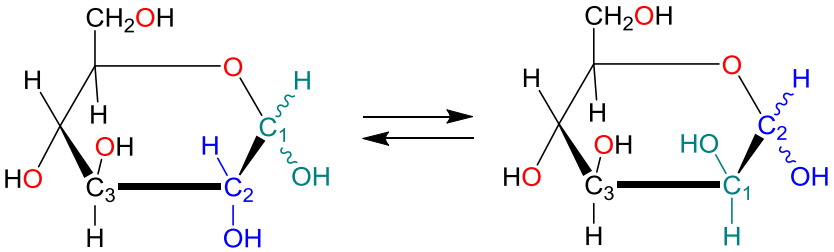
Oxidations



Reductions



Isomerizations



- Detailed mechanism
- Electronic behavior

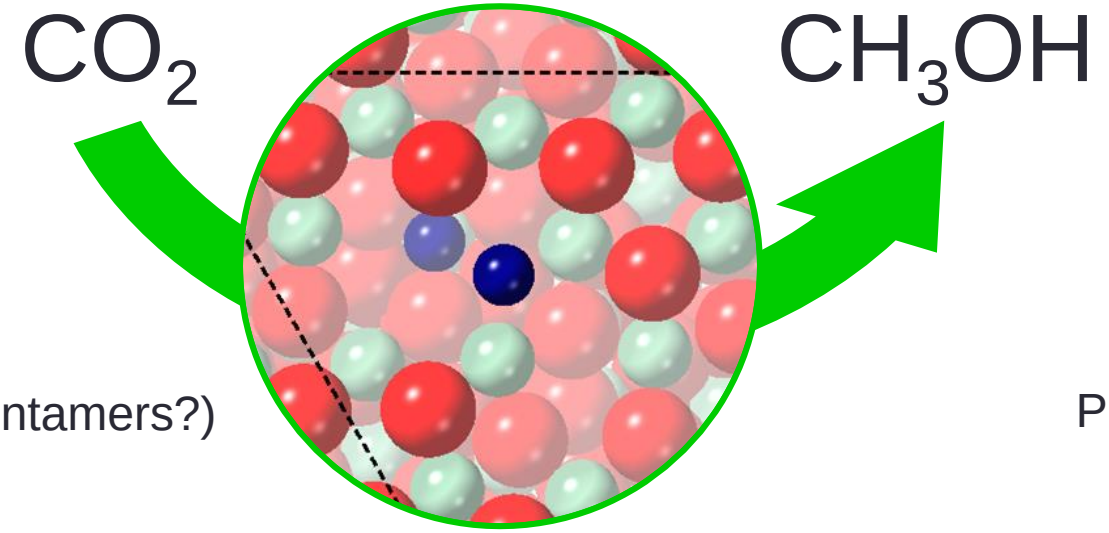
→ To improve catalytic activity and selectivity

Dr. Marcos Rellán

VALIDATION OF EXPERIMENTS

Pd-promoted In_2O_3

CO_2 hydrogenation to methanol



Observations

Pd/In tetramers (pentamers?)

1 or 2 Pd atoms ?

→ What is the active site?

Dr. Rodrigo García-Muelas

Techniques

XPS

EXAFS

Kinetics

TOF-SIMS

HAADF-STEM

Theory

Pd_2In_2 clusters

2 Pd atoms

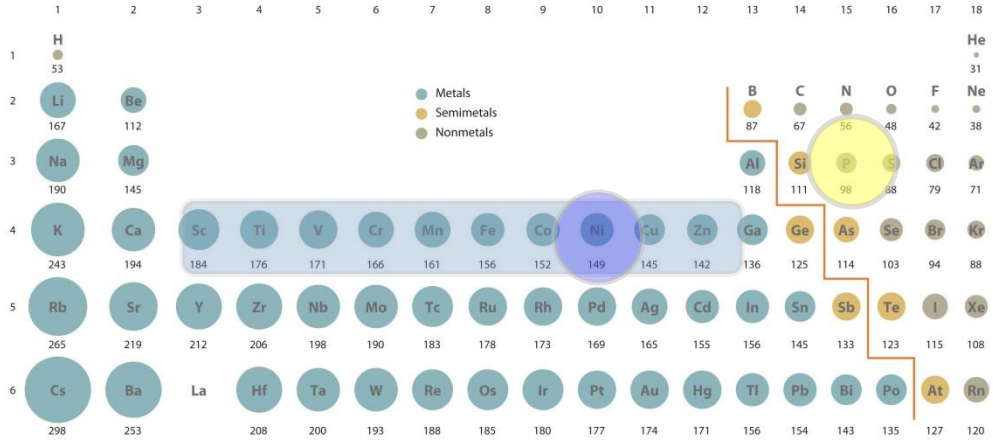
Pd is reduced

Selectivity: 99%

$E_{a,MeOH}$ = 81 kJ mol^{-1}

Nat. Commun. **2019**, *10*, 3377.

Transition Metal Phosphides (TMP)

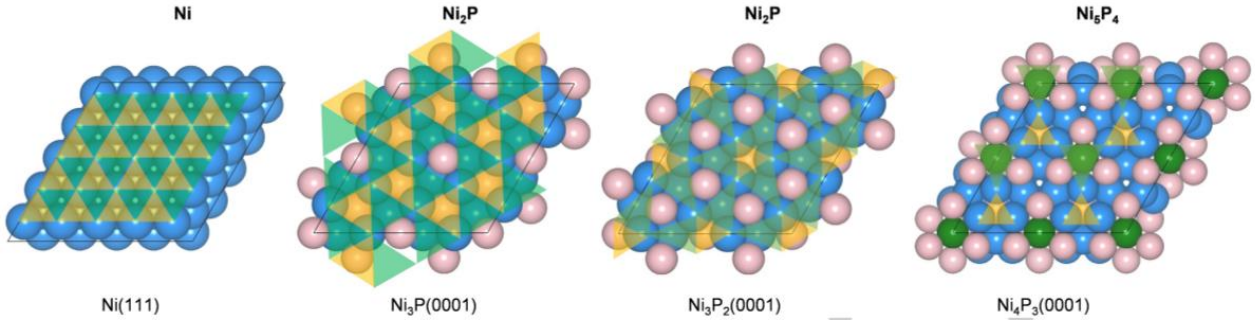


Konstantin Karajovic

- Intrinsic properties
- Semi-hydrogenation
- Hydrogen evolution reaction

Ensembles of Ni_xP_y

- Known TMP
- Controlled surface sites

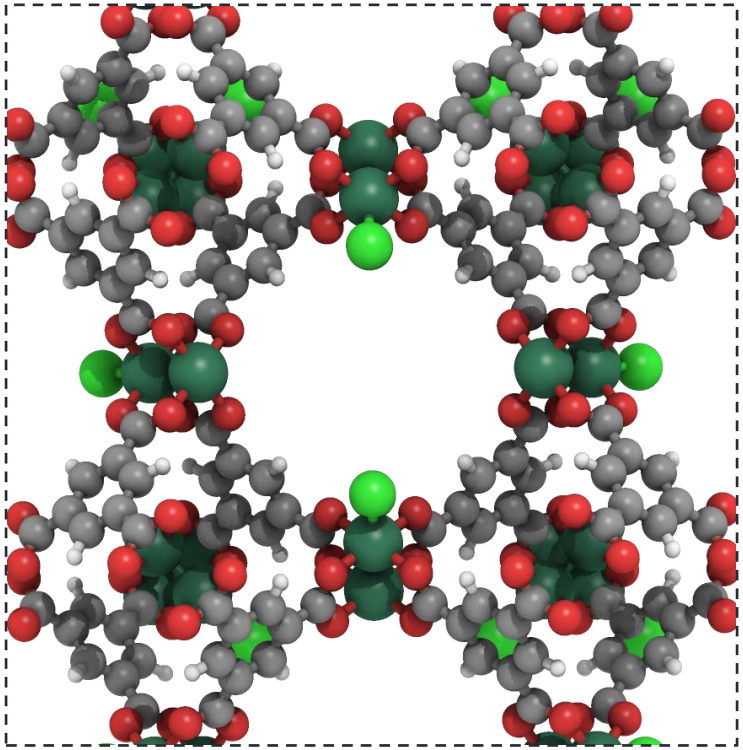


The best of *homogeneous* and *heterogeneous* catalysis

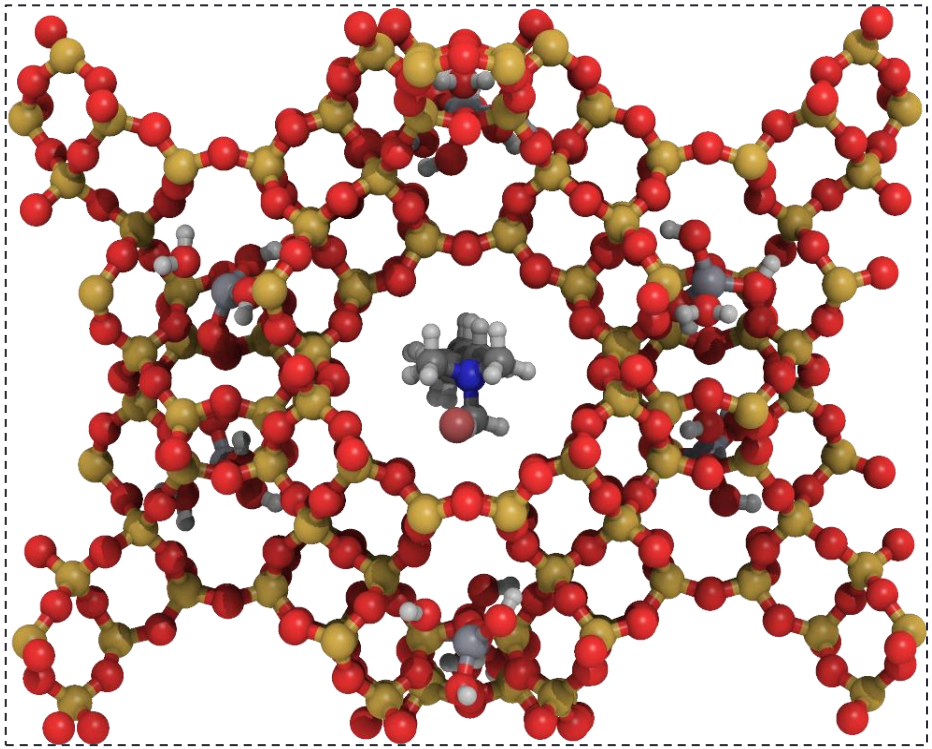
> high selectivity
> easy to tune

> robustness
> easy to recycle

Metal–organic frameworks



Zeolites



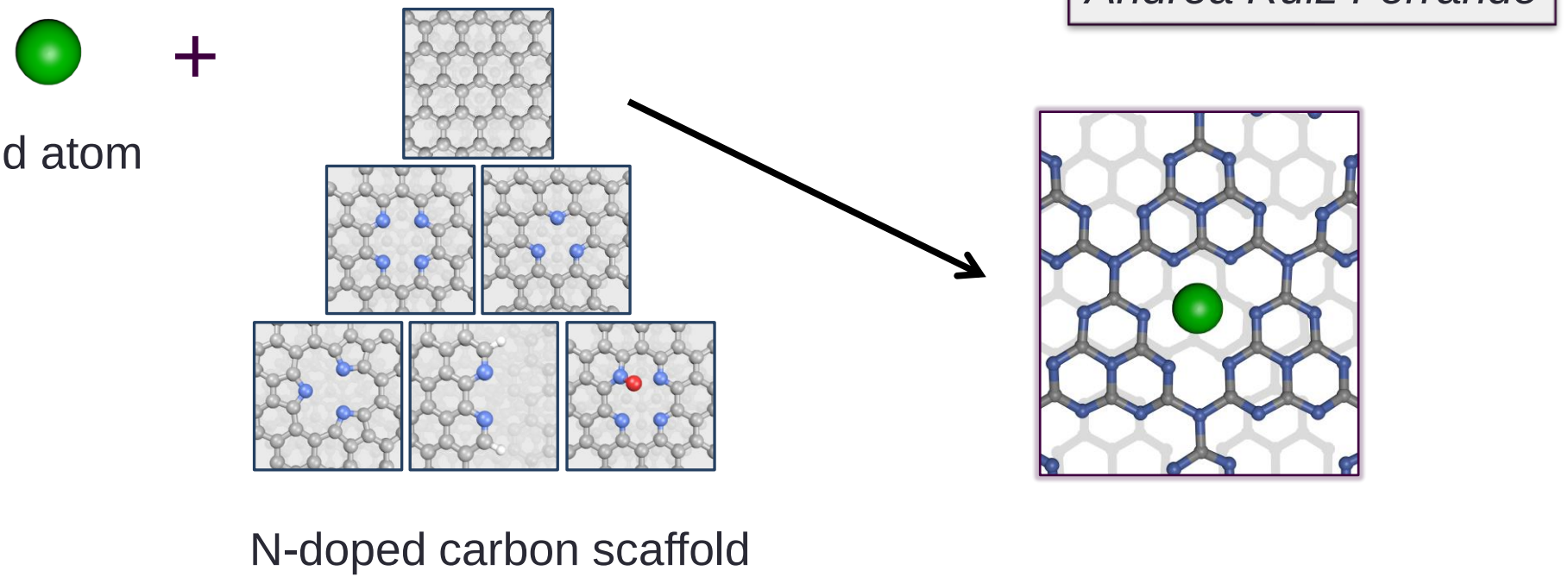
Dr. Manuel Ortuño
Jordi Morales Vidal

Single Atom Catalysis

- Need only a very small amount
- Ultimate surface-to-volume ratio
- Large potential: Application to many different reactions

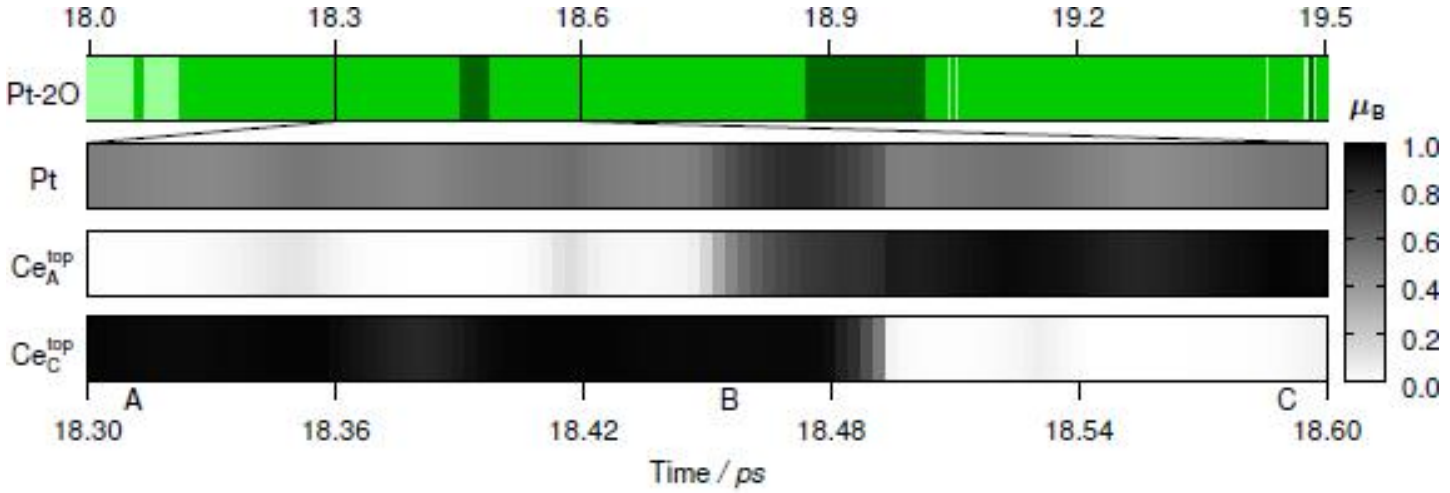
Pt or other single atoms on N-doped carbon host

Edvin Fako
Andrea Ruiz Ferrando

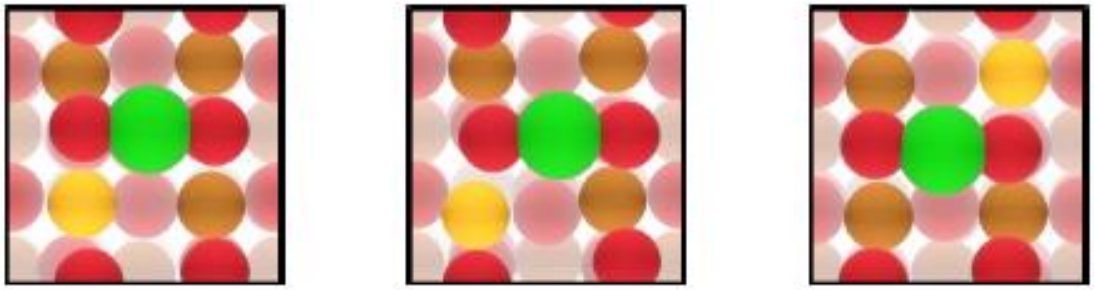


Pt on CeO_2

Monitoring time-dependent charge transfer events



Nathan Daelman



A: 18.306 ps B: 18.450 ps C: 18.585 ps

Green: **Pt**; red: **O**; brown: **Ce^{4+}**; yellow: **Ce^{3+}**.

Random-Walk Kinetic Monte Carlo

Diffusion modeling

- Diffusion on materials can be modeled as a non-reactive process in which a given atom or molecule moves to a neighboring vacancy.

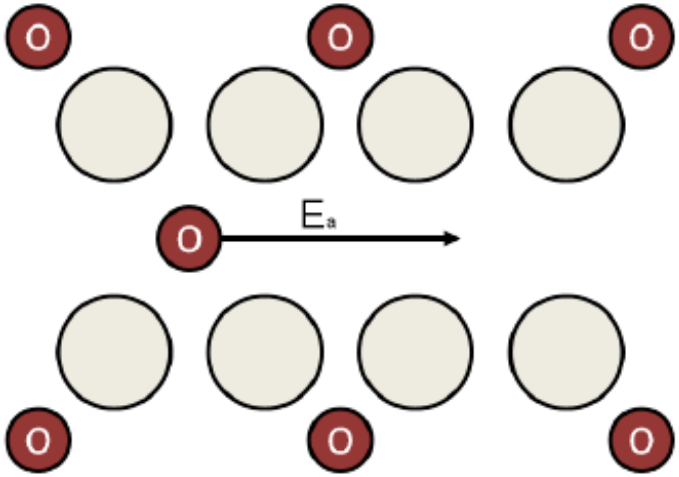


Figure 2: Schematic representation of a (110) zirconia surface. White balls are zirconium atoms and red balls oxygen atoms.

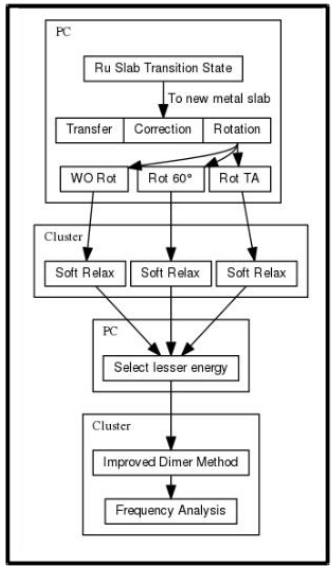
- Stochastic model

Albert Sabadell

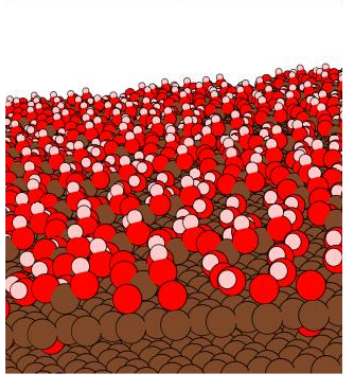
Sergio Pablo García

Robot for automated processes

Avoid humans!

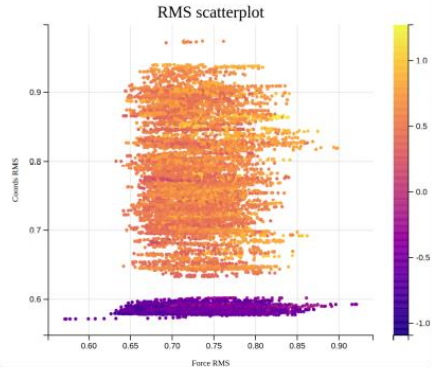


Analyze files!



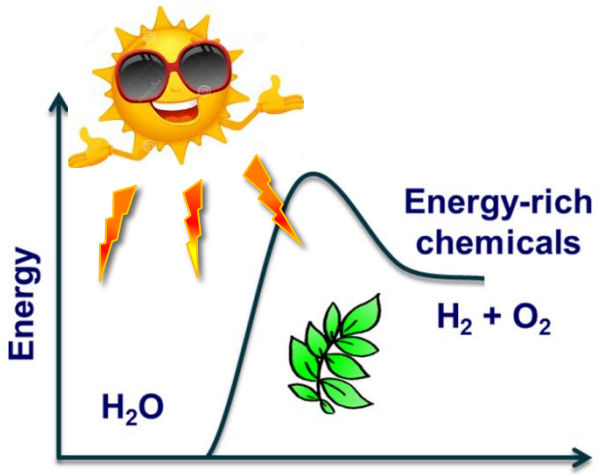
Build models!

Run calculations!



Check results!

ARTIFICIAL PHOTOSYNTHESIS

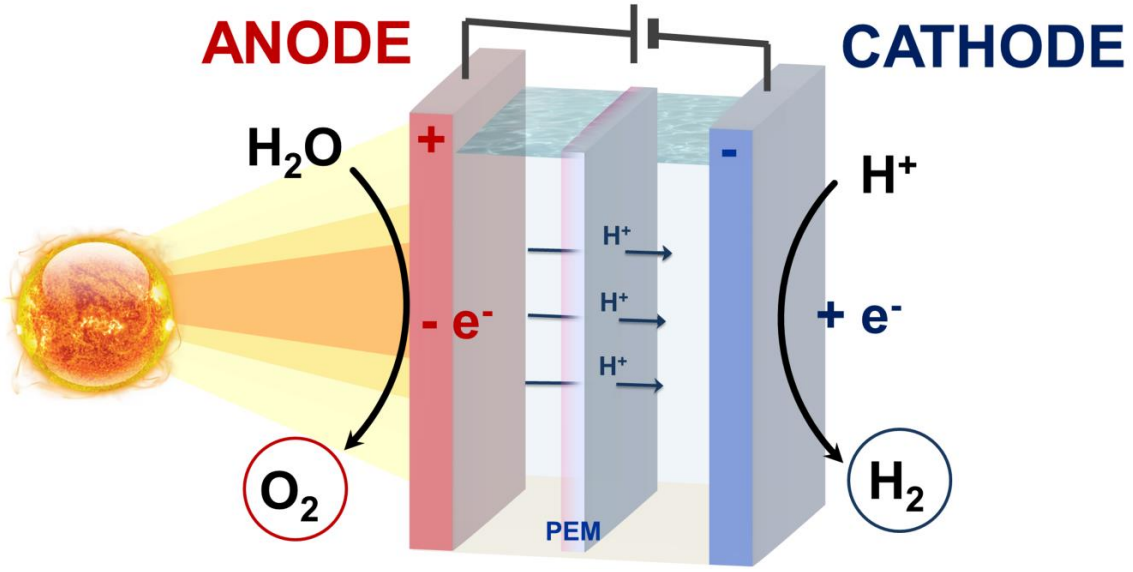


Dr. Nguyen Chuong
Dr. Yecheng Zhou
Dr. Franziska Hegner
Paulina Pršlja
Federico Dattila

- **Water Splitting**
- **CO_2 Conversion**

a-leaf

ELCOREL
Electrochemical Conversion of Renewable Electricity into Fuels and Chemicals



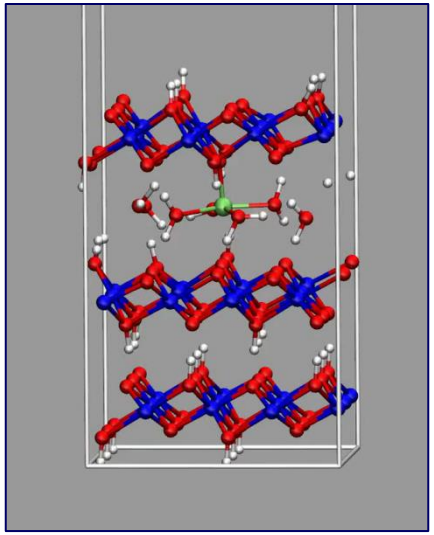
Water Oxidation Catalysts

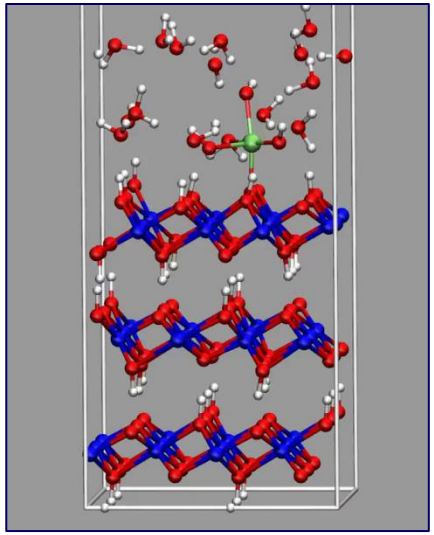
Oxygen Evolution Reaction (OER):

$$2\ H_2O \xrightarrow[(vs\ RHE)]{E^\circ = 1.23\ V} 4\ H^+ + 4\ e^- + O_2$$

Dr. Nguyen Chuong
Dr. Yecheng Zhou
Dr. Franziska Hegner

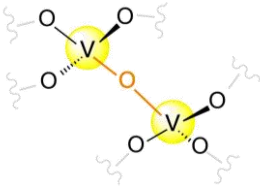
Fe-doped NiOOH

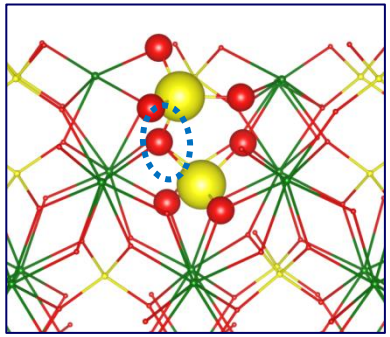




- Intercalation of Fe

$BiVO_4$

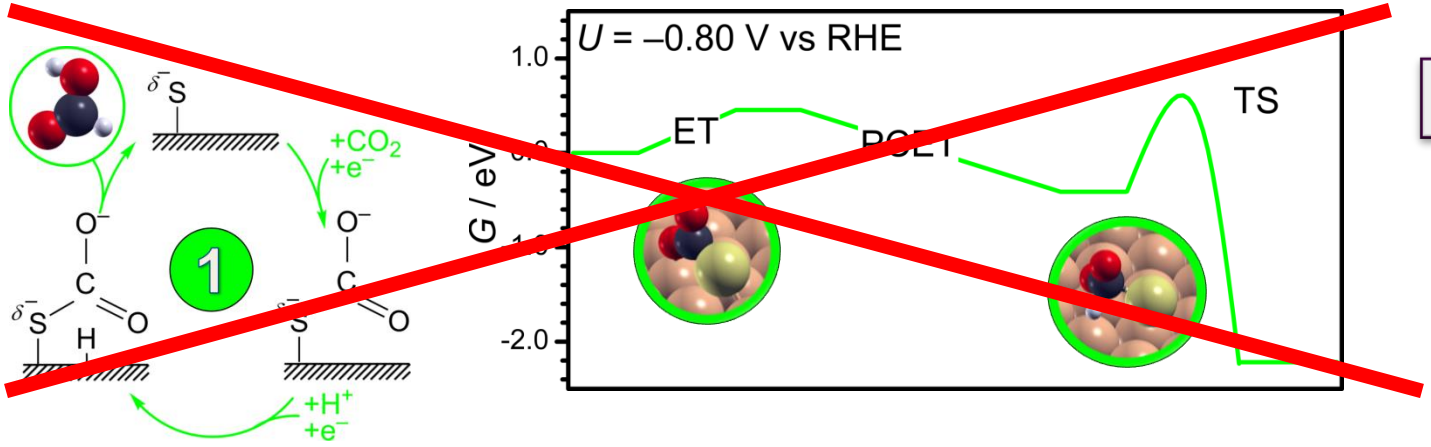




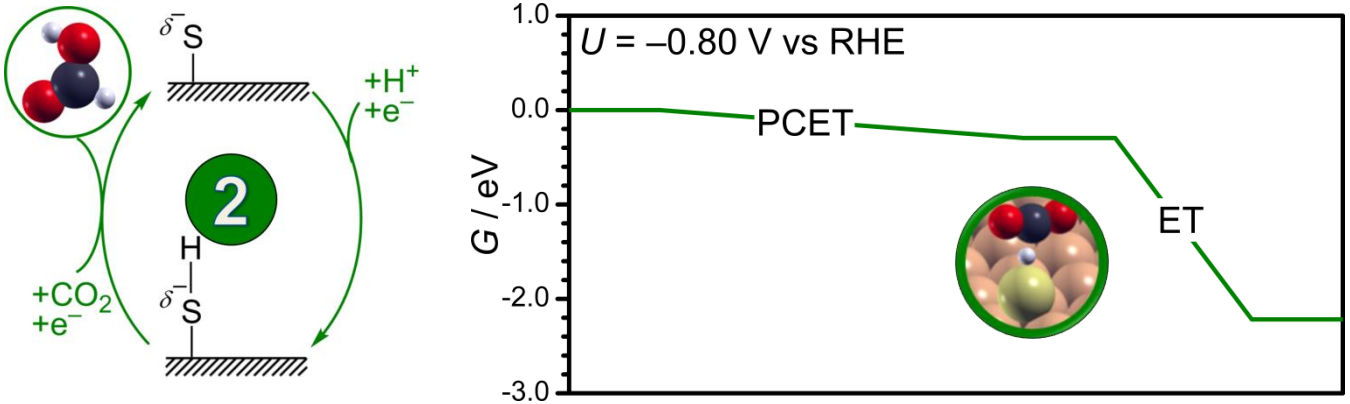
- Oxygen vacancy formation

S-modified Cu

CO_2 reduction to $HCOO^-$

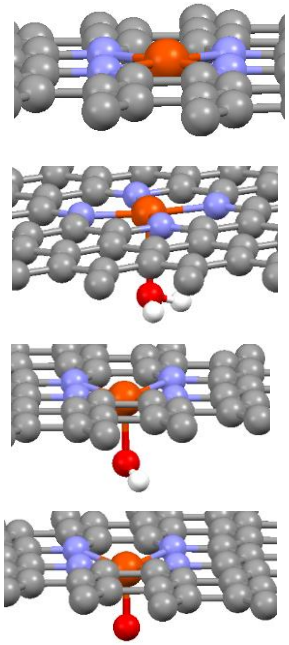


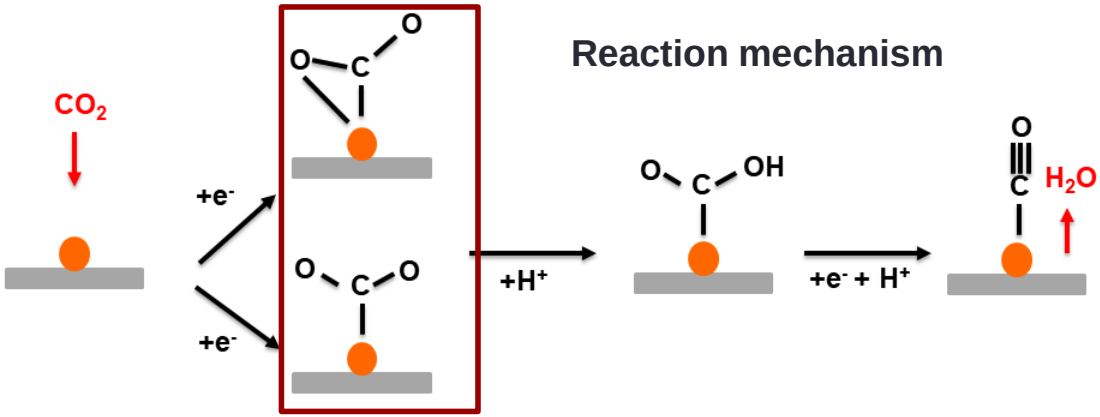
Federico Dattila

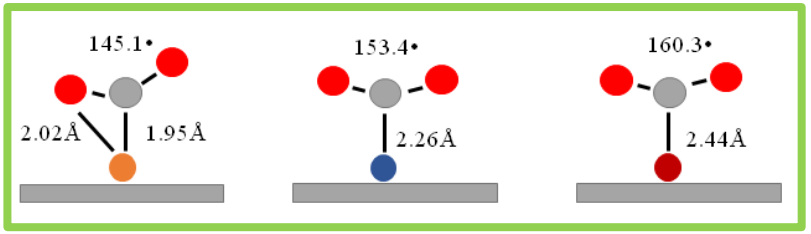


Metal-doped CN nanosheets

CO_2 reduction to CO







Paulina Pršlja

THANK YOU for listening!!!